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EVALUATION OF DIFFERENT ABATEMENT SYSTEMS FOR EMISSIONS OF AMMONIA, PM AND ODOUR IN INTENSIVE LIVESTOCK FARMS

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Abbreviations and acronyms

CC	Climate Change
CH_4	Methane
Cl_2	Chlorine gas
CO_2	Carbon dioxide
Cod	Odor concentration
Covid-19	Coronavirus SARS-CoV-2
EEA	European Environmental Agency
EU	European Union
FAO	Food and Agriculture Organization
FE	Freshwater eutrophication
FEx	Freshwater ecotoxicity
FVW	Fruit and vegetable waste
GC-MS	Gas chromatography coupled with mass spectrometry
GHG	Greenhouse gases
H_2S	Hydrogen sulfide
HTc	Human toxicity, cancer effects
HTnoc	Human toxicity, non-cancer effects
LCA	Life Cycle Assessment
ME	Marine eutrophication
MFRD	Mineral, fossil & renewable resource depletion
N	Nitrogen
N_2O	Nitrous oxide
NO_2	Nitrogen dioxide
NOx	Nitrogen oxides
NECD	National Emission Ceilings Directive
NH_3	Ammonia
OD	Ozone Depletion
PM	Particulate matter
$PM_{2.5}$	Particulate Matter with a diameter ≤ 2.5 µm
PM_{10}	Particulate Matter with a diameter ≤ 10 µm
POF	Photochemical ozone formation
TA	Terrestrial acidification
TE	Terrestrial eutrophication
VOCs	Volatile Organic Compounds

Abstract

Agriculture in the 21st century faces multiple challenges: it has to produce more food to feed a growing population with a smaller rural labour force, adopt and learn new technologies, satisfy consumers' changing tastes and expectations, and at the same time adopt more efficient and sustainable production methods to cope with climate change, soil erosion and biodiversity loss. Livestock contribution to greenhouse gases (GHG), ammonia (NH_3), and odor emissions is relevant. Thus, the identification and adoption of mitigation strategies for intensive livestock farming is necessary, not only to reduce the environmental impact related to livestock production, but also to help solving the issue related to air pollution. Indeed, GHG, NH_3 and odor emission other having a negative effect on the environment, represent a risk for human and animals' health. Air treatment technologies have the double benefit of abating pollutants emissions and improving air quality inside the barns. Alternatively, considering several aspects i) that animal proteins place a heavy burden on our natural resources, ii) the world population and demanding consumers are rapidly increasing, iii) the amount of agricultural land is limited, there is an urgent need to find alternatives to conventional meat products. Terrestrial invertebrates farming represents an environmentally sustainable solution. Earthworms require less land and water, present lower GHG emissions and higher feed conversion efficiency, moreover they are rich in proteins and essential amino acids. The aim of this thesis was to evaluate different mitigation strategies to reduce GHG, NH_3 and odor emissions deriving from intensive livestock farming, also taking in consideration their environmental impact. The Life Cycle Assessment (LCA) methodology was applied to quantify the environmental impact of the strategies proposed. In the first step, livestock contribution to air pollutants emissions was quantified. Agricultural NH_3 emissions did not even reduce during lockdown periods imposed by Covid-19 pandemic. Thereafter, odor annoyance issue was deeply analysed. Finally, environmental benefits achievable by applying different solutions (i.e. air treatment technologies, manure additive, earthworms rearing) were assessed. The results in this thesis outlined that the adoption of mitigation strategies is fundamental and promising, but each one presents different abatement efficiency on air emissions. Thus, a combination of measures is often the best solution to control emissions, at the same time also an integrated approach combining different methods and techniques could represent the best solution to depict exhaustively the problem and define a good strategy for its management. In this regard, LCA helps to identify the most important contributors to the environmental impact.

Riassunto

Nel XXI secolo l'agricoltura dovrà affrontare molteplici sfide: produrre più cibo per sfamare una popolazione in continua crescita avvalendosi di una forza lavoro più ridotta, adottare e apprendere nuove tecnologie, soddisfare i gusti e le aspettative dei consumatori che sono in continuo mutamento e, al tempo stesso, diventare più efficiente e sostenibile per contribuire a ridurre i problemi associati al cambiamento climatico, all'erosione del suolo e alla perdita di biodiversità. Il contributo degli allevamenti alle emissioni di gas serra (GHG), ammoniaca (NH_3) e odori è rilevante. Pertanto, si rende necessaria l'identificazione e l'adozione di possibili strategie di mitigazione da applicare agli allevamenti intensivi così da ridurne l'impatto ambientale e le emissioni ad essi associate. Infatti, GHG, NH_3 e odori non solo hanno un effetto negativo sull'ambiente, ma rappresentano anche un rischio per la salute umana e animale. In questo contesto, le tecnologie per il trattamento dell'aria hanno un duplice vantaggio, da un lato, infatti, sono in grado di abbattere le emissioni di inquinanti che verrebbero rilasciate in atmosfera e dall'altro migliorano la qualità dell'aria all'interno dei ricoveri zootecnici. Oltre alle strategie "tradizionali", il consumo alimentare di insetti svolgerà un ruolo di primo piano nell'affrontare le numerose sfide del presente e del futuro. Gli invertebrati terrestri, infatti, ricchi in proteine e amminoacidi essenziali, pullulano in abbondanza sul nostro pianeta, e presentano molteplici vantaggi: i) richiedono poca terra e acqua, ii) presentano minori emissioni di gas serra e iii) una maggiore efficienza di conversione dei mangimi. Ciò li rende una soluzione ecosostenibile. Lo scopo di questa tesi è stato quello di valutare diverse strategie di mitigazione per ridurre le emissioni di GHG, NH_3 e odori derivanti dall'allevamento intensivo, tenendo anche in considerazione il loro impatto ambientale. La metodologia del Life Cycle Assessment (LCA) è stata applicata per quantificare l'impatto ambientale delle strategie proposte. Nella prima fase è stato quantificato il contributo dell'agricoltura e della zootecnia alle emissioni di inquinanti atmosferici. Le emissioni di NH_3 derivanti dal settore agricolo sono rimaste pressoché invariate anche durante i periodi di *lockdown* imposti dalla diffusione del Covid-19. Successivamente, è stato considerato il problema legato alle molestie olfattive. Infine, sono stati valutati i benefici ambientali ottenibili adottando diverse strategie di mitigazione quali ad esempio tecnologie per il trattamento dell'aria, utilizzo di additivi durante lo stoccaggio delle deiezioni, e l'allevamento di lombrichi su scarti vegetali. In conclusione, i risultati di questa tesi hanno evidenziato che l'adozione di strategie di mitigazione è fondamentale e fornisce dei risultati promettenti. Ciascuna di esse presenta un'efficienza diversa nell'abbattimento delle emissioni in atmosfera. Quindi, utilizzarne più di una è spesso la soluzione migliore per ottenere prestazioni più elevate. Al tempo stesso un approccio integrato può contribuire a definire in modo esaustivo il problema e anche a identificare la corretta strategia per la sua gestione. A tal proposito, l'LCA, attraverso la misurazione dell'impatto ambientale, permette di individuare i processi a maggior impatto e quindi di poter agire su di essi.

CHAPTER 1

1. Introduction

1.1 Population growth and agriculture sustainability

Global agriculture will face many challenges over the coming decades and climate change will complicate this. In 2050, the world's population is expected to grow to almost 10 billion people, boosting agricultural demand by some 50 percent compared to 2013. This implies that agriculture has to produce more, in particular, cereal production is expected to increase by almost one billion tons yearly and meat production would go from 200 million tons to 470 million tons in 2050 (FAO, 2017; Tripathi et al., 2019). Globally, the growth in crop production is expected to come both from higher yields coupled with increased cropping intensity, and from land expansion. In particular, arable land would expand by some 70 million ha, with the expansion taking place mainly in developing countries (i.e., sub-Saharan Africa and Latin America). Irrigated land would expand by 11% (around 32 million ha), while harvested irrigated land would expand by 17% (FAO, 2017). Satisfying these increased demands on agriculture, with existing farming practices, is a pressing challenge. Indeed, agriculture has to produce more, thus implying a more intense competition for natural resources, increased greenhouse gas emissions, erosion of biodiversity, and further deforestation, but at the same time it is requested that agri-food production should be more efficient and sustainable to pose attention to climate change (Tripathi et al., 2019). Also, consumers are posing more attention to sustainability, trying to make food choices that minimize food waste and can be considered "environmental friendly", such as substituting meat with vegetable products (Westhoek et al., 2014). If this is true for developed countries, in other parts of the world hunger and malnutrition remain a huge challenge, that, according to the Food and Agriculture Organization (FAO), will not be eradicated even by 2050 (FAO, 2017). The international community is aware of these challenges. For this reason, in 2015, all United Nations Member States have adopted the 2030 Agenda for Sustainable Development. In particular, the second sustainable development goal aims at ending hunger, achieving food security and improved nutrition, and promoting sustainable agriculture, simultaneously (United Nations, 2015). Optimize, improve and make more efficient use of resources through technological progress, social innovation and new business models are necessary processes to move towards sustainable agriculture. This is a hard work as producing more means a higher environmental impact. At the same time, even if food production is increasing a significant share of the world's population is suffering from under-nutrition or malnutrition (FAO, 2017). Regarding these last aspects, meat plays a crucial role. It represents an important source of nutrition but meat products are among the most energy-intensive and ecologically burdensome foods. They imply high greenhouse gas emissions, agricultural land and freshwater use, especially diets rich in ruminants' products (i.e. cattle) (Opio et al., 2013). According to Gerber et al. (2013), livestock sector contributes 7.1 gigatonnes CO_2 eq (around 14%) to global anthropogenic greenhouse gas (GHG) emissions. In particular, summarized by sector, beef and cattle milk productions are responsible for most emissions (41 and 20%, respectively), followed by pig meat

(9%), and poultry meat and eggs (8%). Thus, the global livestock sector has to face a three-fold challenge: (i) increase the production to meet the global food demand, (ii) adapt to a changing and increasingly variable economic and natural environment, and (iii) improve its environmental performance (Opio et al., 2013).

1.2 Livestock contribution to environmental impact

Carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O) are greenhouse gases (GHG) responsible for the global warming effect and livestock activities emit considerable amounts of these three gases. In 2018, world total agriculture and related land use emissions reached 9.3 billion tonnes of carbon dioxide equivalent (Gt CO_2 eq), of which more than half is generated by crop and livestock activities (FAO, 2020). Direct emissions from livestock come from the respiratory process in the form of CO_2. Among livestock, ruminants are the primary contributors to CH_4 emissions (25-40%), with cattle representing 65% of the livestock sector emissions (Gerber et al., 2013). CH_4 emissions derive both from enteric fermentation and from livestock manure management systems. CH_4 emissions from enteric fermentation are a physiological by-product of herbivores' digestive process, in which carbohydrates are broken down by micro-organisms into simple molecules for absorption into the bloodstream. The amount of CH_4 released depends on various factors, such as the age, weight, and health state of the animal, other than the quality and quantity of the feed consumed (IPCC, 2019). CH_4 emissions from manure management are produced by the decomposition of manure under anaerobic conditions, during its storage and treatment. About 10% of GHG produced by livestock derive from CH_4 emitted during manure storage and processing. Manure management, application and deposition on pastures are also the primary source of N_2O emissions (around 16%) (Gerber et al., 2013). In particular, manure contributes from more than 20% (for pigs) to less than 1% (for poultry) to GHG emissions attributable to livestock sector (Lesschen et al., 2011). N_2O emissions are produced during the nitrification-denitrification of nitrogen contained in livestock waste and the amount released depends on the system and duration of waste management (IPCC, 2019).

Although livestock generates a significant proportion of anthropogenic GHG emissions (Gerber et al., 2013), agriculture is most demonized as the primary source of ammonia (NH_3) emissions (Oenema et al., 2012). In the European Union (EU), agriculture is responsible for about 92% of the NH_3 emissions. The residual part comes from industry, households and traffic (EEA, 2018). Manure management is the main source of NH_3 emissions, followed by the application of manure mineral nitrogen fertilizers (Oenema et al., 2012). According to EEA Report No 6/2018, animal manure applied to soils, inorganic N-fertilizers, and non-dairy manure management jointly make up 52 % of total NH_3 emissions. NH_3 emissions are mostly produced by bacterial and enzymatic decomposition of protein components in the animal waste (EMEP/EEA, 2019). Being NH_3 the most abundant alkaline compound in the atmosphere, it plays an important role in determining the overall acidity of precipitation, cloud water and airborne particulate matter (PM) (Behera et al., 2013). It is a critical N compound that negatively affects both the natural environment and human health. Indeed, i) released into environment it causes soil acidification and water and terrestrial eutrophication, with an adverse effect on biodiversity; ii) in gaseous form it reacts with sulfuric acid and nitric acid to form ammonium sulfate and ammonium nitrate aerosols, leading to the formation of secondary inorganic particulate matter (PM_{10} and $PM_{2.5}$), a threat to human and animal health (Behera et al., 2013; Wagner et al., 2015). Such small PM particles can get deep into the lungs and even pass into the blood circulation, affecting both respiratory and cardiovascular systems (Li et al., 2018). Moreover, prolonged exposure or exposure to high levels of NH_3 are associated with a higher proportion of ill-health in workers, and with disease, or lower production parameters in animals (Maesano et al., 2019; Michiels et al., 2015). According to de Bruyn et al. (2018) the damage to public health and ecosystems, due to NH_3 emissions, can be valued at 17.50€ (±7.50)/kg NH_3. This includes the contribution of NH_3 to acidification, eutrophication and formation of PM and related loss of years of life (de Bruyn et al., 2018). To tackle the problem, a mix of regulation, incentives and education are likely to be necessary to support the implementation of interventions. Some international and national Authorities have already imposed stricter environmental regulations concerning GHG and NH_3 emissions into the atmosphere (Zilio et al., 2020). As an example, at the European level, the Ambient Air Quality Directive (EC, 2008) and the National Emission Ceilings Directive (NECD) (EU, 2016) are the EU’s main legal instruments to reduce overall emissions of air pollution. Moreover, wider education and awareness-raising should be promoted to both help build understanding and stress the importance and benefits of reducing GHG and NH_3 emissions.

1.3 Covid-19 and livestock farming

The outbreak of Covid-19 (coronavirus SARS-CoV-2) has emphasized the attention to air quality and air pollution. Indeed, according to the literature, air pollution is a factor which may facilitate the spread, severity and prognosis of the disease (Manigrasso et al., 2020). The explosion of Covid-19 cases around the world has made it a global pandemic and had devastating consequences on people's livelihoods, behavior, health and the global economy (El Keshky et al., 2020). The first case of Covid-19 was detected in the city of Wuhan in November 2019, after that Italy was the second country affected by coronavirus disease, with the first confirmed case of infection detected on 20th February 2020. To contain the virus, many countries, including Italy, have adopted dramatic measures, more specifically lockdown periods, to reduce human interaction and Covid-19 spread. During the strict lockdown period, people were forbidden to go out except to buy essential goods (i.e., medicines and food). Transport and industrial activities mostly stopped, schools and universities were closed, large-scale gatherings were prohibited, social distancing was encouraged, curfews were imposed, and personal protective equipment should be worn. Just hospitals, food and pharmaceutical supply chains, and the agrifood sector were allowed to remain operational (Venter et al., 2020). These, just mentioned, were just some government actions imposed to contain the spread of the infection. The diffuse periods of lockdown, held in several countries, deeply reduced anthropogenic emissions caused by industries, energy-related industries, and traffic. Many authors observed an appreciable drop in NO_2, NOx, PM_{10} and $PM_{2.5}$ (Collivignarelli et al., 2020; Venter et al., 2020; Zambrano-Monserrate et al., 2020). Conversely, other pollutants emissions were not affected, in particular those associated with agricultural activities as the agricultural sector was not interested in any restriction measures. Focusing on livestock and agricultural activities, no reduction effects on atmospheric NH_3 have been observed, as a consequence of Covid-19 (Deserti et al., 2020; Gualtieri et al., 2020). This implies that also its effects on the environment and on human health were not affected. Europe, and in particular the Po Valley area, is recognized as a zone with high emission rates. This reflects the patterns of animal densities (species and intensive production), and the type and intensity of synthetic fertilizer use. This situation makes it necessary to develop and apply, at a national and regional level, NH_3 emission abatement strategies.

1.4 Livestock odor emissions

Besides being the major causes of NH_3 emissions, agriculture and livestock farming are important sources of odorant compounds that cause conflicts with the neighboring population. Odorous components not only affect the life quality of people living near the production facilities, but are also a concern for the health of animals and agricultural workers (Blanes-Vidal et al., 2012). Indeed, unpleasant odors may cause a variety of emotional and undesirable reactions, ranging from annoyance to documented health effects, such as sensory irritations in eyes, throat, and nose and neurochemical changes (Blanes-Vidal, 2015; Nimmermark, 2004). Odors perception is a complex phenomenon with a strong subjective component. Odor can be defined as the perception given by the interaction of different volatile chemical species (odorous compounds) to the olfactory organ. Odorous compounds include both organic odorants and inorganic molecules; volatile organic compounds (VOCs) are included in the first group, whereas hydrogen sulfide (H_2S), elemental chlorine gas (Cl_2), and NH_3 are examples of the second group of molecules (Guo et al., 2019).

Manure and urine are the main source of odor. However, odor emissions from animal production facilities are a function of many variables including species, housing types, feeding methods, management factors, manure storage and handling methods (Guo et al., 2004; Van der Heyden et al., 2015).

Odors and their discomfort can be measured by applying different approaches and techniques. In particular, their quantitative and qualitative characterization can be carried out thanks to analytical and sensorial methodologies. Gas chromatography coupled with mass spectrometry (GC-MS) and electronic nose are examples of analytical techniques, whereas sensorial techniques include: dynamic olfactometry, field inspection and recordings from residents (Bax et al., 2020; Capelli et al., 2013). GC-MS allows to analytically identify and quantify odorous compounds, but it does not permit to characterize the source and to establish if the olfactory sensation is due to an individual odor constituent or to the whole mixture. Instead, the electronic nose allows a qualitative classification of the analyzed air, attributing the air sample to a specific olfactory class. The output is a pattern, which is typical of the gas mixture. Among sensorial techniques, olfactometry analysis is a standardized methodology (Standard EN 13725:2003) used for determining the concentration of odorous, combining an olfactometer with human panelists. The olfactometer is an instrument that submits the odor sample, diluted with neutral air at precise ratios, to a panel of human assessors. The samples are presented to the panel at increasing concentrations until the panel members start perceiving an odor different from the neutral reference air. The result is the odor concentration (Cod), expressed in European odor units per cubic meter (ouE/m^3). Cod corresponds to the dilution factor necessary to reach the odor threshold, that is the minimum concentration perceived by 50% of population (Bax et al., 2020). Field inspections involve the use of qualified human panel members in the field to directly assess the presence of recognizable odor in ambient air. In the European Standard EN 16841:2016 are described two approaches to assess field inspections. The first one is the grid method, which uses direct assessment of ambient air by panel members to characterize odor exposure in a defined assessment area (Standard EN 16481-1:2016). The second one is the plume method and it is used for determining the extent of the downwind odor plume of a source (Standard EN 16481-2:2016).

Unfortunately, these techniques alone are not sufficient to assess odor impact within a community. Odor dispersion models, combined with odor measurements, allow a better understanding of odor nuisance. Indeed, dispersion models take into account meteorological, topographical and emission data, that influence odors dispersion into the atmosphere. They offer the possibility of mathematically simulating the spatial and temporal variation of odor concentrations and can provide estimates of odor levels in both current and future emission scenarios, predicting the atmospheric impact of a work before being realized (Capelli et al., 2013). These models are useful to determine appropriate setback distances between livestock production facilities and neighboring areas (Guo et al., 2004). Alternatively, in case of already exiting livestock farming causing odor annoyance to nearby residents, different mitigation strategies can be applied, such as nutritional strategies, manure additives, building design, air cleaning systems, manure covers, manure treatment systems and windbreaks. (Ubeda et al., 2013).

1.5 Mitigation strategies and livestock environmental impact

Reduction of livestock pollutants and odors emissions can be realized through mitigation strategies aimed at reducing emissions before they are emitted, or at containing them once produced. As an example, nutritional strategies and appropriate building design fall into the first category, whereas the application of manure covering systems, manure treatments (e.g. anaerobic digestion, solid-liquid separation, nitro-denitro) and air treatment technologies (e.g. bioscrubber, acid scrubber, biotrickling filter) fall under the second one (Ubeda et al., 2013; Van der Heyden et al., 2015).

Modified animal feeding can decrease odors and NH_3 emissions. As an example, in pigs, modifying the inclusion level of proteins and fermentable carbohydrates can affect NH_3 and odor emissions (Le et al., 2005). In particular, low-protein diets could reduce NH_3 emissions up to 63% (Dourmad and Jondreville, 2007;

Portejoie et al., 2004), and odor emissions by 30% (Hayes et al., 2004). Alternatively, increasing the fermentable carbohydrates affects bacterial proliferation in both the gastrointestinal tract and in the manure, modifying N excretion mechanism and thus reducing NH_3 and odor emissions (Le et al., 2005).

NH_3 and odor emission abatement measures are available for all steps in the sequence of animal production and manure management. During storage, covering manure provides a physical barrier to reduce both NH_3 and odors emissions. Indeed, constructing rooftops or covering the surface with different materials reduce the emitting surface of the slurry, obstructing the free exchange of volatile compounds from the underlying liquid to the atmosphere (Portejoie et al., 2003). According to Guarino et al. (2006), vegetable oil is the best floating cover (79.5% to 100% of NH_3 emission reduction), whereas Portejoie et al. (2003) achieved the best results with zeolites compared to oil (71% Vs. 40%). Other strategies to reduce NH_3 volatilization and odor emissions during storage consist in using additives. Numerous types of additives have been investigated over the last decades including masking agents, surfactants, neutralizers, plant extracts, adsorbents, bacterial-enzymatic preparations, but their abating efficiency is still controversial. Acidifying and adsorbent additives, such as sulphuric acid and zeolites, have proven to be effective in reducing NH_3 volatilization but not odors, whereas masking, disinfecting, and oxidizing agents reduce the release of odorous compounds but require frequent reapplication, thus being not economically sustainable (McCrory and Hobbs, 2001). Other technologies with potential impacts on gaseous N and odor emissions include: anaerobic digestion, separation of solid and liquid fractions, reverse osmosis, or acidification. Despite manure treatments are not all specifically designed to abate odors, they may lead to odor emission reduction by altering the anaerobic conditions or eliminating the precursors of odorous compounds (Ubeda et al., 2013).

Another significant part of air pollutants emission occurs during spreading. According to Oenema et al. (2012), low-emission manure application techniques should involve machinery that (i) decreases the exposed surface area of slurries applied to surface soil, and/or (ii) buries slurry or solid manures through injection or incorporation into the soil. Costs of these techniques are in the range of 0.1 to 5 €/kg NH_3-N saved. Other low-emission application methods include band spreading techniques. Finzi et al. (2019) investigated five mitigation techniques (straw, sawdust, clay, oil and sulphuric acid) during storage, coupled with three different spreading methods (broadcast spreading, band spreading, and closed-slot injection). They concluded that all mitigation techniques considered could significantly reduce NH_3 emissions from slurry storage. However, the type of slurry (cattle, pig, or digestate) and its chemical composition affect the mitigation effect. In particular, pig slurry and digestate tend to have higher NH_3 emission potential. Among the mitigation strategies evaluated, acidification provides the best results with an average NH_3 reduction of 95%, followed by covering slurry with oil (87%), whereas the other floating covers (straw, sawdust and clay) were less effective (from 44% to 53% NH_3 abatement efficiency). Finally, regarding the effect of spreading techniques, band spreading or closed-slot injection appear to be both more effective than broadcast spreading.

In general, it should be considered that if emissions reduced immediately after manure production (e.g. diet manipulation), are then not reduced in later stages (e.g. storage or spreading), the emissions benefits at earlier stages are negated. At the same time, if emission reductions obtained during storage are not supported by mitigation strategies during manure application, part of the abatement benefit is wasted. Therefore, interventions need to be used in combination, spanning the whole lifecycle of manure production, storage and application (Oenema et al., 2012).

An alternative to the traditional manure management technologies is the use of air treatment technologies, such as chemical scrubbers or biological, e.g. biofilter, biotrickling filtration, and bio scrubbers (Dumont, 2018). In chemical scrubbers the odorous compound/pollutant is transferred in a packed tower from the gas emission to a chemical oxidant containing an aqueous phase, where they chemically react to be destroyed (Melse and Ogink, 2005). Converserly, biological techniques are based on the enzymatic oxidation of pollutants/odorants once they have been transferred from the gaseous emission to an aqueous phase, where the microorganisms responsible for this enzymatic oxidation are present. For microbial survival two main requirements occur: the presence of an aqueous medium and the availability of macro- and micronutrients (Van der Heyden et al., 2015). Chemical scrubbers are proved to be more effective in abating NH_3 emissions, whereas bioscrubbers and biofilters in reducing odor emissions (Ubeda et al., 2013). Acid scrubbers can remove NH_3 by up to 100%, biotrickling filters and bioscrubbers have an average 70% NH_3 removal efficiency (Dumont, 2018; Melse and Ogink, 2005), and biological air scrubbers fall between 64% and 86% (Van der Heyden et al., 2015; Van der Heyden et al., 2016). For chemical scrubber the NH_3 abatement efficiency is influenced by the kind of acid adopted, better results are obtained with sulphuric acid even if it is more dangerous during manipulation operation. The variation in the odor abatement is high, ranging from a minimum of 3% removal efficiency to a maximum of 51% (Melse and Ogink, 2005).

Compared to the above-mentioned mitigation strategies, air treatment technologies have a double advantage: i) reduce emissions released in the atmosphere, dropping air pollutants environmental impact and ii) improve air quality inside the farms, thus enhancing also health and welfare condition of animals and livestock workers.

In the future, a significant reduction in NH_3, odor, and GHG emissions is expected as a consequence of the abatement measures that are being introduced in agricultural and livestock farms. Other improving air quality they would contribute to achieving important environmental benefits. Reducing NH_3 involves reducing NH_3-related impact categories, such as acidification, eutrophication, and particulate matter formation, whereas cutting GHG helps to slow climate change. In literature, many studies are investigating, through the Life Cycle Assessment (LCA) method, the environmental implications of adopting mitigation strategies with positive results. LCA is a holistic approach for evaluating environmental impacts of products, processes and services throughout their “life cycle”, from production, to use, end-of-life and waste management (ISO, 2006). For example, Miranda et al. (2021) found that acidified slurry, compared to untreated slurry and biochar addition, showed the lowest environmental impact and the benefit was more evident when the impacts were expressed per kilogram of nitrogen in the slurry. Similarly, Fangueiro et al. (2015) reported that the slurry acidification of cattle separated liquid fraction decreased significantly NH_3 emissions and the global warming potential. Promising results can be obtained also through dietary manipulation (Anestis et al., 2020) and air scrubbers (De Vries and Melse, 2017). In particular, wet acid scrubber is the most applied air treatment technology in pig farming. According to Costantini et al. (2020), not only it is a promising technology in environmental terms, as it could play an important role in air pollutants control, but it could also reduce the annual impact on human health.

1.6 Novel food

The world population and the global food demand are increasing (United Nations, 2017), and, at the same time, humankind is consuming more resources than our planet is able to generate. The production of food requires more environmental resources (soil and water) than the available ones, other than economic resources and human labor. In this geo-political scenario, the need of sustainable solutions is required to ensure enough food and to optimize the use of resources in order to mitigate the hunger challenge and the alarming phenomenon of climate change (FAO, 2017). Sustainability is not only the responsibility to conserve natural resources and protect global ecosystems, but includes also reduction of food waste, reduction of overconsumption of calories and a shift towards more sustainable diets (FAO, 2013). It is common knowledge that livestock production plays an important role in GHG emissions and global warming. Besides GHG emissions, livestock production also contributes to natural resource depletion (land, water and fossil energy) and biodiversity loss (Gerber et al., 2013). Halving the consumption of meat would reduce NH_3 emissions by 40% and GHG emissions from 25% to 40% (Westhoek et al., 2014). Meat substitutes can reduce meat consumption. Plant-based alternatives currently play a big role but are not sufficient to meet the increasing global food demand. Edible insects have always been a part of human diets, but in some societies there is a degree of distaste for their consumption (FAO, 2013). It is estimated that in over 130 countries and for around 2 billion people, beetles, maggots and crickets are a traditional part of the everyday diet, but usually not in Western developed countries. Nutritionally, insects are rich in proteins (40% to 70% of dry matter depending on development stage, species and feeding type), with a good amino acid profile, and a high content of essential amino acids, lipids, vitamins, fiber and minerals (FAO, 2013; Rumpold and Schlüter, 2013). Also earthworms are eaten in some non-Western countries and, similarly to insects, are rich in proteins and essential amino acids (Zhenjun et al., 1997). Besides being an alternative source of proteins, earthworms, thanks to their activity in the soil, offer many benefits: increased nutrient availability, better drainage, and a more stable soil structure (Singh et al., 2016). The main advantage of rearing insects/earthworms is that they require less space, feed, and water than traditional livestock farming (van Huis and Oonincx, 2017). The disadvantage is that, if they are not eaten as a whole but as an ingredient included in a product, their production requires more energy inputs than conventional livestock due to the heating and drying processes, needed to transform them into meals (Salomone et al., 2017). In the EU the consumption of insects is regulated by novel food Regulation (EU) 2015/2283, and, at the moment, *Tenebrio molitor*, *Alphitobius diaperinus*, *Acheta domesticus*, *Locusta migratoria*, and *Hermetia illucens* are species admitted and sold in some countries (EU, 2015). Although neo-phobia is the primary instinct towards untraditional and unfamiliar food, the necessity to tackle the climate crisis would move consumers towards more sustainable diets, including artificial meat, insects or pulses as sources for proteins in the human diet. In literature, some studies have already focused on the environmental sustainability of terrestrial invertebrates farming and diets by life cycle assessment. As an example, Smetana et al. (2016) confirmed that the production of insect-based protein powder and meat substitutes, based on food by-products, is 2–5 times more environmentally beneficial than that of traditional products. The environmental advantages of insect farming were also deeply discussed by van Huis and Oonincx (2017). Finally, to summarize, terrestrial invertebrates farming presents different advantages: i) they require little land and water; ii) greenhouse gas emissions are lower; iii) they have high feed conversion efficiencies; and, iv) finally, they can transform low-value organic by-products, such as food waste, into high-quality food or feed.

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CHAPTER 2

2. Framework of the thesis

2.1 Aim

There is a raising interesting in air quality, especially after Covid-19 spread, and in the identification and possibilities offered by mitigation strategies for each specific production sector. Livestock farming largerly contributes to GHG, NH_3 and odor emissions and needs to abate them to for a sustainable production. The general objective of this thesis is to investigate the impact of livestock farming, especially in terms of airborne emissions, and how it could be reduced thanks to the application of abatement measures, suggested to reduce the environmental impact and to improve livestock sustainability. The mitigation solutions were tested in the Italian context, in particular in farms located in the Po Valley, an area deeply affected by air pollution. The environmental benefits were assessed applying the LCA methodology that also allows a broader evaluation of the environmental burdens associated to livestock farming.

The specific objectives of this thesis are:

- Identify the contribution of livestock farming to the global gaseous emissions; evaluating in particular its contribution to NH_3 emissions (especially before and during Covid-19 pandemic);
- Give an overview of techniques and models to assess odor annoyance;
- Describe mitigation strategies to abate odor and gaseous emissions deriving from livestock sector;
- Assess odor and NH_3 removal efficiency of air treatment technologies (i.e. dry filter and wet acid scrubber) in a pig farming context;
- Evaluate the effect of an additive on GHG and NH_3 emissions during cattle slurry storage;
- Estimate the environmental benefits generated by the adoption of mitigation techniques with the LCA approach;
- Investigate alternative sustainable strategies, in particular different protein sources (i.e. earthworms) and calculate their environmental impact.

2.2 Overview of the chapters

The PhD thesis is mainly composed of two sections:

- introduction to the topics (Chapter 1), and goal and scope of the research project (Chapter 2);
- scientific contributions published within the scopes of the Thesis to support the contribution of livestock activities to air pollution and the achievable environmental sustainability of livestock farming by appling different mitigation strategies (Chapter 3, 4, 5, and 6). For each article, the reference with complete bibliographic details is provided.

As shown in Figure 1, the following steps were performed. Firstly, livestock sector is confirmed to play an important role for NH_3 emissions and concentration levels, also during Covid-19 pandemic. In particular, Chapter 3 (*Paper 1 and 2*) describes both the trend of ammonia, particulate matter and nitrogen oxides, and the changes in NH_3 atmospheric concentration in northern Italy during Covid-19 quarantine, paying particular attention on the role and contribution of livestock activities. The focus on norhern Italy is justified by the presence of the Po Valley, one of the most disadvantaged areas in Europe for air quality. Indeed, its geographical and physical characteristics and the high concentration of anthropogenic emissions (intensive livestock farms and industries), both favor pollutants stagnation. As a confirmation of this last aspect, more than 50% of the Italian pig and cattle farms are located in this area.

Then, the second big problem in areas where there is a high density of livestock activities close to residential areas is odor annoyance. Intensive livestock farming, especially swine operations, extensively contributes to NH_3 and odors emissions, that mainly derive from the storage, handling and land application of animals wastes (slurry or manure). Chapter 4 (*Paper 3*) focuses on odor emissions, one of the most relevant air quality issues of intensive livestock production, but also of waste treatment plants. The review wants to give an overview of odor emissions sources, techniques and models for measuring odors and simulating their dispersion, and suggests some mitigation strategies to protect people from odor nuisance.

A further step of the research is presented in Chapter 5 (*Paper 4, 5 and 6*), where mitigation strategies aimed at reducing emissions deriving from pig and cattle farming are discussed. A large number of mitigation techniques are available to reduce NH_3, odor and GHG emissions, but only two kinds of air treatment technologies (i.e. dry filter and wet acid scrubber) and an additive were taken in consideration. Papers 4 and 5 show odor and NH_3 abatement removal efficiencies obtainable from air treatment technologies, in particular from a dry filter and a wet acid scrubber (with citric acid solution). In addition to reduce odor and NH_3 emission, these two tested systems are also useful to improve air quality inside the barns. In Paper 6, an additive based on gypsum was applied on cattle slurry to test its effectiveness on NH_3 and GHG emissions arising during storage. As mitigation strategies could affect livestock environmental impact, in Papers 5 and 6 the Life Cycle Assessment (LCA) method was applied to identify the environmental processes with the greatest impact, and to assess the potential environmental impact related to the implementation of a mitigation solution.

Finally, as mitigation strategies are not always sufficient to abate livestock impact, but sustainable solutions are required to satisfy the increasing world population and global food demand, in the last Chapter - Chapter 6 (*Paper 7 and 8*) – earthworms rearing is presented as an alternative solution. Indeed, edible terrestrial invertebrates could contribute to overcoming the problem of feeding a growing population. To demonstrate their environmental sustainability, also in this case, the LCA method was adopted.

Finally, Chapter 7 discusses and draws the general conclusions of the previous chapters.

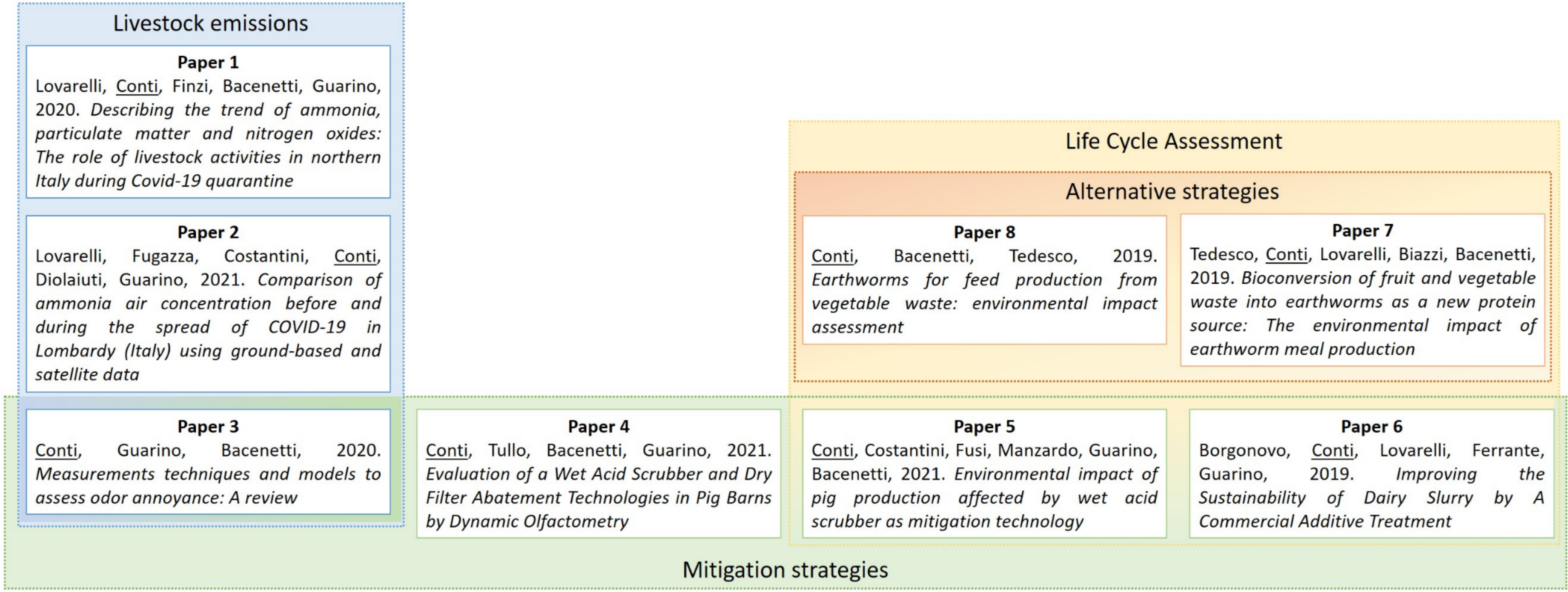

Figure 1. Sequence of scientific contributions.

CHAPTER 3

This chapter consists of two published peer reviewed articles (*Paper 1 and 2*). The aim of this chapter is to evaluate differences in air pollutants emissions and concentrations deriving from livestock activities during lockdown periods imposed by Italian Governemental Authorities immediately after the spread of Covid-19. In both papers the focus was on the Po valley area as it is an area characterized by a large concentration of intensive livestock farms and because it represents an important exceedance zone of the air-quality limit values.

The first paper reports results on NH_3, PM, and NOx emissions differences between quarantine period respect to 2016–2019 using data collected from the database of the Regional Agency for the Protection of the Environment, whereas the second paper focuses on NH_3 concentration before and during the spread of Covid-19 using ground-based and satellite data.

3. Describing the trend of ammonia, particulate matter and nitrogen oxides: The role of livestock activities in northern Italy during Covid-19 quarantine

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Abstract

Nitrogen oxides (NOx), sulphur oxides (SOx) and ammonia (NH_3) are among the main contributors to the formation of secondary particulate matter ($PM_{2.5}$), which represent a severe risk to human health. Even if important improvements have been achieved worldwide, traffic, industrial activities, and the energy sector are mostly responsible for NOx and SOx release; instead, the agricultural sector is mainly responsible for NH_3 emissions. Due to the emergency of coronavirus disease, in Italy schools and universities have been locked down from late February 2020, followed in March by almost all production and industrial activities as well as road transport, except for the agricultural ones. This study aims to analyze NH_3, $PM_{2.5}$ and NOx emissions in principal livestock provinces in the Lombardy region (Brescia, Cremona, Lodi, and Mantua) to evaluate if and how air emissions have changed during this quarantine period respect to 2016–2019. For each province, meteorological and air quality data were collected from the database of the Regional Agency for the Protection of the Environment, considering both data stations located in the city and the countryside. In the 2020 selected period, $PM_{2.5}$ reduction was higher compared to the previous years, especially in February and March. Respect to February, $PM_{2.5}$ released in March in the city stations reduced by 19%–32% in 2016–2019 and by 21%–41% in 2020. Similarly, NOx data of 2020 were lower than in the 2016–2019 period (reduction in March respect to February of 22–42% for 2016–2019 and of 43–62% for 2020); in particular, this can be observed in city stations, because of the current reduction in anthropogenic emissions related to traffic and industrial activities. A different trend with no reductions was observed for NH_3 emissions, as agricultural activities have not stopped during the lockdown. Air quality is affected by many variables, for which making conclusions requires a holistic perspective. Therefore, all sectors must play a role to contribute to the reduction of harmful pollutants.

Keywords: Air quality; Ammonia; Livestock; Particulate matter; Quarantine

3.1 Introduction

Air quality is a big issue in developed countries. The most developed countries and industrialized cities, characterized by important economic exchanges, traffic and highly-density populated commonly have bigger problems than others with air pollution. Several studies are available on these aspects, for example, some Chinese (Xiao et al., 2020; Wang et al., 2015) and European (EEA, 2018; Izquierdo et al., 2020; Koolen and Rothenberg, 2019) cities are recognized as highly polluted.

According to the World Health Organization (WHO) (WHO, 2013), ground-level ozone (O_3), nitrogen dioxide (NO_2) and particulate matter (PM) are the most harmful air pollutants to human health and ecosystems. Among these, PM has been most closely studied due to its adverse health effect. PM identifies fine particles that can have both a primary and secondary origin. In the first case, they are emitted to the atmosphere directly from their sources, such as road traffic and car exhaust gases. Instead, secondary PM precursors are pollutants (e.g. NH_3, NOx, and SO_2) that are partly transformed into particles by photochemical reactions in the atmosphere (Koolen and Rothenberg, 2019). In more detail, air pollution has a direct effect on human health (Boldo et al., 2014), especially for what regards the long exposure to high concentrations of $PM_{2.5}$ (Yang et al., 2019). This is known to cause respiratory and cardiovascular diseases (Dominici et al., 2006) since it can penetrate deeply into the lung and translocate to blood circulation (Li et al., 2018). According to EEA (2019a), in Europe, about 400,000 premature deaths per year are attributable to $PM_{2.5}$ concentrations long-term exposure. Therefore, investigating how to reduce its presence in air is an important issue for society. As regards ecosystems, air pollution is known to cause damages to vegetation and ecosystems, such as eutrophication and soil acidification, which finally lead to biodiversity loss (EEA, 2019b). Moreover, excessive PM concentration causes undoubted problems not only in farms neighboring residents (de Rooij et al., 2017), but also in livestock buildings and on workers' and animals' health (Conti et al., 2020).

On a global scale, the benchmark limit for PM_{10} set at 20 μg/m^3 and $PM_{2.5}$ set at 10 μg/m^3 by the WHO is often not respected (EEA, 2019a). It is estimated that around 90% of the global population in 2018 was breathing polluted air (WHO, 2018), in particular, 6–8% of the European population was exposed to $PM_{2.5}$ exceeding limit and 13–19% to PM_{10} exceeding limit (EEA, 2019a). The European Union set the yearly limit for PM_{10} to 50 μg/m^3 and $PM_{2.5}$ to 25 μg/m^3 (Directive, 2008/50/EC), but although it is less restrictive than the one by WHO, some countries are not able yet to respect it every day of the year; among these, Italy is an example (Kiesewetter et al., 2015). To date, in Lombardy, one of the most productive Italian regions, the measured annual average concentrations of $PM_{2.5}$ range between 10 and 31 μg/m^3 and those of PM_{10} from about 20 to 38 μg/m^3 (Fattorini and Regoli, 2020). In 2017, among all the Lombardy provincial chief towns, only in Lecco, Sondrio, and Varese the annual average concentrations were lower than the limit value. Instead, Milan, Brescia, Cremona and Mantua were the cities that most exceeded the $PM_{2.5}$ annual limit value (PRIA, 2018).

For this series of reasons, some researchers (Wang et al., 2020a; Wu et al., 2020) have started believing that the global spread of Covid-19 has reinforced in areas characterized by bad air quality.

Focusing on Italy, the Po valley – in the Northern part of the country, where also Lombardy region is located - is a highly industrialized and densely populated area characterized by a large concentration of intensive livestock farms (Arvani et al., 2014). These characteristics are also causing pollution, and in addition to them, also geographical and physical characteristics of the Po valley facilitate the persistence of pollutants (Fattore et al., 2011). In particular, they make Po valley one of the most disadvantaged areas in Europe for air quality. First of all, this area is characterized by the presence of the mountain chains of Alps and Apennines on three sides, which affects the pedo-climatic variables, reduces the air exchanges and negatively affects the local air quality. Secondly, the area is poorly ventilated favoring the stagnation of pollutants and consequently contribute to the accumulation of PM (Carugno et al., 2016). Lombardy is located in the middle of the Po valley and it is Italy's leading industrial and agricultural area (Lovarelli et al., 2020), whose emissions (methane, ammonia, dinitrogen monoxide, etc.) worsen the air quality issue (Rebolledo et al., 2013). To confirm this, Lombardy ranks among the most air polluted areas of Europe (Carugno et al., 2016). Several studies highlighted that the unfavorable geographical context, climate characteristics, and intense anthropogenic activities, such as industry and agriculture, of the Po valley promote a high level of air pollution (Arvani et al., 2014; Diémoz et al., 2019; Thunis et al., 2009). For example, Giannakis et al. (2019) found that the highest $PM_{2.5}$ concentration from the agricultural sector in Europe is present in northern Balkan countries and Northern Italy. Similarly, Thunis et al. (2019) identified as "hotspots" regions for PM emissions the Po valley and Eastern Europe. In addition, by 2050, global ammonia (NH_3) emissions are estimated to further increase because of agricultural intensification (Rebolledo et al., 2013). Emissions of NH_3 are an important aspect to evaluate, in fact it was estimated that 640 kg of PM_{10} can be formed per ton of NH_3 emitted and that 880 kg PM_{10} are formed per ton of nitrogen oxides (NOx) emitted (De Leeuw, 2002).

With the 2020 quarantine caused by the pandemic coronavirus, in Italy, many production and industrial activities have started being locked down from March, except for agricultural ones. The main evidence of this is related to the reduction in traffic: respect to the first part of February 2020, the beginning of March 2020 points out a reduction of about 90% of car traffic and about 50% of heavy vehicles (Buganza et al., 2020). Consequently, it is interesting to identify if and how air emissions are affected by agricultural activities in a polluted area such as Northern Italy, and in particular Lombardy, the Italian region most affected by Covid-19 (Fattorini and Regoli, 2020) and where the lockdown was the most severe (Bontempi, 2020), in a period in which most of the productive activities have been stopped while agricultural-related ones have remained active.

This study aims to analyze the main air emissions related to livestock activities in the cities and provinces of Lombardy that are mostly dedicated to livestock productions. This is carried out to evaluate if and how air emissions have changed in the quarantine period. In particular, from the comparison between the data about air emissions in data stations located in the main cities and those located in small cities or countryside stations, where livestock is the main activity, it is expected to identify a reduction in emissions due to the strong limitation of the industrial sector and transport, but a low reduction of those emissions related to agricultural activities.

3.2 Background

With the lockdown of work activities as a consequence of Covid-19, it can be expected that:

(i) anthropogenic emissions caused by industries, energy-related industries, and traffic deeply reduce,

(ii) emissions caused by agricultural activities should maintain a usual trend.

In Italy, agriculture is known to be responsible for more than 90% of NH_3 emissions (EEA, 2019c), in particular in Lombardy region they account for around 97% of all NH_3 emissions, corresponding to yearly 94,000 tons (INEMAR, 2020). Therefore, it can be assumed NH_3 did not vary in this analyzed quarantine period, except for possible different meteorological aspects. In particular, NH_3 emissions from the agricultural sector derive mainly from manure management and the application of fertilizers (Oenema et al., 2012), among which the superficial slurry spreading. Slurry spreading is generally carried out previous to soil preparation for crops sowing. Because this operation occurs seasonally every year and because agricultural activities have not been stopped during quarantine, emissions of NH_3 should not be subject to variations caused by the quarantine, instead of by the meteorological trend. In fact, NH_3 is a volatile gas whose amount is influenced by temperature, wind speed, and rainfall, other than by livestock housing, storage, and field spreading practices (Anderson et al., 2003; Welch et al., 2005). Given the dependence of agricultural field operations from weather conditions and given the European regulation for organic fertilizers spreading (Nitrate Directive) (Directive 91/676/EEC), the slurry is commonly spread between the end of winter and early spring before soil tillage and summer crops' sowing, as well as in autumn after the harvest of summer crops and before sowing of winter crops. To this, the Directive norms also organic nitrogen application (Directive 91/676/EEC, 1991). In particular, this Directive obliges to avoid the slurry spreading in the months in which no crop requirements for nutrients occur, and consequently in which runoff and leaching occur the most. Moreover, it is also forbidden to spread slurry when rainfall occurs or is expected to occur in the following days. For these reasons, NH_3 emissions can differ on a seasonal and a daily basis. One additional aspect related to NH_3 is that it plays an important role in atmospheric chemical reactions that bring to the formation of secondary PM_{10} and $PM_{2.5}$ (Koolen and Rothenberg, 2019). However, it should be considered that the quantity of secondary particulate matter that is formed in the atmosphere starting from the latter is variable in time and space and depends on non-linear processes and meteorology (Marongiu et al., 2020). PM can be classified as primary or secondary according to its origin and it can be both organic and/or inorganic in nature (Cambra-Lopez et al., 2010). In the atmosphere, NH_3 reacts with atmospheric nitric and sulfuric acids to form particulate sulphate (SO_4^{2-}), nitrate (NO_3^-) and ammonium (NH_4^+) compounds, which constitute the major fraction of secondary inorganic $PM_{2.5}$ (Behera et al., 2013; Wang et al., 2020a). With the reduction in the release of NOx and sulphur oxides (SOx) from industrial activities, energy-related industries and traffic, $PM_{2.5}$ reduces only partially (EEA, 2018). It can be assumed that if NH_3 is not subject to changes during the Covid-19 quarantine

(Marongiu et al., 2020), $PM_{2.5}$ may also reduce only partially, because the decrease in pollutant emissions caused by transport and industrial activities are not sufficient to avoid the chemical reaction (Wu et al., 2016). Regarding the agricultural sector, the main sources of PM emissions are buildings housing livestock, in particular those in which are carried out the feed operations, which account for 80%–90% of total PM emissions from the agriculture sector. Pig and poultry livestock farms are the main sources of PM (EEA, 2019b) from the agricultural sector. Other than the just mentioned sources, particulate matter emissions from pig houses arise also from skin particles, faeces, and bedding, while emissions from poultry housing from feathers and manure (EEA, 2019b). Together with these categories, cattle farming brings to relevant NH_3 and PM emissions, mainly from feed operations (Brown et al., 2018), feedlots (McGinn et al., 2010), barn, and storage tanks. In Lombardy, in 2017, $PM_{2.5}$ and PM_{10} emissions deriving from agriculture accounted for 4% and 6%, respectively (INEMAR, 2020).

3.3 Materials and methods

3.3.1 Monitored area

Lombardy houses 38% of the farms specialized in cattle milk production (ISMEA, 2019a), 10% of the farms specialized in cattle meat production (ISMEA, 2019b), and 11% of the pig farms for heavy pig production (ISMEA, 2019c), reaching a total number of reared animals equal to 1,543,639 cattle and 3,984,633 pigs (ISTAT, 2019).

Four provinces have been identified as the most livestock-intensive of the region. They are Brescia, Cremona, Lodi, and Mantua that altogether represent about 77.5% of the total Lombardy livestock production (ERSAF, 2019). Such a livestock intensity makes pressure on the territory, making most of the region susceptible to nitrates leaching, and most of the fields are recognized as Nitrate Vulnerable Zones (NVZ) in the context of the Council Directive 91/676/EEC. In NVZ, 170 kg/ha of N from organic origin can be spread on fields. Commonly, farms are equipped with traditional machinery for slurry spreading, which consists of a superficial slurry spreading through slurry tankers equipped with diverter plates (Bacenetti et al., 2016a). This machine undoubtedly contributes to the volatilization of NH_3 during spreading and also in the few subsequent hours. NH_3 volatilization worsens also when optimal meteorological conditions occur (e.g., mild temperatures, low wind speed, and no rainfall) (Brentrup et al., 2000).

3.3.2 Data collection

Meteorological data and air quality data were collected from the database of the regional agency for environmental protection (ARPA, 2020). Data of ARPA (ARPA, 2020) were downloaded from different ground-based stations per province. The period investigated was January, February, and March of the years 2016–2019 and 2020. Data of years 2016–2019 were averaged to reduce the annual seasonality and were compared with 2020. These months were chosen since Covid-19 started spreading in the Lombardy region from January and almost all production activities have been locked down from February, except for agricultural ones.

The meteorological data used were air temperature (T, ◦C), relative humidity (RH, %), wind speed (W, m/s), and rainfall (R, mm). The air quality data used were ammonia (NH_3, μg/m^3), nitrous oxides (NOx, μg/m^3), and secondary particulate matter ($PM_{2.5}$, μg/m^3). SOx were not included since these data were not available on the ARPA website. Particulate matter on the order of 10 μm (PM_{10}) was investigated only in an initial phase because it is caused by multiple sectors (transport, heating systems, energy sector, etc.) therefore its formation is not only due to agricultural activities and attributing its effect to one single sector may be misleading (Ansari and Pandis, 1998). Instead, since with PM_{10} are intended particles with a diameter equal or less than 10 μm, PM_{10} includes the smaller $PM_{2.5}$, and their trends can be compared. Moreover, because secondary $PM_{2.5}$ is partially formed from NH_3 released from livestock activities and the effect of $PM_{2.5}$ on health and ecosystem is damaging (Wang et al., 2020b), in this study $PM_{2.5}$ was analyzed in more detail instead of PM_{10}.

Fig. 1 summarizes the research framework and air quality detection and meteorological stations referred to every province analyzed. In every province (i.e. Brescia, Cremona, Lodi, and Mantua), the stations with the availability of the pollutants NH_3, NOx, and $PM_{2.5}$ were used, except for Brescia where no detection stations for NH_3 emissions were available. All of them were distinguished in stations in the city and the countryside, to investigate the effect of pollutants in the city where most people live, traffic jams may occur and industrial activities are carried out, respect to the countryside, in which the main activities are related to livestock farms. Therefore, with “city” authors refer to those data stations located in a densely populated settlement whose members work primarily on non-agricultural tasks, whereas with “countryside” those located in a rural area mainly used for farming activities (field cultivation and livestock). As mentioned, together with pollutants, the meteorological data (temperature, relative humidity, wind speed, and rainfall) were collected by the same stations for the identified periods. In total, 14 data stations were analyzed for each of the 2 periods (January–March 2016–2019 vs January–March 2020). The final dataset was made of about 6000 data for every emission and meteorological variable. Statistical analyses were conducted using SAS version 9.4 (SAS Institute, Cary, NC, USA) statistics software. The mean and standard deviation of weather parameters for the two periods considered were calculated. Descriptive statistics were calculated using a means procedure in SAS. Principal Components Analysis (PCA), Factor Analysis (FA), and a general linear model (GLM) were used to identify relationships among variables and test the resulting model.

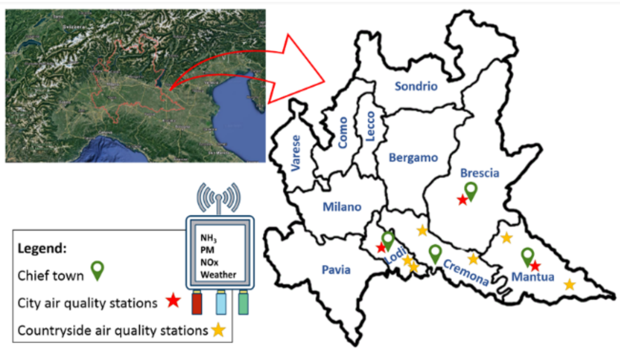

Figure 1. Research framework for data collection

3.4 Results and discussion

For what concerns the analysis of the meteorological aspects, Table 1 reports the average data of the 4 provinces for mean, standard deviation (SD) and minimum and maximum values for temperature (T), relative humidity (RH), wind speed (W) and rainfall (R) in the period of January–March for 2016–2019 and 2020. From the results, it emerges that the weather conditions show reduced differences in the selected years 2016–2019 vs 2020. No events of strong wind speed or heavy rainfall were highlighted for these periods. Moreover, no differences can be observed between city and countryside stations in regard to average weather parameters, since in Italy, especially in the Po valley, agricultural areas are closed to cities due to population density.

Table 1. Mean, standard deviation (SD), minimum and maximum values for daily weather parameters in the weather stations of the Lombardy region during the evaluated periods.

Year	Parameter	January		February		March	
		mean (SD)	Min-Max	mean (SD)	Min-Max	mean (SD)	Min-Max
2016-2019	W (m/s)	1.26 (0.3)	0.79-2.04	1.47 (0.45)	0.94-3.35	1.58 (0.28)	1.23-2.24
	RH (%)	75.72 (8.4)	60.99-95.6	80.2 (8.25)	64.23-98.63	67.99 (7.8)	55.63-85.48
	T (°C)	3.18 (0.84)	1.58-5.32	6.1 (0.82)	4.5-8.91	10.15 (2.04)	6.3-14.25
	R (mm)	0.53 (0.89)	0-3.99	2.48 (3.56)	0.01-18.65	1.25 (1.59)	0-5.06
2020	W (m/s)	1.29 (0.3)	0.88-2.2	1.69 (0.76)	0.83-3.48	1.75 (0.68)	0.98-3.35
	RH (%)	90.91 (7.61)	66.58-99.53	71.1 (20.5)	31.73-99.63	73.21 (14.64)	40.4-97.5
	T (°C)	4.19 (1.94)	0.35-7.28	8.32 (1.64)	5.45-11.03	9.86 (2.73)	6.05-14.9
	R (mm)	0.68 (2.32)	0-12.7	0.08 (0.18)	0-0.85	1.61 (3.23)	0-10

Notes: W = wind speed; RH = relative humidity; T = temperature; R = rainfall.

To support the fact that PM_{10} and $PM_{2.5}$ maintain a similar trend over time, Fig. 2 reports the average daily trend of PM_{10} and $PM_{2.5}$ in the city of Brescia for the period January–March 2019, to which are associated the wind speed and rainfall events of the same period. In particular, it is possible to notice that air pollutants generally reduce when wind and rainfall events occur. Although not reported, a similar trend was observed also for the other 3 provinces considered (Cremona, Lodi, and Mantua) (available in Supplementary Material).

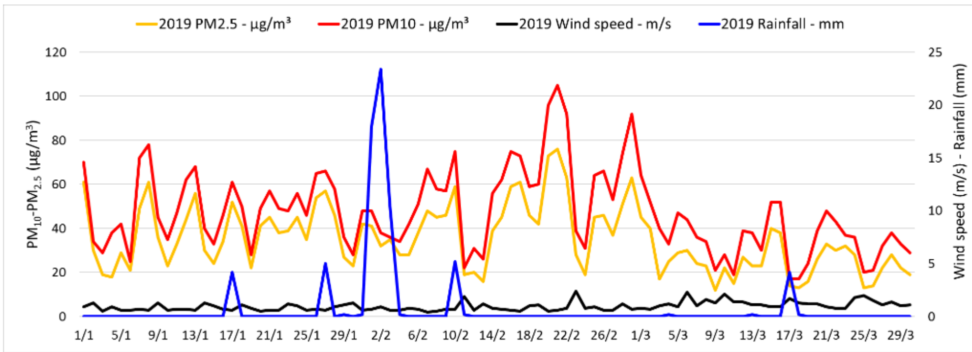

Figure 2. Trend of PM_{10} (red line) and $PM_{2.5}$ (yellow line) in the period January-March 2019 for the city of Brescia. Wind speed (black line) and rainfall events (blue line) are also shown.

Analyzing these data, the average presence of $PM_{2.5}$ in air changes as a consequence of meteorological variables; $PM_{2.5}$ in the air after rainfall events is on average 28.14 (±9.97) µg/m^3, whereas if no rainfall occurred this value was equal to 37.07 (±14.76) µg/m^3. Similarly, although the wind speed is generally low (<1.6 m/s or <6 km/h), when higher wind speed occurred, $PM_{2.5}$ for the studied period of January–March was on average 17.17 (±4.62) µg/m^3, while with low wind speeds it was equal to 40.36 (±13.87) µg/m^3.

In order to analyze the trend of emissions in the two periods considered, the air quality stations were grouped as follows: all data stations were split between stations located in the city ("city") and the countryside ("country"). Average values were calculated with data from Brescia, Cremona, Lodi, and Mantua data stations for each pollutant. Hence, for each pollutant (NOx, NH_3, and $PM_{2.5}$) and each period (2016–2019 and 2020) are available average data for "city" and "country" stations. Fig. 3 reports these results.

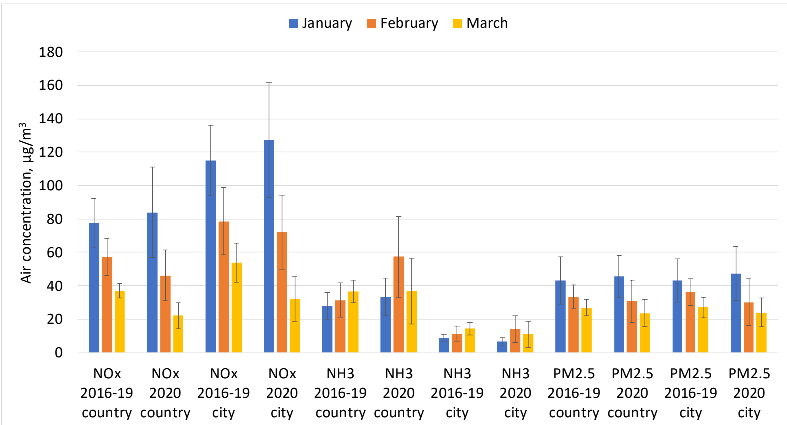

Figure 3. Results of average NOx, NH_3 and $PM_{2.5}$ emission in country and city stations of Lombardy for the period January-March of 2016-2019 and 2020. Bars refer to standard deviation.

From these results emerges that emissions of NOx and $PM_{2.5}$ are in all cases higher in January and follow a common reduction trend in February and March, which is common in all periods analyzed. Moreover, the emission of NOx and $PM_{2.5}$ were higher in January 2020 than in the same month of the previous years, although standard deviations are quite wide. NOx records the highest values in January (the coldest month), with an average of the city stations equal to 115.1 µg/m^3 in 2016–2019 and to 126.0 µg/m^3 in 2020. In the countryside stations, these values were equal to 76.4 μg/m^3 in 2016–2019 and 83.5 μg/m^3 in 2020. Indeed, NOx emissions gradually decrease from January to March in both periods considered, mainly because of the lower use of heating systems with milder temperatures.

In 2020, and in particular, in February and March, the reduction in NOx and $PM_{2.5}$ emissions is bigger than in the previous period and is even bigger in the city stations than in those in the countryside, where are mostly located the livestock and agricultural activities. NOx in March 2020 amounted to 22.03 (±7.72) μg/m^3 in countryside stations and 31.92 (±13.45) μg/m^3 in city stations, whereas in the previous period 2016–2019 they amounted to 36.88 (±4.28) μg/m^3 and 53.84 (±11.85) μg/m^3, respectively in countryside and city stations. This highlight considerable reductions in both city and countryside stations, partially motivated by the mentioned milder temperatures (on average for January–March 2020, 7.46°C) that allowed reducing home heating systems. Therefore, following the adoption of the Ministerial Decree (DPCM March 8, 2020), which introduced measures to limit travels, and following the reduced adoption of home heating system, NOx emissions decreased in March more than in the other months. In particular, the difference between March and February 2016–2019 ranged from 22.5% to 41.9%, while in the same period for 2020, NOx reduced by 43.3%–61.5%.

Regarding $PM_{2.5}$, the emission in the cities for March ranges between 19% and 32% respect to February for the period 2016–2019 and between 21% and 41% respect to February in 2020, thus with a considerable reduction that characterized firstly the area of Lodi that was the first to be locked down (DPCM February 23, 2020). Respect to January, in some of the evaluated provinces, the reduction of $PM_{2.5}$ even reached 65% in 2020, while in the previous period 2016–2019 it did not exceed 46%. In the countryside stations, the reduction of $PM_{2.5}$ in March respect to February ranged between 16% and 26% in 2016–2019 and between 18% and 43% in 2020. Respect to January 2020, also in the countryside areas the $PM_{2.5}$ reduction in March reached 57%. Such a reduction for $PM_{2.5}$ is due to the fact that some of the pollutants co-participating to the formation of $PM_{2.5}$ have reduced. As reported in Section 1.1, in fact, $PM_{2.5}$ derives from a chemical reaction in which NH_3, NOx, and SOx pollutants can participate. This latter aspect has been highlighted by the study of Collivignarelli et al. (2020), and even if they focused on the area of Milan, they found that $PM_{2.5}$ concentrations were almost halved during the lockdown period probably due to the precursors (e.g. NOx) reduction. However, focusing on March 2020, $PM_{2.5}$ was equal on average to 23.48 (±8.17) μg/m^3 and 23.99 (±8.59) μg/m^3, respectively in countryside and city stations, while in the period March 2016–2019, the average values were 26.8 (±4.8) μg/m^3 and 26.92 (±6.04) μg/m^3, respectively for countryside and city. For $PM_{2.5}$, therefore, no evident differences emerge between countryside and city stations; however, some small differences emerge between 2020 and the previous years, which can be mainly due to the reduction in car traffic during the lockdown.

As emerges from Fig. 3, however, the emission of NH_3 did not reduce in 2020 respect to the same period of 2016–2019. The reason is related to the fact that for agricultural activities no restriction was imposed during the quarantine. Therefore, agricultural activities, and in particular slurry spreading on the field, took place (similar to previous years) in the analyzed period.

Values of NH_3 in the countryside are similar between 2016 and 2019 and 2020 in January (28.0 ± 8.1 and 33.1 ± 11.3 μg/m^3 in 2016–2019 and 2020, respectively) and March (36.5 ± 6.6 and 36.8 ± 19.8 μg/m^3 in 2016–2019 and 2020, respectively), which was expected; however, they were higher in February 2020 respect to the previous years, probably because of the lack of possible slurry spreading events in the previous autumn. This condition was caused by a particularly rainy period that obliged farmers to avoid the slurry spreading in autumn and introduce more spreading interventions in February 2020. In February 2020, in fact, NH_3 emissions are about the double than in the 2016–2019 period (31.4 ± 10.2 μg/m^3 in 2016–2019 and 57.5 ± 24.2 μg/m^3 in 2020). The higher NH_3 emission in the countryside is also reflected by slight increase respect to February 2016–2019 in the stations located in the cities (11.3 ± 4.6 μg/m^3 in 2016–2019 and 14.0 ± 7.8 μg/m^3 in 2020). Respect to the NH_3 emission in the city stations occurred in March, the average values were found equal to 14.2 ± 3.5 μg/m^3 in 2016–2019 and 10.9 ± 7.8 μg/m^3 in 2020, thus with values considerably lower than in the countryside areas. No strong distances can be observed between city and countryside areas, but specifically for NH_3 emission this difference can be relevant.

From the statistical analysis carried out with these data, a Pearson's correlation matrix is reported in Table 2 and in Table 3, where the main relationships among the identified parameters can be highlighted for the studied periods 2016–2019 and 2020. The statistical analyses were carried out separately between 2016 and 2019 and 2020 in order to better focus on emissions during Covid-19 quarantine. The considered parameters include air quality data of NOx, NH_3, and $PM_{2.5}$ from city and countryside stations as well as weather data of temperature, wind speed, relative humidity, and rainfall for city and countryside stations. A good correlation has been considered for values equal to or higher than 0.6. In particular, a good correlation emerges among pollutants, especially between NOx and $PM_{2.5}$ ($r \geq 0.74$), both for city and countryside data. Instead, NH_3 is well correlated only between NH_3 in the countryside station and NH_3 in the city station ($r = 0.88$). Relative humidity and temperature have good correlations with $PM_{2.5}$, NOx and NH_3 ($r \geq 0.60$), while wind speed and rainfall show small correlations. Regarding 2020, the correlations are similar: NH_3 is well correlated with itself ($r = 0.86$), while $PM_{2.5}$ and NOx are well correlated with each other ($r \geq 0.76$). Once more, correlations are obtained with temperature and relative humidity but not interestingly with wind speed and rainfall.

Table 2. Pearson's correlations for the period 2016-2019. In bold correlations r ≥ 0.60.

Parameters	NOx 2016-19 city	NH_3 2016-19 country	NH_3 2016-19 city	$PM_{2.5}$ 2016-19 country	$PM_{2.5}$ 2016-19 city	Wind 2016-19	RH 2016-19	T 2016-19	Rain 2016-19
NOx 2016-19 country	**0.97**	0.30	0.05	**0.81**	**0.79**	0.13	**0.68**	-0.35	0.03
NOx 2016-19 city		0.26	0.01	**0.74**	**0.74**	0.10	**0.64**	-0.40	-0.02
NH_3 2016-19 country			**0.88**	0.42	0.42	0.46	0.47	**0.60**	-0.12
NH_3 2016-19 city				0.25	0.26	0.47	0.36	**0.73**	-0.03
$PM_{2.5}$ 2016-19 country					**0.98**	0.14	**0.74**	-0.14	0.00
$PM_{2.5}$ 2016-19 city						0.14	**0.77**	-0.16	0.03
Wind 2016-19							0.56	0.59	0.26
RH 2016-19								0.15	0.35
T 2016-19									0.09

Table 3. Pearson's correlations for the period 2020. In bold correlations r ≥ 0.60.

Parameters	NOx 2020 city	NH_3 2020 country	NH_3 2020 city	$PM_{2.5}$ 2020 country	$PM_{2.5}$ 2020 city	Wind 2020	RH 2020	T 2020	Rain 2020
NOx 2020 country	**0.96**	0.14	-0.04	**0.79**	**0.80**	-0.08	**0.65**	-0.33	-0.09
NOx 2020 city		0.19	-0.05	**0.76**	**0.77**	-0.07	**0.62**	-0.32	-0.09
NH_3 2020 country			**0.86**	0.32	0.28	0.12	0.20	0.57	-0.08
NH_3 2020 city				0.23	0.20	0.03	0.18	**0.63**	-0.01
$PM_{2.5}$ 2020 country					**0.99**	-0.02	**0.76**	0.03	-0.10
$PM_{2.5}$ 2020 city						-0.08	**0.75**	-0.03	-0.08
Wind 2020							0.22	0.54	0.13
RH 2020								0.13	0.21
T 2020									-0.01

Similar information emerges also from the Principal Components Analysis (PCA) and Factor Analysis (FA). Fig. 4 reports the first graph relating Component 1 and Component 2 of PCA for years 2016–2019 and the year 2020, respectively. These components together explain >60% of the variability.

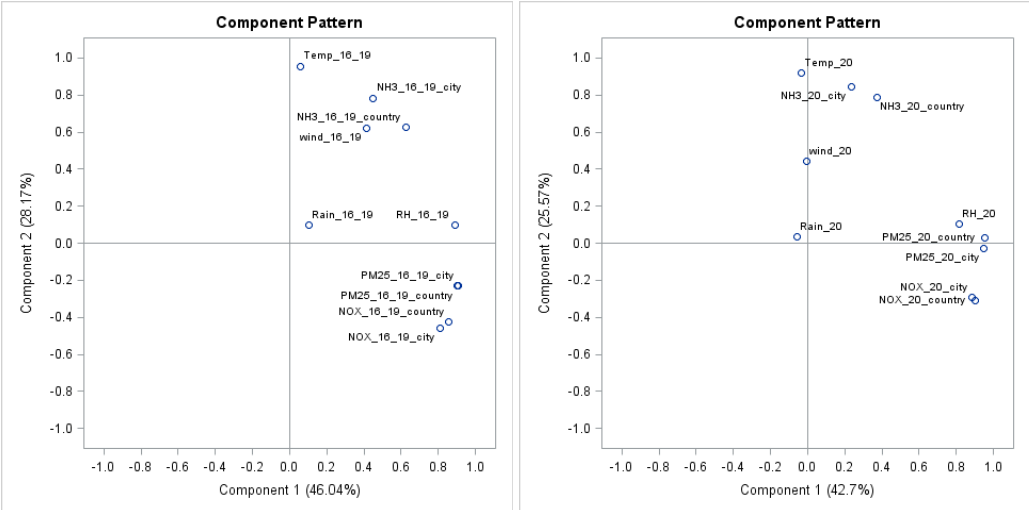

Figure 4. PCA, on the left for the period 2016-2019, on the right for 2020.

In more detail, PCA shows that every pollutant averaged for city and countryside stations is positioned close to each other. NOx and $PM_{2.5}$ are also close, while NH_3 is positioned in the upper quarter. For the 2016–2019 period, wind speed and temperature are quite close to NH_3 emission, while relative humidity and especially rainfall are quite isolated. In 2020, rainfall is isolated, relative humidity is closed to $PM_{2.5}$ emission, while temperature and wind speed are quite far but slightly closer to NH_3.

With FA, a clear distinction emerges between factors. In particular, 3 factors are identified that describe the components. Factor 1 can be entitled as "$PM_{2.5}$, NOx and RH", Factor 2 as "NH_3, wind and temperature" and Factor 3 as "rain" for the analysis on 2016–2019. In 2020, the factors are much similar, although a small change occurred between Factor 2 that was characterized as "NH_3 and temperature" and Factor 3 that was "wind and rain". The results of FA are reported in Table 4 and Table 5.

Table 4. Factor Analysis for the period 2016-2019.

Parameters	Factor1	Factor2	Factor3
NOx 2016-19 country	**0.86**	-0.42	-0.01
NOx 2016-19 city	**0.81**	-0.46	-0.04
NH_3 2016-19 country	0.63	**0.63**	-0.35
NH_3 2016-19 city	0.45	**0.78**	-0.26
$PM_{2.5}$ 2016-19 country	**0.90**	-0.23	-0.11
$PM_{2.5}$ 2016-19 city	**0.91**	-0.23	-0.08
Wind 2016-19	0.42	**0.62**	0.38
RH 2016-19	**0.89**	0.10	0.33
T 2016-19	0.06	**0.96**	0.00
Rain 2016-19	0.10	0.10	**0.91**

Table 5. Factor Analysis for the period 2020.

Parameters	Factor1	Factor2	Factor3
NOx 2020 country	**0.90**	-0.31	-0.01
NOx 2020 city	**0.89**	-0.29	-0.02
NH_3 2020 country	0.37	**0.78**	-0.31
NH_3 2020 city	0.24	**0.84**	-0.32
$PM_{2.5}$ 2020 country	**0.95**	0.03	-0.03
$PM_{2.5}$ 2020 city	**0.95**	-0.03	-0.03
Wind 2020	-0.01	0.44	**0.69**
RH 2020	**0.81**	0.10	0.40
T 2020	-0.04	**0.92**	0.15
Rain 2020	-0.06	0.03	**0.68**

Finally, a General Linear Model (GLM) was carried out with SAS software, from which emerged interesting results. In particular, the model resulted significant, with $r^2 = 0.83$. Table 6 reports the estimates of GLM for NH_3 in 2020 in the city station, from which can be highlighted the effect of NH_3 emitted in the country and of wind and temperature.

Table 6. General Linear Model results.

Parameter	Estimate	S.E.	t Value	Pr > |t|
Intercept	-0.431	4.823	-0.090	0.929
NOx 2020 country	0.087	0.050	1.760	0.085
NOx 2020 city	-0.096	0.035	-2.730	0.009
NH_3 2020 country	0.249	0.028	9.050	<.0001
$PM_{2.5}$ 2020 country	0.028	0.165	0.170	0.864
$PM_{2.5}$ 2020 city	-0.020	0.141	-0.140	0.889
Wind 2020	-2.371	0.954	-2.490	0.016
RH 2020	0.045	0.043	1.040	0.301
T 2020	0.417	0.271	1.540	0.130
Rain 2020	0.156	0.311	0.500	0.618

The interesting aspect that can be gathered from this study is that among the pollutants, NH_3 is mostly evident in the countryside stations, therefore confirming what reported by EEA (2018) and Pozzer et al. (2017). Moreover, NOx and $PM_{2.5}$ are well correlated, but NH_3 highlights a specific trend, only partially correlated with $PM_{2.5}$. Since NH_3 also contributes with NOx and SOx to $PM_{2.5}$ formation, if NH_3 reduced from the agricultural activities, $PM_{2.5}$ would reduce even more, as confirmed also by Zhao et al. (2017). This would involve additional positive benefits on the environment, ecosystems, and human health. As an example, Zambrano-Monserrate et al. (2020) highlighted as positive effects of Covid-19 quarantine an improvement of air quality associated with $PM_{2.5}$ and NO_2 emissions reduction, improved appearance of beaches and a reduction of the environmental noise level. Moreover, also in the study of Pozzer et al. (2017), it is reported that reducing by 50% the agricultural emissions of NH_3, a reduction of $PM_{2.5}$ equal to 2.4 μg/m^3 could be obtained in the Po valley region. In this way also global mortality and respiratory diseases due to $PM_{2.5}$ could be reduced. This reduction value is important considering that Po valley basin is among the European areas at greatest risk of exceeding the threshold limits of air quality due to its geographic conformation and to the high level of industrialization and anthropization. Regarding the reduction of NOx and partial reduction of $PM_{2.5}$, this effect is more evident in the cities than in the agricultural areas; therefore, their reduction can be partially attributed to the reduction in traffic and interruption of many industrial and energetic activities. As reported by Marongiu et al. (2020), during March 2020 NOx deriving from transport reduced by 60%, and NOx from energy production and industries decreased by 4% and 13%, respectively. Also, Buganza et al. (2020) and Chauhan and Singh (2020) observed a reduction in $PM_{2.5}$ concentrations in the period characterized by the Covid-19 emergency. Improving these aspects of air quality is very important, as reported also by a preliminary study conducted by ARPA and Lombardy region (Buganza et al., 2020). Considering all these emissions, it is important to note that the current strong reduction in traffic and industrial activities has helped reduce $PM_{2.5}$ (Wu et al., 2016), therefore a combined reduction of all air pollutants should be promoted.

Being the agricultural sector responsible for the widest part of NH_3 emissions, this sector should adopt measures for its reduction, providing interventions to improve the agricultural impact on the environment (Zhao et al., 2017). Policymakers and stakeholders should promote policies, incentives and disseminate knowledge to farmers about the need of abating NH_3 emissions with the already widely studied solutions: covering tanks for slurry and digestate storage instead of adopting open tanks (Bacenetti et al., 2016b; Finzi et al., 2019; Guarino et al., 2006), introducing treatment systems for slurry (anaerobic digestion, solid-liquid separation, nitro-denitro, air treatment, additives, etc.) (Dinuccio et al., 2011; Fangueiro et al., 2009; Finzi et al., 2020), removing frequently slurry and manure from the barn (Hoff et al., 2006) and spreading slurry on-field through injection techniques that permit to spread slurry into the soil through anchors and avoiding the superficial spreading with diverter plates (Hansen et al., 2003; Mattila and Joki-Tokola, 2003) that instead favor the conditions for NH_3 volatilization. These just mentioned are all strategies related to manure/slurry management, however, other strategies allow reducing pollutants inside livestock houses, such as biofilters, bioscrubbers (or biotrickling filters), dry filters, water scrubbers, and wet acid scrubbers (Dumont, 2018; Van der Heyden et al., 2015). All these air cleaning systems improve the air quality that animals and farmers breathe daily, with positive effects on animal welfare and thus on-farm performance and profitability, but also assuring a healthier environment for animals and workers. Finally, it could be useful to set a benchmark limit not only for $PM_{2.5}$ and nitrogen application, but also for NH_3, NOx, and SOx. To date, the only limit set by the National Emission Ceilings Directive, 2016/2284/EU regards the obligation in European countries to abate NH_3 emission by 6% by 2020 (Directive (EU) 2016/2284).

3.5 Conclusions

From this preliminary study about air quality variation during the Covid-19 quarantine period, some conclusions can be drawn, and some key aspects can be opened for discussion on the agricultural sector, but also on industrial activities, energy sector, and traffic. The achieved results allowed to confirm what was initially expected. Probably as an effect of the quarantine, some emissions caused by industries, energy production, and traffic deeply reduced (e.g., NOx and $PM_{2.5}$) at least in the cities areas considered, while some emissions caused by the agricultural activities did not change (e.g., NH_3) because no variations occurred for agricultural activities within the quarantine framework. However, further studies focused on agricultural emissions considering more data air quality stations are needed, also over a longer period of time. These could give the opportunity to better monitor the emission of NH_3 on the territory and introduce targeted interventions for its reduction. This study may be considered as a preliminary reference to future evaluations on agricultural emissions.

From some current discussions, it could be concluded that somehow air quality has slightly improved; but this last conclusion cannot be drawn at the time being and relatively to this study. This research aimed to focus on agricultural activities, therefore data stations were selected based on this need. Moreover, air quality is affected by a big series of factors, among which other pollutants such as SOx, local weather conditions, regional air exchanges, traffic, energy-related industry and industrial activities that in this study were not evaluated. For what regards the responsibility of the agricultural sector to $PM_{2.5}$ emission, the need for abating NH_3 emissions is highlighted. Since agricultural NH_3 emissions derive mainly by livestock housing, manure storage and manure spreading, the already studied solutions of covering tanks, introducing additives, removing and treating slurry, using air cleaning systems in barns, and improving the spreading techniques should be promoted by policymakers and stakeholders. This last point can be carried out through the promotion of policies and incentives, and disseminating knowledge to farmers who are the final decision-makers for investing in the improvement of air quality. Moreover, much need to be done to comply with air quality regulations in order to not exceed PM limits and also implement more restrictive rules related to agricultural NH_3 emissions reduction. In any case, to improve the air quality, a combined role of all productive sectors is fundamental because pollutants, and in particular $PM_{2.5}$, derive from the co-presence of multiple pollutants in the air.

Supplementary materials

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envres.2020.110048.

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4. Comparison of ammonia air concentration before and during the spread of COVID-19 in Lombardy (Italy) using ground-based and satellite data

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Abstract

Several anthropogenic activities have undergone major changes following the spread of the COVID-19 pandemic, which in turn has had consequences on the environment. The effect on air pollution has been studied in detail in the literature, although some pollutants, such as ammonia (NH_3), have received comparatively less attention to date. Focusing on the case of Lombardy in Northern Italy, this study aimed to evaluate changes in NH_3 atmospheric concentration on a temporal scale (the years from 2013 to 2019 compared to 2020) and on a spatial scale (countryside, city, and mountain areas). For this purpose, ground-based (from public air quality control units scattered throughout the region) and satellite observations (from IASI sensors on board MetOp-A and MetOp-B) were collected and analyzed. For ground-based measurements, a marked spatial variability is observed between the different areas while, as regards the comparison between periods, statistically significant differences were observed only for the countryside areas (+31% in 2020 compared to previous years). The satellite data show similar patterns but do not present statistically significant differences neither between different areas, nor between the two periods. In general, there have been no reduction effects of atmospheric NH_3 as a consequence of COVID-19. This calls into question the role of the agricultural sector, which is known to be the largest responsible for NH_3 emissions. Even if the direct comparison between the two datasets shows little correlation, their contextual consideration allows making more robust considerations regarding air pollutants.

Keywords: Ammonia; Ground-based measurements; IASI; Po valley; Lombardy; COVID-19

4.1 Introduction

Ammonia (NH_3) is an air pollutant of increasing environmental concern, whose emissions are primarily anthropogenic, released mainly from the agricultural sector by field application of synthetic fertilizers and manure management (Van Damme et al., 2015). NH_3 causes a series of cascading negative effects that damage both ecosystem biodiversity due to acidification and nitrogen enrichment (EMEP Centre on Emission Inventories and Projections, 2020; Erisman et al., 2007; European Commission, 2005) and human health (Van Damme et al., 2014), being a precursor of secondary fine particulate matter ($PM_{2.5}$) (Lovarelli et al., 2020; Perone, 2021). The damages to public health and ecosystems have been evaluated in 10–25 €/kg NH_3

(Executive Body of the Convention on Long-Range Transboundary Air Pollution, 2019). For these reasons, in recent decades, international integrated policies have been increasingly interested in quantifying, monitoring, and limiting NH_3 emissions. Despite the reduction obtained in absolute terms since 1990, in 2018, agriculture accounted for 93% of NH_3 emissions in relative terms in the European Union, still showing the criticality of this sector (EEA, 2019).

The reduction of NH_3 emissions is a complex process. First of all, measuring this compound is not easy, which also makes accurate monitoring difficult. The most widely used methodologies to date are measurements through ground-based instruments and satellite-based remote-sensing (Nair and Yu, 2020). However, each of these two techniques has advantages and problems. The ground-based measurements currently allow understanding the evolution of the atmospheric concentrations over time at ground level, but they are affected by local spatial and meteorological variability. Indeed, NH_3 volatilization from main primary sources (fertilizers and manure) is influenced by weather parameters such as wind speed, rainfall, and temperature, among others (Brentrup et al., 2000). Given the high variability related to NH_3 emissions, a geographically dense network of control units can be a useful method to fulfill monitoring requirements. However, due to the stickiness of NH_3 to observational instruments, the control units that collect NH_3 data are normally fewer per unit area than those used to measure other air pollutants (Van Damme et al., 2015). Satellite remote sensing, on the other hand, is based on the distinction of the NH_3 spectrum in the gas-phase by means of infrared spectrometers and allows to obtain a broad spatial coverage (both superficial and vertical) of NH_3 atmospheric concentrations, but the measurements available to date suffer from temporal discontinuity because deployed instrumentation is not onboard geostationary satellites (Nair and Yu, 2020). Furthermore, the reliability of the measurements at night or in the presence of clouds decreases. Ultimately, since the two methods compensate, at least partially, the respective limitations, considerations on atmospheric NH_3 pollution made by integrating both types of measurement can be more solid and comprehensive.

During the worldwide spread of Coronavirus Disease (2019) (COVID-19) in 2020, attention on air quality and the need to understand changes in the presence of pollutants in the atmosphere increased considerably. In literature, several studies have focused on the relationships among the COVID-19 outbreak, government actions to contain the spread of the infection and air pollution (Nuñez-Delgado et al., 2021; Zambrano-Monserrate et al., 2020). Among the European countries, Italy was the first in which the infection was detected in 2020, and which suffered a rapid spread of the infection in the first months of the year; this led the government to implement partial restrictions (e.g., establishment of a “red zone” in some municipalities in Northern Italy on 23 February, and subsequent interruption of school and university teaching in attendance). Then, heavy restrictions were introduced starting on 8 March, until on 23 March a nationwide lockdown was declared. Only industries deemed essential, such as food and pharmaceutical supply chains, and the agrifood sector were allowed to remain operational. This lasted officially until 3 May (DPCM, 2020).

As concerns the Po Valley in Northern Italy, where air pollution is recognized as being normally high (Raffaelli et al., 2020), the lockdown period has led to significant reductions in atmospheric concentrations of pollutants such as $PM_{2.5}$, PM_{10}, nitrogen oxides (NOx) and others (e.g., carbon monoxide and benzene) (Buganza et al., 2020; Collivignarelli et al., 2020; Deserti et al., 2020). On the other hand, the effect of the pandemic on NH_3 concentration has received comparatively less attention (Gualtieri et al., 2020; Lovarelli et al., 2020), although Northern Italy, and in particular Lombardy, is one of the leading regions for agriculture. Here, livestock production accounts for around 52% of pigs (ISMEA, 2019a), 20% of meat and dairy cattle (ISMEA, 2019b, c), and 17% of poultry (ISMEA, 2020) of the whole Country. The lower interest towards NH_3, compared to PM concentrations could be explained by the identification of PM particles as possible vectors for transporting the SARS-CoV-2 virus and for their responsibility for respiratory and cardiovascular diseases (Li et al., 2018; Srivastava, 2021). However, NH_3 influence on secondary aerosol is significant as it is a recognized precursor (Perone, 2021). Moreover, Zheng et al. (2020) and Manigrasso et al. (2020) discussed the possibility that SARS-CoV-2 spread is favored by a mild alkaline pH of airborne particles, and thus related to ammonia-polluted environments such as the Po Valley. To support this hypothesis, in the world 28 hotspots were identified with an NH_3 column concentration above 0.5 mg/m^2, which were linked to either biomass burning and fires or (and especially) agricultural areas, in particular with agricultural valleys surrounded by mountains, such as the case of Po valley in Northern Italy. Perone (2021) also identified cities in Northern Italy as those with the highest mortality risk in the country.

The aim of this study is to analyze NH_3 air concentration in Lombardy, the region most affected by the pandemic (Altuwayjiri et al., 2021; Bonati et al., 2021; Perone, 2021), by using both data from control units at ground level and satellite observations to evaluate the temporal and spatial scale of NH_3 concentration before and during lockdown (i.e., the strict national lockdown occurred in Spring, 2020), highlighting relationships, similarities or differences between these two measurement solutions.

4.2 Methods

4.2.1 Data collection of ground-based observations

Ground-based observations were retrieved from the database of the Regional Environmental Protection Agency of Lombardy (ARPA Lombardia, 2020). Data from all the control units collecting NH_3 measurements in the Lombardy Region were considered; overall data from 12 control units were available. These latter measure NH_3 indirectly through special analyzers using the chemiluminescence technique, by which NH_3 is first oxidized to nitrogen oxide (NO) and its concentration in the air sample is measured alongside NO and NO_2 (nitrogen dioxide). In particular, hourly data on air NH_3 concentration (expressed as μg/m^3) collected by the control units in question were sourced.

Furthermore, from the same ARPA Lombardia control units and for the same period, the daily weather parameters of temperature (T; °C), relative humidity (RH; %), rainfall (R; mm) and wind speed (W; m/s) were downloaded to consider their effects on NH_3 air concentration. The main characteristics of the control units used to retrieve these data are shown in Table 1.

Table 1. List of the control units used. Their location is provided as well as the grouping in different zones.

Province	Station Name	Analyzer model	Longitude (E°)	Latitude (N°)	Altitude (m)	Zoned as
Cremona	Corte de Cortesi	API 201E	10.0062	45.2785	57	Country
Cremona	Fatebenefratelli	TEI 17i	10.0438	45.1425	43	City
Cremona	Gerre Borghi	TEI 17i	10.0692	45.1095	36	City
Lecco	Colico	TEI 17i	9.3847	46.1381	229	Mountain
Lecco	Moggio	TEI 17i	9.4975	45.9128	1194	Mountain
Lodi	Bertonico	API 201E	9.6663	45.2335	65	Country
Mantua	Schivenoglia	TEI 17i	11.0761	45.0169	12	Country
Milan	Pascal	API 201E	9.2355	45.4790	122	City
Pavia	Folperti	TEI 17i	9.1646	45.1947	77	City
Pavia	Sannazzaro	ENVEA AC32e	8.9042	45.1028	87	Country

All control units were distinguished in "city", "country" (short for countryside) or "mountain" stations, to investigate the effect of COVID-19 on NH_3 air concentration in the city, most densely populated and characterized by traffic jams and industrial activities, in the countryside, where agricultural and livestock activities are most concentrated, and in mountain areas. This grouping was based on the zoning of Lombardy for air quality monitoring according to the Italian legislative decree 155/10 under the directive 50/08/CE on ambient air quality and cleaner air for Europe (ARPA Lombardia, 2020). According to the previous regulations, the stations located in *urban agglomerations* or *highly urbanized plains* have been considered as "city" stations, those located in the *plain area* as "country" and finally those located in the *pre-Alps*, *Apennines* and *mountains* as "mountain". The same grouping of stations was also adopted by Lonati and Cernuschi (2020). The zone shapefile was obtained from the Lombardy region geo-portal website (http://www.geoportale.regione.lombardia.it/) with the zone classification carried out at the municipality level. Fig. 1 shows the regional zoning for Lombardy and the geographical position of each control unit. To further investigate the land cover in each zone, the Corine Land Cover (CLC) classification (Feranec, 2016) from 2018 was reclassified to a binary layer with the classes agriculture/other. Agricultural land cover was 87% in the country class, 55% in the city class and 14% in the mountain class.

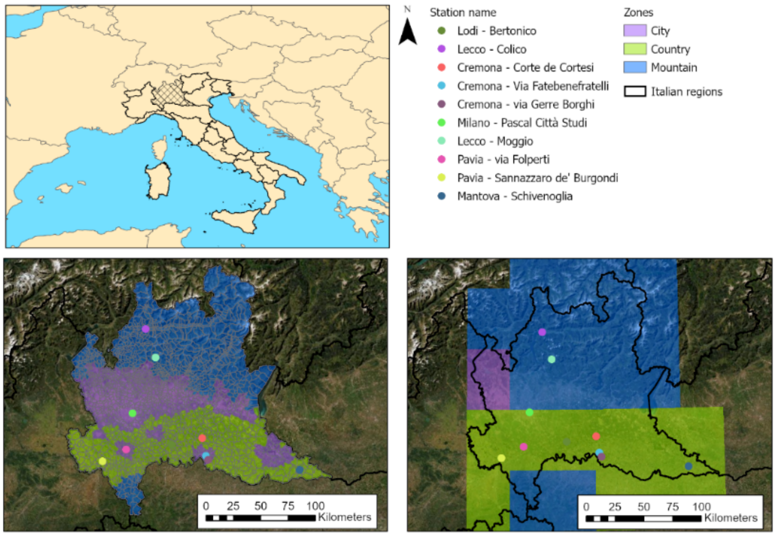

Fig 1. Zoning of the Lombardy region (Northern Italy) in city (violet colored), country (green colored), and mountain (blue colored) zones. Colored dots represent the position of every ground-based control unit. Bottom-left: original zoning; bottom-right: the zoning resampled to IASI pixels

The temporal coverage of the collected data goes from the beginning of 2013 to the end of October 2020. This period was selected because the same data were available also from the satellite dataset. Some data were lacking or were characterized by excessive uncertainty; therefore, the dataset was cleaned before data processing. With respect to groundbased control units, NH_3 concentrations not included in ±3 standard deviations were excluded from the dataset. Data were averaged as daily measures in order to be merged with the weather data for further analyses. All data were grouped in 2 periods to allow comparisons between normal living conditions and the period of the pandemic: the first included data from 2013 to 2019, while the second those of 2020.

Statistical analyses were conducted using SAS version 9.4 (SAS Institute, Cary, NC, USA) statistical software. Descriptive and multivariate statistics were carried out on each meteorological variable and on NH_3 concentration. A general Linear Model procedure (GLM Proc) was carried out to identify a model predicting the air concentration of NH_3 based on the unit zoning, local weather, monitoring period, and their interactions. In particular, in the model the following class parameters were included: (i) the period, with the 2 levels of 2013–2019 and 2020, and (ii) the zone, with the 3 levels of city, country, and mountain; the weather variables of temperature, rainfall and wind speed and the months of the year were included as well, together with their interactions for (i) year, zone and month, and (ii) temperature, rainfall and wind speed.

4.2.2 Data collection of remote sensing observations

Remote sensing data were retrieved from the online freely available database of the IASI sensor (https://iasi.aeris-data.fr/nh3/), which is the Infrared Atmospheric Sounding Interferometer onboard the ESA’s (European Space Agency) MetOp satellites. In particular, data observed by the MetOp-A and MetOp-B satellites were obtained for the period 2013–2020, as MetOp-B became operational in early 2013, and thus data from two satellites were available to obtain a higher number of observations. While a third satellite, MetOp-C has also been operational since 2019, IASI data from this platform were not used to avoid introducing a bias in the later period of the dataset. For both satellites, daily level 2 products were downloaded, which report total column NH_3 in molecules/cm^2 and their relative uncertainty (in percentage) for discrete points observed by the satellite with an approximate footprint of 12 km (at nadir). Only daytime observations were used, as these are considered more accurate owing to the larger thermal contrast compared to nighttime observations (Van Damme et al., 2014). As cloud cover and thermal contrast in the atmospheric column greatly reduce the number of available measurements, daily observations lying partially or entirely within the Lombardy region were spatially re-gridded to a 0.5° × 0.5° grid (for a total of 26 grid points) and temporally averaged to obtain monthly means. This grid size is similar to the choice adopted by Van Damme et al. (2014) and allowed us to obtain on average 100 measurements per grid cell per month, thus producing more statistically robust NH_3 monthly estimations. The averaging was weighted based on the uncertainty of each measurement, following the procedure described by Van Damme et al. (2014), i.e. (equation (1)).

$$\bar{x} = \frac{\sum w_i x_i}{\sum w_i} \qquad (1)$$

where x_i is a IASI measurement contained in the 0.5°x0.5° cell, w_i is the weighting factor, equal to $1/\sigma^2$ and σ is the error of the total column retrieval on a pixel basis.

The uncertainty of each monthly average was then expressed as

$$\bar{\sigma} = \frac{\sum \frac{1}{\sigma_i}}{\sum \frac{1}{\sigma_i^2}} \qquad (2)$$

Moreover, it was established to discard monthly means for which the uncertainty was higher than 75% and measurements were fewer than 10 (30% of a month), as per recommendations by Van Damme et al. (2014). Similar to the methodology adopted for the ground-based control units, satellite-based measurements were also divided into two periods, i.e., 2013–2019 and 2020, and classified as “city”, “country” or “mountain” based on the areas defined by ARPA Lombardia. In this case, the analysis was limited to the months from January to June owing to the unavailability of IASI data from June 2020 onwards at the time of writing. The zone shapefile was transformed to a raster and resampled to the same 0.5°×0.5° grid used for IASI observations; each IASI pixel was then classified by assigning to it the zone with the largest count within the pixel (see Fig. 1). In addition, the CLC 2018 was resampled to the IASI grid and the percentage of agricultural land use was counted for each pixel. In the country zone, agricultural land use ranged between 50% and 83% except for one pixel (21%). In the mountain zone, it ranged between 4% and 21% while in the city zone it was 13%.

Statistical analyses were also conducted using SAS version 9.4 (SAS Institute, Cary, NC, USA) statistical software, with a General Linear Model procedure (GLM Proc) similar to the ground-based dataset; also in this case, a model predicting NH_3 air concentration based on the units zoning, local weather, monitoring period and their interactions was carried out for satellite data. The statistical model used for satellite data was the same as for ground-based data, to allow the best comparability of results; therefore, the parameters included in the model were class parameters for the period (levels of 2013–2019 and 2020) and for the zone (3 levels of city, country, and mountain) and the weather variables of temperature, rainfall and wind speed, the months of the year and the interactions among year, zone and month, as well as temperature, rainfall and wind speed.

To complement satellite observations with meteorological data in a similar way as done for ground NH_3 measurements, daily temperature, rainfall and wind speed from the reanalysis model ERA5 (European center for meteorology and weather forecast reanalysis, (Hersbach et al., 2020)) at 0.25° × 0.25° resolution were obtained. The values were then regridded to the same 0.5° × 0.5° grid of IASI observations and averaged to monthly values to conduct the GLM procedure. The steps of the adopted methodology are summarized for both ground-based and remote sensing observations in Fig. 2.

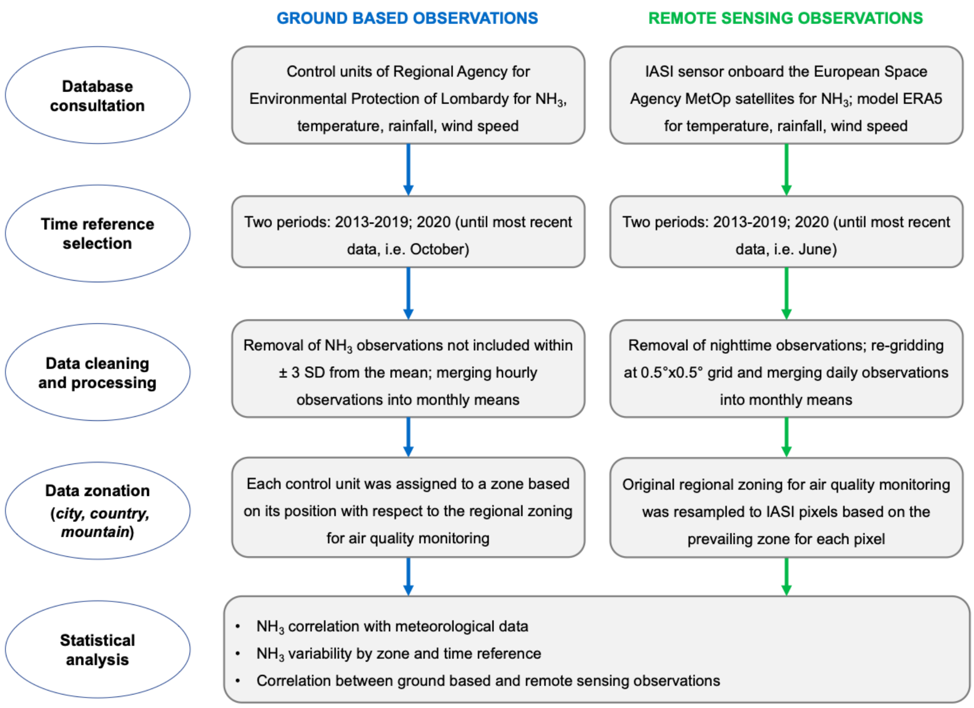

Fig. 2. Flow chart summarizing the main phases of the methodology adopted to organize and analyze the dataset.

4.3 Results

4.3.1 Ground-based measurement

For what concerns NH_3 air concentrations, Fig. 3 reports the average concentration (μg/m^3) for the control units according to the classification in the zones of city, country, and mountain, with the mean and standard error for each month of the two periods 2013–2019 and 2020.

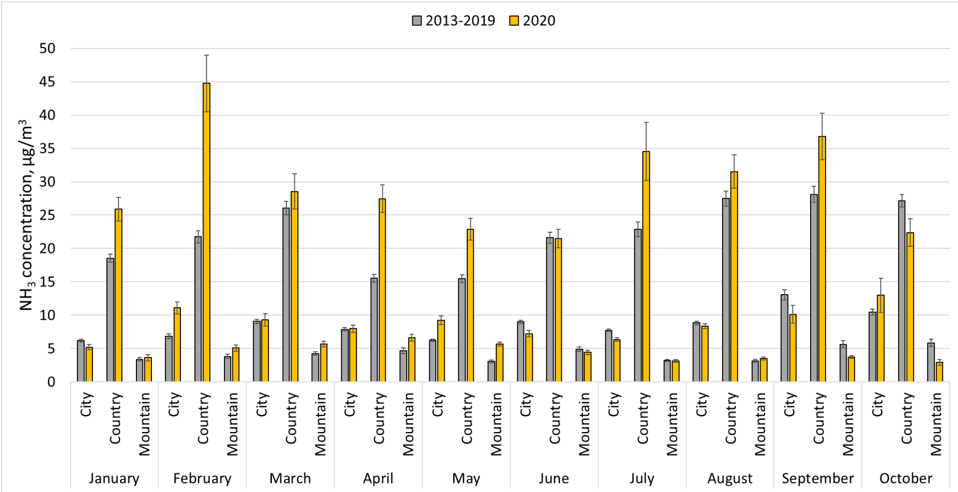

Fig. 3. Mean and standard error of NH_3 concentration, expressed in µg/m^3, per zone, month and period.

In both periods, the highest NH_3 concentrations can be observed for countryside stations. These stations are located in areas near agricultural production sites, whose seasonal practices affect mostly NH_3 emissions. Statistically significant differences can be found among the three zones, with country zones showing much higher concentrations than stations located in city and mountain zones. In particular, the mean yearly value for country zones was 22.5 and 29.5 µg/m^3 for 2013–2019 and 2020, respectively. The highest concentrations were recorded in August and September for 2013–2019 (27.5 and 28.1 µg/m^3, respectively) and in February and September for 2020 (44.7 and 36.8 µg/m^3, respectively). The lowest average value of NH_3 concentration in the country zone (21.5 µg/m^3, June 2020) was higher than the highest of city and mountain zones (13.0 and 6.6 µg/m^3, October and April 2020, respectively).

In all cases, mountain stations show the lowest concentrations, with 2020 higher than 2013–2019 in all months except for June, July, September, and October. The yearly average is equal to 4.2 and 4.4 µg/m^3 in 2013–2019 and 2020, respectively. The city zone has intermediate values, with a yearly average equal to 8.6 and 8.8 µg/m^3 for 2013–2019 and 2020, respectively. Although for half the months considered, the city zone NH_3 concentration was higher in 2020 than in the previous period (i.e., February, March, April, May, and October), the annual average is still slightly higher in 2020. Interestingly, the months in which NH_3 in the city was higher than in the previous periods were the same months in which the strictest lockdown was in progress (i.e., spring 2020). A similar observation can be done with respect to the mountain zone.

Both country and city zones showed higher NH_3 concentration in 2020 than in the previous period in February, March, April, and May. Such higher concentration, especially the peak observed in February, can be attributed to manure management and manure field spreading. In particular, because field application of manure can be carried out depending on the constraints fixed by crops cultivation (sowing periods), by laws (European nitrates directive 91/676/EEC and the regional action programs for the protection of water pollution, Nitrates Directive (Eu), 2016), as well as by weather conditions (field spreading can be carried out when field conditions permit it, such as when no rainfall occurs), quite strict temporal windows can be identified during the year. Therefore, slurry application, and the related NH_3 air concentration were higher at the beginning of 2020 than in previous years probably because the unfavorable weather conditions of the previous autumn (i.e., in October 2019 high rainfall was observed at country zone stations with average rainfall of 115 mm, equal to 15% of the year) did not favor slurry spreading. Hence, a massive field application of slurry occurred at the beginning of 2020, which led to the consequent observation of high NH_3 concentrations. Moreover, during the months of lockdown, transport and industrial activities mostly stopped, and consequently also the related emissions decreased. The reduction of other air pollutants, such as sulfuric acid and nitric acid, with which NH_3 combines to form secondary PM (Ge et al., 2020), may have contributed to binding less NH_3 and a higher chance to find it in its free form.

The main zone differences mentioned above are reported in Fig. 4. Here, NH_3 air concentrations are reported split into 2 periods and 3 zones. In 2020, NH_3 concentration recorded at country stations were 69% of the total and were 31% higher than in 2013–2019. No significant differences between NH_3 values in 2013–2019 and 2020 can be observed for the city zone (+2%). In the mountain zone, instead, NH_3 concentration participates for 10% of the total, with +6% in 2020 compared to 2013–2019. The three zones of city, country, and mountain show statistically significant differences.

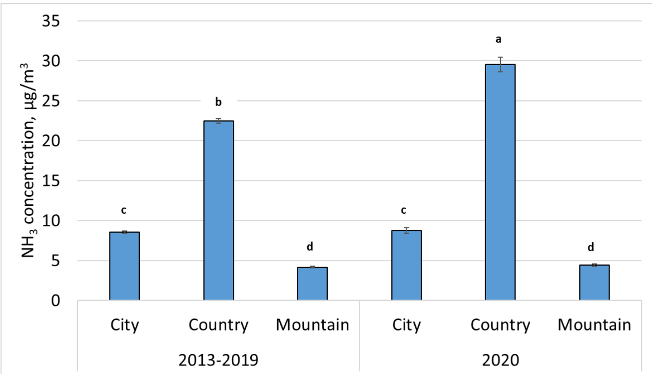

Fig. 4. Mean and standard error of NH_3 concentration, expressed in µg/m^3, per zone and period.

4.3.1.1 Meteorological observations in relation to NH_3 air concentration

Fig. 5 reports the average meteorological data of the analyzed provinces, with control units grouped by zone in the two selected periods for temperature (Fig. 5-top), rainfall (Fig. 5-middle), and wind speed (Fig. 5-bottom). Considering that Po Valley is located in Northern Italy and that it is mostly characterized by a temperate climate, without a dry season and with a hot summer (Beck et al., 2018), the meteorological data show a pattern consistent with these characteristics. In particular, regarding temperature a cold winter and a warm summer can be identified; rainfall is concentrated in spring and autumn and wind speed is low on average throughout the year.

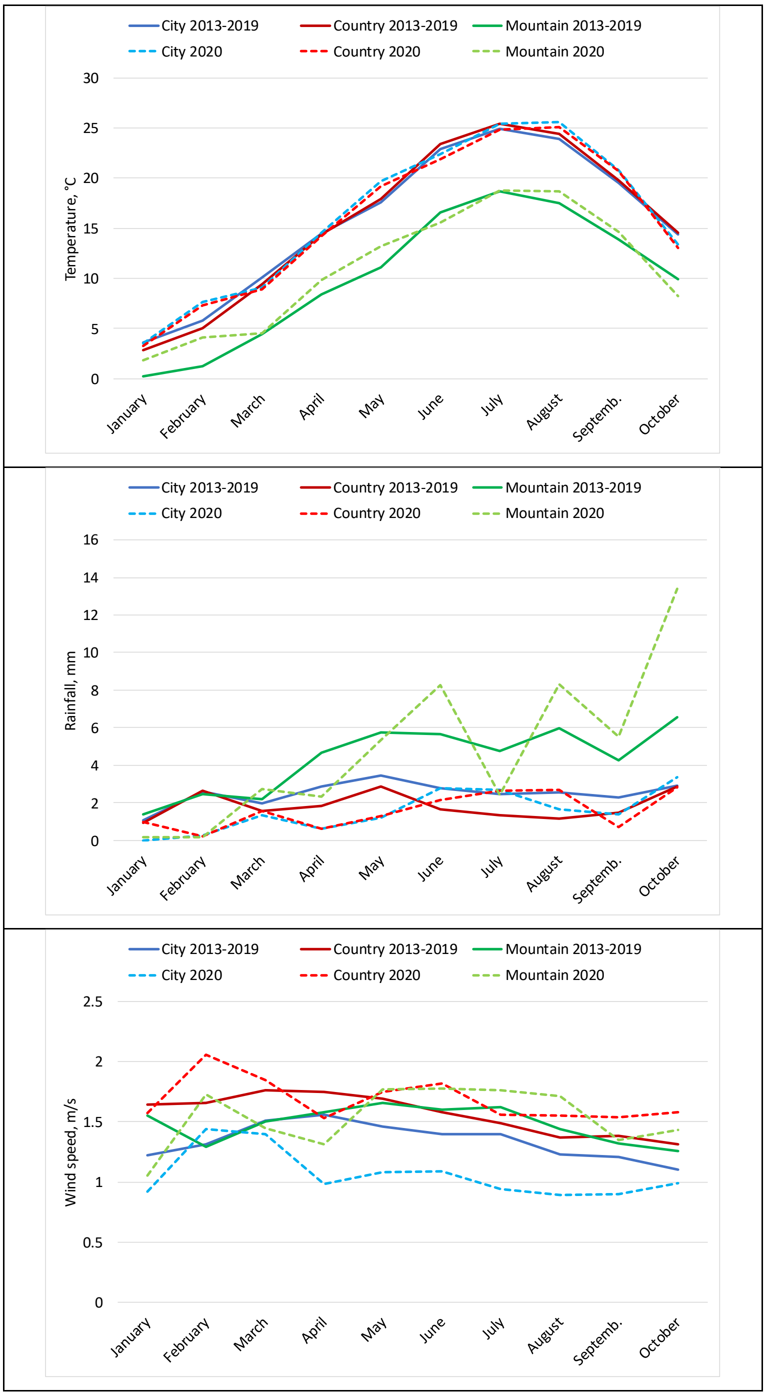

Fig. 5. Mean meteorological data (temperature -top, rainfall – middle, and wind speed -bottom) per zone (city, country, and mountain), month (Jan–Oct) and period (2013–2019, 2020).

In general, it can be observed that both city and country station units recorded similar average yearly values for temperature, with 2020 showing slightly higher values (15.7 and 16.2°C in 2013–2019 and 2020 in the city zone and 15.7 and 15.8°C in the country zone, respectively). In the mountain zone they were lower (10.2 and 10.9°C in 2013–2019 and 2020, respectively). The average rainfall was lower in 2020 compared to the previous period in the city (2.5 and 1.9 mm/d in 2013–2019 and 2020, respectively) and country (1.8 and 1.6

mm/d) zones, while in the mountain zone rainfall was higher (4.4 and 4.9 mm/d). As regards wind speed, all values were low: 1.3 and 1.1 m/s as average of 2013–2019 and 2020 in the city zone, country averaging 1.6 and 1.7 m/s and mountain averaging 1.5 m/s in both periods. Pearson correlations were calculated (data not shown) and significant differences were found among all the meteorological parameters considering the different zones and months. Although the linear relationship is not very strong, temperature is significantly negatively correlated with rainfall and wind speed. In contrast, rainfall and wind speed are positively correlated with each other. The changes in NH_3 air concentration and meteorological parameters during the period of observation are reported in Table 2.

Table 2. Percentage change between 2020 and 2013–2019 for NH_3 air concentration, temperature (T), rainfall (R) and wind speed (W). When the resulting changes are positive (year 2020 > years 2013–2019) cells have a green background.

Month	Zone	NH_3	T	R	W
January	City	-15.4%	-3.3%	-34.0%	-24.6%
	Country	39.6%	15.5%	1.0%	-4.3%
	Mountain	6.5%	682.6%	-87.7%	-32.3%
February	City	62.4%	32.6%	-90.0%	9.9%
	Country	105.6%	44.4%	-91.3%	24.1%
	Mountain	32.8%	231.2%	-92.7%	34.1%
March	City	2.8%	-10.3%	-31.0%	-7.3%
	Country	9.5%	-5.7%	1.3%	5.1%
	Mountain	33.2%	1.6%	26.0%	-3.3%
April	City	1.1%	0.5%	-78.7%	-37.2%
	Country	76.7%	-1.0%	-64.9%	-12.6%
	Mountain	41.5%	16.1%	-50.2%	-17.1%
May	City	48.2%	11.9%	-64.9%	-26.0%
	Country	48.2%	6.9%	-54.3%	3.6%
	Mountain	83.8%	19.1%	-7.3%	6.6%
June	City	-19.7%	-2.3%	1.1%	-22.1%
	Country	-0.6%	-6.6%	28.1%	15.2%
	Mountain	-9.8%	-6.1%	47.0%	11.3%
July	City	-17.5%	2.2%	9.7%	-32.9%
	Country	51.4%	-2.2%	94.8%	4.7%
	Mountain	-1.9%	0.8%	-49.2%	8.6%
August	City	-5.2%	7.1%	-34.6%	-27.6%
	Country	14.7%	2.5%	133.9%	13.1%
	Mountain	11.9%	6.8%	39.0%	18.8%
September	City	-22.3%	6.3%	-40.7%	-25.6%
	Country	31.0%	4.5%	-52.7%	11.6%
	Mountain	-33.7%	5.5%	28.7%	2.3%
October	City	24.2%	-7.3%	15.3%	-10.0%
	Country	-17.6%	-10.3%	-1.4%	20.6%
	Mountain	-49.7%	-16.8%	104.1%	13.5%

The relative difference between the periods 2020 and 2013–2019 is carried out by using the data reported in Figs. 3 and 5. From these data, it emerges that NH_3 concentration in air was higher in 2020 than in the previous period from February to May for all zones and this occurred, in most cases, together with higher temperatures. Except for these four months and October, in city zones NH_3 was lower in 2020 than over the previous years, which occurred with changes in the meteorological parameters that, however, do not allow to identify clear links between NH_3 and weather patterns, and especially with rainfall/wind speed.

To clarify the relationships among these variables, a GLM procedure was performed to obtain a predictive model for NH_3 concentration in air. The model was generated on the classes of year (2 levels: 2013–2019 and 2020) and zone (3 levels: city, country, and mountain) and was found statistically significant, although with a very low coefficient of determination ($R^2 = 0.19$). All variables were also significant with "year" and "zone" very significant ($p < 0.0001$) and month, weather parameters and their interactions significant ($p < 0.05$). Among the meteorological parameters, rainfall was the variable with the lowest significance for the prediction of NH_3. The calculated Least Squares Means (LS Means) were all significant ($p < 0.0001$). However, the statistical significance of LS Means for the effect year*zone showed no difference between the 2 periods for the city and the mountain zones as reported in Table 3.

Table 3. Statistical significance of Least Squares Means resulting from the GLM procedure for the effect year*zone and NH_3 as dependent variable.

i/j	2013-2019 country	2013-2019 mountain	2020 city	2020 country	2020 mountain
2013-2019 city	***	***	n.s.	***	***
2013-2019 country		***	***	***	***
2013-2019 mountain			***	***	n.s.
2020 city				***	***
2020 country					***

This result shows that, in the two studied periods, both city zones and mountain zones have comparable results. Since NH_3 is mostly released in countryside areas and normally deposited within a short radius from the emissive source, it can be expected that lower concentrations reach cities and mountain zones and that these latter do not show significant differences in the analyzed periods. Since agricultural activities are more subject than others to annual variability and seasonality, the fact that NH_3 in countryside areas is significantly different between 2013 and 2019 and 2020 prompted us to investigate the effects of seasonality on NH_3 every year. Thus, the model was relaunched evaluating the effect of each year on NH_3, focusing only on the country zone. This model was found significant with the coefficient of determination $R^2 = 0.29$ and all weather parameters significant ($p < 0.05$). Significant differences for NH_3 concentration emerged in some years, while no differences were found between (i) 2013 and 2016, (ii) 2014 and 2015, and (iii) 2017, 2018 and 2020. The results of this second analysis suggest that meteorological variability can play a role on NH_3 air concentration in country zones.

4.3.2 Satellite measurements

The NH_3 total column observations from IASI data reported in Fig. 6 show slightly different patterns compared to ground measurements, although it is a shared feature that country zones show the highest values in all months during 2013–2019 and in 2020. The highest total column NH_3 was recorded for grid points classified as "country" in June, both over 2013–2019 ($5.89e^{-4}$ mol/cm^2) and in 2020 ($5.31e^{-4}$ mol/cm^2); over 2013–2019, May was the second highest month with respect to NH_3 values ($4.59e^{-4}$ mol/cm^2), while in 2020 it was March, when the lockdown started ($5.22e^{-4}$ mol/cm^2). In March 2020, mountain grid points also show very high values ($5.11e^{-4}$ mol/cm^2), possibly because of the actual inclusion of agricultural areas within grid points classified as mountain at the IASI scale (maximum agricultural cover was 21% in mountain grid points from CLC, 2018). As a further hint of this, compared to ground observations, city zones show generally lower total column NH_3 than mountain zones, with the lowest values overall recorded for city zones in January 2020 ($1.34e^{-4}$ mol/cm^2). For country zones, all months except June show an increase in total column NH_3 compared to the average of 2013–2019. For the other zones, the pattern is more variable: city grid points show a higher NH_3 total column in February, March, April, and June 2020 while for mountain stations the months with a higher NH_3 total column are January, March (with an increase of 108% compared to the same month over 2013–2019), April and June.

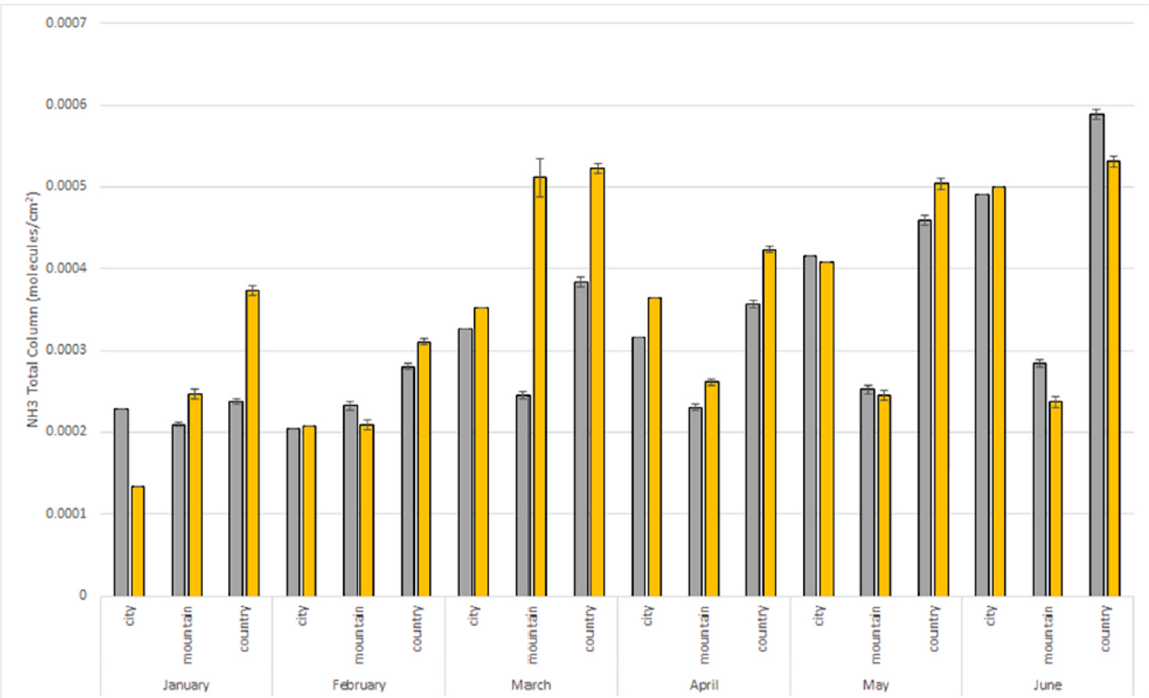

Fig. 6. Mean and standard error of total column NH_3 from IASI observations for each month (January–June) over the period 2013–2019 and 2020 for each of the three zones considered (city, mountain, country).

As shown in Fig. 7, averaging all monthly observations between January and June, mountain and country zones show an increase in NH_3 total column in 2020 compared to 2013–2019, with similar values (18% for mountain zones and 15% for country zones).

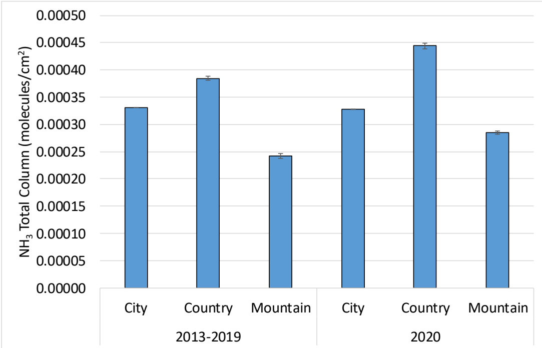

Fig. 7. Mean and standard error of NH_3 total column for January–June in 2013–2019 and 2020 for the three zones considered (city, mountain, country).

As for ground observations, the NH_3 total column for city zones is unvaried between the two periods (-0.73%). In contrast with ground observations, however, the share of NH_3 for the different zones is more uniform: in 2020, country zones accounted for 42% of the total column, mountain zones for 27% and city zones for 31%. This might be caused by the coarse spatial grid of IASI measurements and mix between different land use classes at the IASI scale.

4.3.2.1 Meteorological observations in relation to NH_3 air concentration

As for ground observations, monthly meteorological data and NH_3 total column divided in the two periods were compared. Table 4 reports such results.

Table 4. Percentage change between 2020 and 2013–2019 for NH_3 total column, temperature (T), rainfall (R) and wind speed (W) from IASI and ERA-5 observations. When the resulting changes are positive (year 2020 > years 2013–2019) cells have a green background.

Month	**Zone**	**NH_3**	**T**	**R**	**W**
January	City	-41.30%	12.53%	-81.74%	-15.22%
	Country	57.09%	8.67%	-66.28%	-5.94%
	Mountain	17.62%	50.67%	-69.93%	-2.42%
February	City	1.53%	50.45%	-87.94%	-1.07%
	Country	10.67%	48.31%	-90.94%	48.47%
	Mountain	-9.82%	449.99%	-66.97%	32.27%
March	City	7.72%	-5.11%	-4.30%	4.73%
	Country	36.02%	-4.51%	-22.97%	46.01%
	Mountain	108.41%	-5.71%	14.73%	-9.26%
April	City	15.62%	3.15%	-57.28%	-47.02%
	Country	18.64%	-0.05%	-52.21%	-24.42%
	Mountain	13.32%	9.78%	-57.93%	28.69%
May	City	-1.68%	9.32%	-10.92%	24.81%
	Country	9.80%	9.55%	-36.25%	46.39%
	Mountain	-2.54%	16.17%	-12.84%	26.45%
June	City	1.84%	-7.49%	49.63%	-11.88%
	Country	-9.85%	-5.65%	47.79%	41.90%
	Mountain	-16.59%	9.58%	44.44%	32.56%

In most months, temperature was higher in 2020 than in 2013–2019, particularly in the mountain zones where it was always higher except for March, with an increase of 449.99% in February 2020 compared to 2013–2019. In contrast, precipitation was lower in most months, except for mountain zones in March and all zones in June, while wind speed showed a more varied pattern with a decrease in January 2020 and an increase in May 2020 in all zones.

A GLM procedure was carried out to identify a prediction model for NH_3 total column, in a similar way as done for ground-based measurements. The model was found significant with a coefficient of determination of 0.55. The main difference with the GLM for ground-based observations is related to the fact that none of the meteorological parameters was found statistically significant, and only the zone parameter and month*zone effect were significant in the prediction of NH_3. In addition, no statistical difference emerged from the evaluation of LS Means for year*zone, as reported in Table 5.

Table 5. Statistical significance of LS Means resulting from the GLM procedure for the effect year*zone and NH_3 as dependent variable.

i/j	2013-2019 country	2013-2019 mountain	2020 city	2020 country	2020 mountain
2013-2019 city	n.s.	n.s.	n.s.	n.s.	n.s.
2013-2019 country		n.s.	n.s.	n.s.	n.s.
2013-2019 mountain			n.s.	n.s.	n.s.
2020 city				n.s.	n.s.
2020 country					n.s.

The result of the GLM procedure with satellite NH_3 observations and meteorological parameters shows no statistical difference with respect to the NH_3 concentration in the air between the two periods. This is in contrast with the ground-based measurements, which showed a significant difference for NH_3 concentrations in country stations, between 2013 and 2019 and 2020. The reason for this result may be due to the susceptibility of control units to a series of site-specific aspects that, instead, cannot be measured in the satellite coarse grid, and the fact that satellites measure total column NH_3, which might be the result of emissions originating in a different grid point than the footprint observed by the satellite at the time of data acquisition.

4.4 Discussion

This study was carried out with a focus on comparing the period (i.e., from year 2013–2019) before the lockdown determined by the emergency of COVID-19 and the subsequent period during the pandemic (i.e., year 2020, with the strict lockdown lasting from March–June 2020), adopting two measurement systems. Remote sensing datasets show no significant difference in NH_3 air concentration between the period before the spread of COVID-19 and the one during the pandemic. From the ground based analysis, NH_3 was observed higher than other years in city and mountain, and significantly in country zones. As NH_3 is strongly dependent on agricultural and livestock activities, its concentration was found subject to seasonality, weather conditions and to agricultural management. This aspect has been underlined also in the study of Lonati and Cernuschi (2020) and Deserti et al. (2020). The same result was observed also by Gualtieri et al. (2020) who state that, in 2020, NH_3 concentration increased in Italy compared to 2019. They also report that more than 90% of NH_3 is of agricultural origin in Milan, while this contribution decreases to 71% in Rome and 62% in Bologna, suggesting once more the relevance of livestock activities in Northern Italy. Therefore, the role of agriculture and livestock appears to be the largest influence on NH_3 air concentrations. Given the relationship of weather and seasonality with agricultural field activities, and the characteristics of NH_3 emissions (Brentrup et al., 2000; Sutton et al., 2013), it can be expected that NH_3 concentration is higher in summer (high temperatures) than in winter (when regulations prohibit field spreading due to nitrate leaching), and that peaks are observed in periods before crop cultivation (generally from February to June and from August to October in the analyzed area, where double cropping is widespread) when base-dressing fertilizers are applied on the field (Guido et al., 2020; Pedersen et al., 2020). All these aspects are confirmed also by the present study. Independently from the period considered, the highest NH_3 concentration can be observed in February–March and July–October, compared to the other months.

Moreover, different values in NH_3 concentration are strictly linked to manure storage (Zilio et al., 2020) and to field application, since this operation can be carried out when no rainfall occurs and in agreement with the European nitrates directive 91/676/EEC and the regional action programs for the protection of water pollution caused by nitrates from agricultural sources. These aspects have been widely investigated in literature, such as by Skjøth et al. (2011), Ramanantenasoa et al. (2018), and Ge et al. (2020).

4.4.1 Limitations of ground-based and satellite observations

In this study, measurements from both ground control units and satellite sensors were used to investigate air concentrations of NH_3. The two techniques showed similar patterns, such as the higher yearly average concentrations in country zones in 2020 compared to 2013–2019 but also some differences for individual months and in the relative contribution of the different zones to total NH_3 concentrations, which are related to the limitations inherent in measurement techniques.

For the entire Lombardy region (surface area about 23000 km^2), the availability of NH_3 measured data for the analyzed period was limited to 10 control units for the assessment using ground stations. However, NH_3 concentration may vary considerably within few kilometers (Lonati and Cernuschi, 2020). This confirms the scarcity of NH_3 control units compared to other air pollutants; in fact, in the same area available stations amount to 90 units for PM_{10} and 38 units for $PM_{2.5}$ (ARPA Lombardia, 2020). Moreover, for the same control units, air concentrations of different pollutants, such as NH_3 and PM, are not always available, making comparisons more difficult.

As concerns satellite images, the number of measurements, after filtering for observations with high uncertainty, did not allow us to create a grid finer than 0.5 × 0.5°, which would have introduced a large number of data gaps. The coarse grid however complicates the assessment of local variability; the comparison between data gathered from each ground-based control unit and satellite observations is problematic because of 1) the different spatial resolution; in fact, ground stations classified as “city” might be included in country zones in IASI pixels, with a much larger contribution of NH_3 air concentration from agricultural areas; 2) the different unit of measure and assessment method, as one is a ground level observation, the other is a column observation. An attempt to correlate monthly data from ground and satellite observations showed an r^2 of 0.21–0.29; this is in line with Van Damme et al. (2015), who report Pearson’s correlations of 0.28 comparing monthly data from IASI and ground observations from a global network, ranging from 0.81 (Russian Fyodorovskoye site, with very high NH_3 concentrations owing to fire events) to negative correlations at several sites in Finland, probably caused by the low concentrations at these sites in association with low temperatures and thus the low thermal contrast of IASI.

Given the limitations of both measurement approaches, among which the temporal and spatial resolution, improvements should be introduced both in the number and density of ground stations, especially in areas where agriculture is widespread, and in the availability of satellite data with a higher spatial resolution, which might also make validation efforts easier in comparison with ground observations.

4.4.2 Prospects for improving air quality and opportunities for further study

In the future, a significant reduction in NH_3 air concentrations is expected as a consequence of the abatement measures that are being introduced in agricultural and livestock farms (e.g., closed tank storages, manure and slurry treatments, precision application of slurry on field) (Finzi et al., 2019; Zilio et al., 2020), although these measures are currently present only in few contexts. Considerable improvements have already been introduced in the European Union, which brought to a strong reduction of NH_3 air concentration, reaching -24% from 1990 to 2017 (Costantini et al., 2020). However, this trend of reduction is proceeding further; in fact, in some countries, regions, and farms additional improvements are being introduced or are under study. For example, Miranda et al. (2021) investigated through the Life Cycle Assessment method, the environmental implications of treating slurry and found positive results for the reduction of gases, among which NH_3, with the addition of sulfuric acid to slurry. Slurry acidification was found effective in reducing NH_3 and other greenhouse gases (GHGs) also by Fangueiro et al. (2015). However, supporting farmers in the direction of abating NH_3 is fundamental to increase the spread of such ameliorative techniques and solutions. The chief aspects on which to focus include efficient livestock rearing techniques supported by technology, balanced animal feed rations, air scrubbers in barns, adequate manure management with frequent removal from the barn, manure treatments such as anaerobic digestion, solid-liquid separation, slurry acidification, proper manure storage with closed tanks and proper field application with precision application equipment (Guido et al., 2020; Regueiro et al., 2016).

Importantly from the environmental point of view, avoiding NH_3 losses can lead to maintaining nutrients in the manure/slurry and therefore applying more nutrients to the soil when spreading manure or slurry, thus requiring fewer mineral fertilizers and bringing benefits to the environmental sustainability, plus valorizing an already available resource that is free of charge. Finally, while high NH_3 concentration in air may represent a local issue, damages to ecosystems from NH_3 are not only local and are widely investigated due to the main effects of acidification and eutrophication on biodiversity loss in coastal and estuarial areas, such as in the study by Vetterli et al. (2016).

Despite the improvements in agricultural practices, this study has shown that NH_3 concentrations in air remained high in areas such as the Po Valley even during the lockdown caused by the spread of COVID-19. To further validate the considerations made regarding NH_3 air concentrations during 2020, future research should consider a wider area (e.g. Italy, Europe), as well as expand the analysis to other air pollutants. To this end, it would also be interesting to integrate ground-based and satellite datasets with physico-chemical dispersion models (e.g. LOTOSEUROS, Schaap et al., 2008; also employed by Van Damme et al., 2014, 2015) to better understand their origin and interactions, while in this study a purely statistical comparative approach was adopted.

4.5 Conclusions

This study aimed to evaluate the atmospheric concentrations of NH_3 during the spread of the COVID-19 pandemic in Northern Italy and the related changes in anthropogenic activities. For this purpose, ground-based and satellite measurement data relating to 2020 were analyzed and compared with previous years (2013–2019). Ground-based measurements showed statistically significant differences between the two periods in the country areas, where NH_3 was found higher in 2020 (+31% compared to the 2013–2019 average); on the other hand, no significant differences emerged for the city and mountain areas. Satellite data show similar patterns, but no significant differences between the two periods and less spatial variability between city, country, and mountain areas, probably due to the coarse spatial grid of IASI measurements. Contrary to other air pollutants, it can be concluded that no NH_3 reduction effect has occurred as a consequence of the anti-COVID-19 measures, which can be explained by the non-interruption of agricultural activities, the main emissive source of this pollutant. The integration between datasets from different measurement sources allows having a broader understanding of atmospheric phenomena, since both methods alone have their limitations. These considerations offer insights into the physico-chemical modeling of this pollutant and the actions aimed at its mitigation.

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CHAPTER 4

Intensive livestock farming other being responsible for the vast majority of NH_3 emissions in the European Union, extensively contributes to odor emissions, producing complaints from people living nearby. This chapter consists of one published peer reviewed review (*Paper 3*) that summarizes: i) odor measurement techniques; ii) air dispersion models applied to assess odor annoyance; iii) sources of odor nuisance; iv) mitigation strategies against odor impact.

5. Measurements techniques and models to assess odor annoyance: A review

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Abstract

Odors have received increasing attention among atmospheric pollutants. Indeed, odor emissions are a common source of complaints, affecting the quality of life of humans and animals. The odor is a property of a mixture of different volatile chemical species (sulfur, nitrogen, and volatile organic compounds) capable of stimulating the olfaction sense sufficiently to trigger a sensation of odor. The impact of odors on the surrounding areas depends on different factors, such as the amount of odors emitted from the site, the distance from the site, weather conditions, topography, other than odors sensitivity and tolerance of the neighborhood. Due to the complexity of the odor issue, the aim of this review was to give an overview of: i) techniques (sensorial and analytical) that can be used to determine a quantitative and qualitative characterization; ii) air dispersion models applied for the evaluation of the spatial and temporal distribution of atmospheric pollutants in terms of concentration in air and/or deposition in the studied domain; iii) major sources of odor nuisance (waste and livestock); iv) mitigation actions against odor impact. Among sensorial techniques dynamic olfactometry, field inspection, and recording from residents were considered; whereas, for analytical methodologies: gas chromatography-mass spectrometry, identification of specific compounds, and electronic nose. Both kinds of techniques evaluate the odor concentration. Instead, to account for the effective impact of odors on the population, air dispersion models are used. They can provide estimates of odor levels in both current and future emission scenarios. Moreover, they can be useful to estimate the efficiency of mitigation strategies. Most of the odor control strategies involve measures oriented to prevent, control dispersion, minimize the nuisance or remove the odorants from emissions, such as adequate process design, buffer zones, odor covers, and treatment technologies.

Keywords: Odor, nuisance, odor measurement, air dispersion models, waste, livestock

5.1 Introduction

The growing interest of people towards the environment and the greater attention to the quality of life have led to defining odors as harmful atmospheric pollutants (Capelli et al. 2013b; Henshaw et al. 2006), since malodorous conditions are mostly associated with unhealthy air situations (Aatamila et al. 2011). Because of accelerated urbanization and the lack of suitable sites, urban areas are sometimes built directly within or close to existing waste treatment plants and farms (Peters et al. 2014). Nuisance due to odor generation by waste treatment plants (e.g. landfill and composting plants) (Blanco-Rodríguez et al. 2018; Rincón et al. 2019), and animal production operations is one of the major sources of complaints of people living near these facilities (Keck et al. 2018), and has triggered increased emphasis on controlling the impact of atmospheric pollutants on neighboring areas (Bibbiani and Russo 2012; Hayes et al. 2014). Unpleasant odors may cause a variety of emotional and undesirable reactions in people, ranging from annoyance to documented health effects, leading to a reduced quality of life (Blanes-Vidal 2015; Domingo and Nadal 2009; Palmiotto et al. 2014).

The odor is defined by ISO 5492:2008 as an organoleptic attribute perceptible by the olfactory organ (including nerves) on sniffing certain volatile substances (International Organisation for Standardization 2008). Thus, the odor can be defined as "perception of smell" or "a sensation resulting from the reception of stimulus by the olfactory sensory system". Whereas odorant is a substance which stimulates a human olfactory system so that an odor is perceived (Blanco-Rodríguez et al. 2018). The odor is given by the interaction of different volatile chemical species, including sulfur compounds (e.g. sulfides, mercaptans), nitrogen compounds (e.g. ammonia, amines) and volatile organic compounds (e.g. esters, acids, aldehydes, ketones, alcohols) (Barth et al. 1984). Odorous compounds include both organic odorants and inorganic molecules that contribute to odor level (Zhu et al. 2016). Volatile organic compounds (VOCs) are a large group of organic chemicals, formed by molecules with different functional groups, having different physical and chemical behaviors, but characterized by certain volatility (Komilis et al. 2004), such as volatile fatty acids, alcohols, aldehydes, amines, carbonates, esters, sulfides, disulfides, mercaptans, and heterocyclic nitrogen compounds (Fang et al. 2012). On the other hand, inorganic compounds (H_2S, NH_3, Cl_2) due to their low molecular weights can bind olfactory receptors and affect odor level (Heaney et al. 2011; Huang et al. 2014; Pagans et al. 2006).

Different approaches and techniques have been used for measuring odors, both physical and chemical measurements (Capelli et al. 2013b; Munoz et al. 2010), and for simulating their dispersion in the atmosphere to plan setback distances, aimed at maintaining adequate buffer zones between livestock units and residents (Guo et al. 2004; Jacobson et al. 2005; Jacobson et al. 2000; Nimmermark et al. 2005; Schauberger et al. 2012).

Odors have always represented a social problem, but recently public concerns about their potential impact on health and wellbeing have been raised. This has led public opinion to concern on air quality issues and to complain to local Authorities (Brancher et al. 2019), with waste treatment plants and livestock farms representing the major sources of complaints (Keck et al. 2018; Sironi et al. 2005).to local Authorities (Brancher et al. 2019).

Thus, the aim of this review was to present, through the analysis of the published literature, an overview of techniques and models for measuring odors and simulating their dispersion, other than of odor emissions from waste management plants and livestock farms.

With this review we wanted to answer the following questions:

- Which techniques can be adopted for measuring odors in the field or in the laboratory?
- How can odor annoyance be assessed?
- Where do the biggest odors come from?
- How to protect people from odor nuisance?

According to these questions, the review has been organized as follows: section 2 will be focused on odor measurements, section 3 on dispersion models, section 4 will be on the major sources of odor, waste and livestock, respectively, and, finally, section 5 will be about strategies to protect people from odor nuisance.

5.2 Technical approaches to odor measurement

Quantitative and qualitative characterization of odors can be carried out by direct or indirect methods. In 2018, the European Union published a BREF (Bat REFerence document) on emissions monitoring "JRC Reference Report on Monitoring of Emissions to Air and Water from IED Installations" (Brinkmann et al. 2018). Regarding odor emissions, the following approaches are mentioned: dynamic olfactometry, dispersion models, field inspection, electronic noses, and odor surveys. Overall, in order to quantify the effective odor discomfort, sensorial techniques or analytical methodologies, based on human examiners, and on instruments, respectively, can be used (Munoz et al. 2010). According to this criterion, in sensorial techniques are included: dynamic olfactometry (sub-section 2.1.1), field inspection (2.1.2), and recording from residents (2.1.3); whereas, in analytical methodologies are mentioned: gas chromatography-mass spectrometry (2.2.1), identification of specific compounds (2.2.2), and electronic nose (2.2.3).

5.2.1 Sensorial techniques

5.2.1.1 Dynamic olfactometry

Olfactometry analysis is a standardized methodology (CEN EN 13725) used for determining the concentration of odorous, combining an olfactometer with human panelists. The EN 13725:2003 standardized the procedures and methods of analysis, making the dynamic olfactometry a reliable and consolidated measurement method (EN 13725 2003).

The olfactometer submits the odor sample, diluted with neutral air at precise ratios, to a panel of human assessors (Munoz et al. 2010). The samples are presented to the panel at increasing concentrations until the panel members start perceiving an odor different from the neutral reference air. The result is the odor concentration (Cod), expressed in European odor units per cubic meter (ouE m^{-3}), which corresponds to the dilution factor necessary to reach the odor threshold, that is the minimum concentration perceived by 50% of the population (Blanco-Rodríguez et al. 2018). The odor concentration is calculated as a geometric mean of at least 12 odor detection threshold values of each member of the panel. The advantage of using a human nose is its higher sensitivity, even if it suffers from a lack of specificity for individual odorants (Barth et al.

1984). During the analysis of the air samples it is necessary to monitor the following parameters: temperature, spare parts/hour, soundproofing and behavior code of the panel (not smoking or using perfume, be cold, be stressed, etc. before performing the analysis to avoid jeopardizing the results) (Brinkmann et al. 2018; Hayes et al. 2014). During the evaluation, panelists should be located in a dedicated and comfortable laboratory with temperature control and they must not be influenced by the response of other panelists or by the panel operator (Capelli et al. 2010; Dravnieks and Jarke 1980).

Although all the panelists are selected according to their individual sensitivity and repeatability regarding the reference gas, *n*-butanol, they must be continuously screened and trained (Brattoli et al., 2011). For this latter purpose, samples of *n*-butanol at different concentration shall be used so that the panel members will not be able to guess the right answer (Capelli et al. 2010).

On the market, there are different types of olfactometers, but two are the most common. The first can be called "yes/no", from the sniffing port, odorless air or air with odor comes out alternatively and the panelist shall indicate on the evaluation card if he/she detects the odor or not. The second olfactometer, called "forced choice" presents two or three different sniffing ports and each panelist shall indicate from which sniffing port the odor comes from (Guffanti et al. 2018; Munoz et al. 2010).

Actually, the EN 13725 edited in 2003 is under revision with respect to storage and materials for olfactometry, sampling techniques, reference material for panel selection, panel size and panel management procedures (EN 13725 2003). The reviewed EN 13725:2003 should be available by the end of 2019.

5.2.1.2 Field inspection

Field inspection methodology is standardized by EN 16841:2016, it is a field analysis that uses a panel of people (from 2 to 8) who assess the presence or absence of an odor directly in the ambient air. EN 16841 (starting from pre-existing VDI Guideline 3940-Part 1 and Part 2) establishes two different methods: Grid and Plume methods (EN 16841-1 2016; EN 16841-2:2016 2016). The application of the grid method is odor exposure, whereas of the plume method is odor extent from a specific source (Capelli et al. 2013b).

Grid method consists in designing a grid around the odor source to cover all the sensitive receptors and areas where complaints were recorded (Dentoni et al. 2013). Each panelist has a specific path that covers different intersection points (which correspond to evaluation points) of this grid and walks inside and outside the mapped area to prevent odors habituation. Prior to the field inspection, odor bags shall be collected in order to train the panelists for the specific odor/odors they need to recognize (Dentoni et al. 2013; Guillot et al. 2012). The duration of a single evaluation is 10 minutes, during which every 10 seconds the panelist shall indicate on his/her card if he/she perceives no odor (e.g. 0 = no odor), one of the training odors (e.g. 1 = landfill, 2 = livestock, etc.), the mixture of the training odors (e.g. 3 = 1+2) or a different odor that is no under evaluation (e.g. 4 = barbecue) (Dentoni et al. 2013; Diallo et al. 2018). Each measurement is defined as "odor hour" if 10% of the measurements is attributable to that/those specific odor/odors considered (Diallo et al.

2018; Guillot et al. 2012). For each area, the EN 16841 establish 104 evaluations (or 52 for 6 months) given by the sum of the four intersection points considering that in each point the panel goes 26 times a year. At the end of the field inspection, the result will be a map with different squares where will be reported the odor frequency, expressed as a percentage, derived from the sum of "odor hours" (EN 16841-1 2016). The minimum period for this evaluation is at least 6 months; however, the evaluation of one year is recommended to take into account the seasonality.

The Plume method is used to determine the extent of detectable and recognizable odor from a specific source using direct observation in the field by panelists under specific meteorological conditions (Guillot et al. 2012). The extent of the plume is assessed as the transition from zones in which odor is not perceived to zones in which is perceived. It is used to verify the outputs of odor dispersion modeling (Capelli and Sironi 2018). The plume method includes two approaches: the stationary and the dynamic method (Dentoni et al. 2013; Van Elst and Delva 2016).

According to the stationary method, five panelists move in the field upwind, approaching the odor source and along parallel lines, that are perpendicular to the plume extent (Van Elst and Delva 2016). Panelists follow the same procedure illustrated for the grid method, except for the fact that their path will consist of parallel lines and they do not have to identify odors but only the presence or absence of recognizable odors (Capelli et al. 2013b). A measurement cycle shall consist of at least 20 single measurements (four intersection lines each consisting of five single measurement points), and eight transition points. Transition points are those between points with absence and presence of odor. The distance between the line without odor presence and the nearest line with odor presence shall be less than 20% of the maximum odor plume, otherwise, the odor plume remains too undefined.

According to the dynamic method, two panelists follow exactly the same indications of the stationary method with the only difference that they will move zigzag inside and outside from the plume, to identify the presence or absence of recognizable odors (Brinkmann et al. 2018; EN 16841-2:2016 2016; Guillot et al. 2012; Van Elst and Delva 2016).

Regardless of the method adopted, field inspections must occur on different days, with different weather conditions and prior to the analyses, an inspection should be planned to decide the optimum location and distribution of sampling points, accordingly to the topography and also to the different kind of odor sources (Sówka 2010). To define the inspection area, information on the prevailing wind direction and wind speed have to be taken into account. Moreover, the panel should be composed of qualified assessors, selected using the criteria proposed for the dynamic olfactometry procedure (Guillot et al. 2012).

In conclusion, the grid method uses panelists to characterize odor exposure in a defined assessment area over a sufficiently long period (typically one year) to include all different meteorological conditions of that location. Instead, the plume method uses panelists to determine the extent of the odor impact, under specified emission situation (based on the source characteristics) and meteorological conditions (including specific wind direction, wind speed, and boundary layer turbulence).

5.2.1.3 Recordings from residents

This method is based on reporting cards filled out by the population living near the odor source, engaged as regular odor monitors (Capelli et al. 2013b). It can be useful to identify the origin of the odor episodes when the odor episode is geolocalized or when a new plant is built (Aatamila et al. 2011). The database can be built using the reporting cards, where residents have to sign their name, their position when they perceive the odor, the date and duration (start and end time) of the odor episode, and a description of both quality and intensity of the odor (Blanes-Vidal et al. 2012; Gallego et al. 2008). Data obtained from social participation can subsequently be associated with the meteorological parameters recorded during the episodes detected (Sironi et al. 2010).

Usually, this method takes a long time, but it is not expensive. However, the limit is the weak scientific stability of the data due to the psychological effect of the population involved that need to be formed to have similar recordings (Brinkmann et al. 2018; Capelli et al. 2013b; Sironi et al. 2010). Even if they are trained, they cannot be considered experts like panelists used in the above-mentioned sensorial techniques. Moreover, data variability based on people's characteristics must be considered. Indeed, for example, people living in rural place probably will not associate livestock activities to malodor, instead, people living in big cities will consider them a source of odor nuisance. Finally, since participants are recruited on a voluntary basis, it is necessary making them actively involved in the odor impact assessment, for example, asking them to regularly complete the reporting cards. Social participation can be useful to identify odor episodes or record odor incidents (Capelli et al. 2013b) and allows to build sensory databases (based on completed questionnaires) (Gallego et al. 2008). Data obtained from residents need to be associated with meteorological parameters recorded during the perceived odor episodes, thus allowing the comparison to dispersion models (Sironi et al. 2010).

5.2.2 Analytical techniques

5.2.2.1 Gas chromatography-mass spectrometry, GC-MS

In the chemical analysis, the use of gas-chromatographic techniques coupled with mass spectrometry allows to identify and quantify odorous compounds, even if the source cannot be characterized and it is not possible to know if the olfactory sensation is due to an individual constituent or to the whole mixture (Blanco-Rodríguez et al. 2018; Cadena et al. 2018).

The principle of the gas chromatographic method is the separation of the components from the odor mixture according to their affinity with the stationary phase in the column (Munoz et al. 2010). Since each type of molecule has a different rate of progression, the various components of the analyte mixture are separated as they progress along the column and reach the end of the column at different times (retention time). According to the retention time, components are qualitatively identified. Then they are quantitative identified thanks to mass spectrometry (Dincer et al. 2006). The association of these two techniques amplifies their potential and enables to lower the detection limits, allowing to identify analytes in very low concentrations (Guffanti et al. 2018).

A problem that can arise during GC is the elution problem due to poor chromatographic separation. It occurs when two (or more) compounds due to widely differing retention properties do not chromatographically separate because the late ones remain in the column too long. Changing the chemistry of the mobile phase, stationary phase, temperature, and column or plane length are good methods to increase the separation (Brattoli et al. 2013; Delahunty et al. 2006).

An alternative approach to GC-MS is gas chromatography-mass spectrometry coupled with olfactory analysis (GC-MS/O). The process is equal to classical GC-MS with the only difference that at the end of GC, the sample is split between an MS detector and the trained human panelists (Munoz et al. 2010). Panelists shall press a button and record what they are sniffing. In the end, the olfactogram and the chromatogram are combined. This represents an improvement compared to GC-MS because the human nose is more sensitive, but there is subjective components and panelists shall be really concentrated during the analysis (Brattoli et al. 2013; Guffanti et al. 2018; Hayes et al. 2014) because peaks are eluted very quickly and the panelist at the same time has to both recognize odors and provide a description (Brattoli et al. 2013). Since the odor detection is linked to the human perception, as for dynamic olfactometry, the panel must be periodically screened and trained, and observe a simple behavior code (Capelli et al. 2010). Moreover, in order to obtain reproducible data, panel members are selected according to their sensitivity, and the ability to recall and recognize odor qualities (Brattoli et al. 2013).

Finally, being the GC-MS/O sessions quite long, the position of the sniffing-port must be comfortable (long transfer line) and the panelist should be seated far from the hot chromatograph components during detection to avoid the smell of hot metal (Delahunty et al. 2006).

5.2.2.2 Identification of specific compounds

Analysis of a single gas, for example, NH_3 or H_2S, can be useful when these gases are tracer and representative of a specific source, such as livestock farming and landfill, respectively. However, it is not possible to identify tracer compounds for all situations and or sources, nor to relate analytical concentrations to odor properties, thus considering a single compound may not be enough to determine the effective odor perception (Capelli et al. 2013b). On the other hand, alternative approaches have been established for analysis of VOCs. One approach is to use real-time detection devices, such as PID (photo ionization detectors). PID is a broadband detector that detects ionized species with a UV lamp. The output of this kind of sensors is a non-specific total concentration of organic compounds, expressed in ppm (or ppb) equivalent (Biasioli et al. 2011; Chen et al.

2013). Alternatively, colorimetric sensors can be used, which are easy to use and cost effective. Thanks to the interactions between the analyte and the responsive colorants VOCs are identified (Lin et al. 2011).

5.2.2.3 Electronic nose

The electronic nose allows a qualitative classification of the analyzed air, attributing the air sample to a specific olfactory class. It is composed of an array of sensors that simulate the receptors of the human olfactory system and a computer that simulates the response of the human brain. The output is a pattern, which is typical of the gas mixture (Blanco-Rodríguez et al. 2018; Capelli et al. 2014a). The electronic nose includes three major parts: a matrix of sensors, a data processing system, and a pattern recognition system. They simulate three components of the human olfactory system: nose receptors, olfactory bulb, and brain. Odor interacts with the surface of the sensor and causes a change in certain chemical and/or physical properties, these variations are converted to an electronic signal which is sent to the data processing unit. Here feature extraction is performed, followed by an explorative analysis which converts features in results and representations that can be easily interpreted, thanks to statistical analysis. Finally, odor samples thanks to a series of algorithms are classified into clusters (Guffanti et al. 2018). Prior to the measurements, the electronic nose needs to be trained with qualified samples to build a database of reference. For the training, odor bags near the odor source need to be collected to train the nose for odor or odors it has to identify (Brinkmann et al. 2018). For this reason, training is always site-specific. In case of long monitoring times (> 1 year) the nose needs to be controlled to verify if the training is still valid and if sensors have been damaged by adverse weather conditions. Indeed, during analysis, it is important to always keep temperatures and humidity monitored to avoid damaging the sensors (Hayes et al. 2014). Thanks to this method, since the instrument is used in injection, the air is monitored continuously (if installed at the borders of the installation) and at the receptor (sensitive even a few kilometers away). Unfortunately, there is no standard that provides minimum requirements for this type of analysis (Capelli et al. 2014a).

5.2.3 Conclusions on technical approaches to odor measurement

Regardless of the measurement technique adopted (sensorial or analytical techniques), the quality of the results obtained is heavily dependent on appropriate sampling, which is one of the main issues relating to odor characterization and measurement (Capelli et al. 2013a). The purpose of sampling is to obtain representative information on the typical characteristics of odor sources by collecting a volumetric fraction of the effluent (Lucernoni et al. 2016). Unfortunately, these techniques alone are not sufficient to assess odor impact within a community. Indeed, both sensorial (dynamic olfactometry) and analytical techniques are used for the evaluation of odor concentration. In the first case, the concentration is given by a panel of experts based on samples of air, in the second one, it is assessed by means of complex physical and chemical analyses (Mielcarek and Rzeznik 2015).

Odor concentration is necessary to find the Odor Activity Value (OAV), which is defined as the concentration of a single compound divided by the odor threshold for that compound. Knowing the OAV of an odorous compound allows to understand which is the contribute of that specific odorant to the overall odor (Brattoli et al. 2013). However, an approach based just on OAVs is imprecise due to the different values of the odor threshold reported in the literature (Capelli et al. 2013b).

In odor measurement, the evaluation of odor concentration alone is not sufficient. Also, the air flow associated with the monitored odor source have to be taken into account, being these parameters interrelated in most cases. Thus, Odor Emission Rate (OER) has to be evaluated, which is the quantity of odor emitted per unit of time and it is expressed in ouE s^{-1} (Capelli et al. 2013a) (OER will be better described in Section 3 – Air dispersion models).

In Table 1 and 2 are reported the main advantages and disadvantages of sensorial techniques and analytical methodologies, respectively. Odor dispersion models, combined with odor measurements, allow a better understanding of odor nuisance and odor characterization.

Table 1. Advantages and disadvantages of sensorial techniques.

Sensorial techniques	**Advantages**	**Disadvantages**	**Reference**
Dynamic olfactometry	• Recognized and standardized technique • Endpoint assessment • High sensitivity • Possible implementation to atmospheric dispersion models • Sensitivity of human nose is higher than electronic instruments	• Only quantitative characterization • Impossibility of continuous measurements and monitoring • High measurement uncertainty • Not applicable for low odor concentrations • Time consuming • Lower repeatability compared to chemical analysis • Subjectivity of human perception • Psychological factors could affect the evaluation	Blanco-Rodríguez et al. (2018); Cadena et al. (2018); Capelli et al. (2014a); Guffanti et al. (2018); Hayes et al. (2014); Van Elst and Delva (2016)
Field inspection	• Direct determination of the odor impact in terms of frequency (Grid method) or area of impact of the odor at the receptor (Plume method) • Possibility of comparing the results with other methods • Possibility to validate air dispersion method • Sensitivity of human nose is higher than electronic instruments	• High cost • Many data to be processed • Logistical difficulties related to the planning of the evaluation card and to the identification of paths (plume method) • Difficulty of finding an adequate panel, available and not directly involved (grid method) • Lack of reference acceptability values • Time consuming • Subjectivity of human perception • Psychological factors could affect the evaluation	Both (2001); Hayes et al. (2014); Van Elst and Delva (2016)
Recordings from residents	• Low or no cost	• Management difficulties in collecting similar data	Di Francesco et al. (2001); Hayes et al. (2014); Sironi et al. (2010)

	• Useful for involving and sensitizing citizenship (psychological effect) • Sensitivity of human nose is higher than electronic instruments	• Poor scientific stability of the data (due to psychological effect) • Lack of reference acceptability values • Possibility of bias • Long response times • Subjectivity of human perception	

Table 2. Advantages and disadvantages of analytical techniques.

Analytical techniques	**Advantages**	**Disadvantages**	**Reference**
Gas chromatography-mass spectrometry (GC-MS)	• Recognized and repeatable technique • Possibility to identify and quantify single components • Possibility to perform the analysis at the emission and at the receptor • Possible implementation to atmospheric dispersion models • Objectivity of the evaluation	• No information on the odor impact provided • Non-reliable in case of mixture of many odorants at very low odor concentration • High technical requirements • Detection limits below the odor detection threshold of the compounds • Precise calibration required • Interaction between odorants not detected • Possible masking effects with complex odor mixtures	Blanco-Rodríguez et al. (2018); Cadena et al. (2018); Capelli et al. (2014a); Guffanti et al. (2018); Hayes et al. (2014); Mielcarek and Rzeznik (2015)
Identification of specific compounds	• Possibility to make measurements at the receptor • Possibility to control accidental emissions • Objectivity of the evaluation • User-friendly (PID)	• Non-reliable in case of mixture of many odorants at very low odor concentration • Possible only with a source with a particular type of emission • Results depend on the type of instrument and sensor • High costs • Impossible correlation with odor concentration (different response factors, different OTV) • Inability to recognize source (many interferers, no speciation)	Capelli et al. (2013b); Hayes et al. (2014)
Electronic nose	• Continuous analysis of the ambient air at the receptor • Direct determination of the presence/absence of odors where odor annoyance is lamented • Determination of odor provenience in case of multiple sources • Odor recognition/classification (at the source) • Possibility of comparing the results with other methods • Objectivity of the evaluation • Positive effect on population which is comforted by the presence of an instrument that perform a continuous analysis of the ambient air	• Absence of a specific regulation that standardize the method • Instrument complexity: precise procedures for its use (training and data processing) • Odor impact assessment based only on the quantification of the frequency of odor episodes and not on their intensity • Interaction between odorants not detected	Blanco-Rodríguez et al. (2018); Cadena et al. (2018); Capelli et al. (2014a); Di Francesco et al. (2001); Guffanti et al. (2018); Hayes et al. (2014)

5.3 Air dispersion models

Different types of models, that take into account meteorological, topographic and emission data, can be used to determine odors dispersion into the atmosphere (Yu et al. 2010). They offer the possibility of mathematically simulating the spatial and temporal variation of odor concentrations (Zhou et al. 2005) and can provide estimates of odor levels in both current and future emission scenarios, predicting the atmospheric impact of a facility before being realized (Sheridan et al. 2004). Therefore, these models are useful to determine appropriate setback distances between production facilities (farms, industries, and waste treatment plants) and neighboring areas (Capelli et al. 2013b; Danuso et al. 2015; Guo et al. 2004).

Most dispersion models are Gaussian models, which assume the concentration profile across the plume to follow a Gaussian probability curve (Daly and Zannetti 2007). The other models follow the Lagrangian or Eulerian approaches (Danuso et al. 2015). They are mathematical models used to estimate (in case of existing facilities) or to predict (when planning new operations or evaluating the efficacy of mitigation strategies) the downwind concentration of air pollutants emitted from sources such as farms (Zhou et al. 2005).

Gaussian models assume "steady-state" conditions. The meteorological conditions are assumed to remain constant during the dispersion from source to the receptor, which is effectively instantaneous. Also, emissions are considered time-invariant and, for this reason, calculations refer to periods of one hour or less (Danuso et al. 2015). However, emissions and meteorological conditions can vary from hour to hour thus the model simulate hourly-average concentrations, so that calculations in each hour are independent of those in other hours. The plume formula has the uniform wind speed in the denominator and hence breaks down in calm conditions (Danuso et al. 2015). It is usual to specify a minimum allowable wind speed for the model.

Assumptions in plume Gaussian modeling: i) continuous emission, ii) conservation of mass, iii) steady-state conditions, iv) absence of wind in calm conditions. Gaussian dispersion models are the most widely used (Yu et al. 2010) since they are quite easy to be applied, indeed assuming "steady-state" conditions they do not require significant computer resources (Zhou et al. 2005). However, compared to other models, they do not consider topography, so they give a precise evaluation only in flat terrain (Danuso et al. 2015).

Puff models (no steady-state) are an evolution of classical Gaussian models; the plume is presented by a series of independent elements (puff) that evolve over time according to spatial and meteorological characteristics (Zhou et al. 2005). These models are based on sets of equations describing the three-dimensional space concentration generated from a point source and are able to encounter with changing wind and emission data (Jung et al. 2003; Yu et al. 2010).

Lagrangian models (also known as particle models) describe the motion in space of individual, non-interacting elementary odor particles (Danuso et al. 2015). Lagrangian models are based on the idea that pollutant particles in the atmosphere move along trajectories determined by the wind field, the buoyancy, and the turbulence effects (Wilson and Sawford 1996). The final distribution of randomly moving particles gives a stochastic estimation of the concentration field; this means that these models require high computing power as they simulate several trajectories of elementary particles to achieve an adequate accuracy level (Flesch et al. 1995). Lagrangian models are exceptionally efficient close to the source and are particularly suited for elevated point sources. Puff models are far less computationally expensive than particle models but are not as realistic in their description of the pollutant distribution.

In Eulerian models (grid models), the area under investigation is divided into grid cells, both in vertical and horizontal directions and in each grid is calculated the average concentration of pollutant particles. Their limit is the high computing power required, due to the fact they allow a more correct spatio-temporal representation (Danuso et al. 2015). The main difference between Lagrangian and Eulerian models is related to the perspective of atmospheric motion (Nguyen et al. 1997). The first ones take the perspective of an air particle, whereas the second ones define specific reference points in a gridded system and treat the particle phase as a continuum.

Dispersion models usually calculate the hourly mean odor concentrations for every receptor for every hour of the simulation domain. However, the sensation of the odor depends on the momentary (“peak”) odor concentration. For the assessment of peak values, a so-called “peak-to-mean” factor is used in order to account for these fluctuations (Piringer and Schauberger 2013). The goal of the use of peak-to-mean factors is to simulate the rapid response of the human nose. The pick-to-mean factor is calculated by dividing the peak concentration by the 1-h averaged (mean) concentration (Schauberger and Piringer 2012). The applied peak-to-mean factors differ from country to country (Piringer and Schauberger 2013). For example, in Italy, DGR n. IX/3018 15/02/2012 suggests a constant peak-to-mean factor of 2.3. Results are represented as a map of the 98th percentile of the peak odor concentration values (D.G.R. 15 febbraio 2012 e n. IX/3018 Regione Lombardia 2012). Austria suggests variable peak-to-mean factors in function of the atmospheric stability and of the distance from the source (Schauberger and Piringer 2012). The United Kingdom establishes peak odor concentration values at 1.5 ouE m^{-3}, 3.0 ouE m^{-3} and 6.0 ouE m^{-3} (at the 98th percentile), for high, medium and low “offensiveness” industry types, respectively (UK-Environmental-Agency 2002).

Models modified in this way are then able to calculate separation distances for so-called odour impact criteria, a combination of odour concentration (mostly an odour threshold) and a pre-selected exceedance probability according to land use. An overview of various national odour impact criteria can be found in Sommer-Quabach et al. (2014) and Piringer et al. (2016).

A summary of principal dispersion models with their characteristics, advantages, and limitations that have been applied to simulate odor dispersion from waste and agricultural sources are reported in Table 3.

Table 3. Principal air dispersion models with related characteristics, advantages, and limitations.

Model name	Type of model	Characteristics	Consider: Topographic data	Consider: Meteorological data	Advantages	Limitations	Reference
AERMOD (U.S.)		It enables comparison of predicted and measured data and among different kind of source (point, volume, and area sources). T	✓ (includes simple and complex terrain because it is an advanced plume model)	✓	• It follows the default regulatory options consistent with the Guideline on Air Quality Models (U.S. EPA). • Topographic features and meteorological conditions: considered. • Suited for near-field dispersion.	• Under-predict odor concentrations when configured as an area source.	Schulte et al. (2007); Baawain et al. (2017)
LODM (Canada)		Designed for livestock facilities for predicting hourly odor frequency with input hourly meteorological data.	✓	✓	• Adequate in predicting both odor concentration and frequency. •	• Surface parameters (roughness, albedo, and Bowen ratio) essential input required to predict odor concentration and frequency. • Stack height, stack diameter, and exit velocity affect concentration values.	Yu et al. (2013a); Yu et al. (2013b)
STINK (Australia)	Gaussian plume model	It calculates odor emission rates from ground level area sources. It predicts the dispersion of odors downwind of area sources of finite size and any orientation with respect to the wind direction.	X	✓ (averaged over a period of 1 hour or less)	• Applicable to sources of irregular shape or of limited lateral extent. • Any location can be used for the measurements providing the location, the size of the source and the wind direction.	• An accurate description of the source geometry is requested. • Not applicable to wind directions perpendicular to the source.	Galvin et al. (2004); Smith (1995)
OdiGauss software (Italy)		Free multilingual software application for estimating odor dispersion from multiple point sources and to generate the related maps.	X (also parameters of odor emission sources required)	X (for all the odor sources, the weather conditions are considered to be the same)	• User-friendly. • Available in several languages. • Little training required. • Fast in performing simulations. • It can simulate the dispersion of particles (e.g. PM10)	• It allows only short-term simulations. • It does not consider inversion processes of the plume. • It considers only the mean hourly odor concentration. • • Not suitable for complex orographic conditions. •	Danuso et al. (2015)
INPUFF-2 (U.S.)	Gaussian puff model	This model can simulate airborne pollutants dispersion of from semi-instantaneous or continuous point sources.	✓	✓ (actual, not empirical formula)	• It can handle single and multiple odor sources. • It can deal with multiple receptors at the same time. • It uses actual odor emissions.	• It correlates best with the field data at 100 m distance and worst at 400-m and 500-m distances. • The scaling factors used are only valid for this model.	Zhu et al. (2000); Asadollahfardi et al. (2015)
CALPUFF (U.S.)	Lagrangian model	It is a multi-layer, multi-species non-steady-state dispersion model that simulates the effects of time- and space-varying meteorological conditions on pollution transport, transformation and removal. .	✓	✓	• It contains modules for complex terrain effects, over-water transport, coastal interaction effects, building downwash, wet and dry removal and simple chemical transformation. • It keeps track of recent past pollution information (e.g. over a few hours); as a result, pollutant concentrations that occurred during these previous hours have an impact on current and future pollutant dispersion.	• Applied best only to long term modeling. • It tends to underestimate peak intensities close to the source. • It needs realistic emissions rates, that are difficult to estimate. • It requires a great number of inputs.	de Melo et al. (2012); Ranzato et al. (2012)

					• It can predict actual and potential effects of odor emissions at any point in space with a high temporal resolution. • It can identify the odor nuisance generated by single emission sources.		
CALGRID (U.S.)	Eulerian model	It is a photochemical transport and dispersion model that requires three-dimensional fields of air temperature and vertical velocity.	✓	✓ (Precipitation rate not considered)	• It includes modules for: horizontal and vertical advection/diffusion, dry deposition, photochemical mechanism • Transport dispersion is more realistic • It can simulate multi-day scenarios • More accurate compared to the other models	• It requires a great number of inputs. • It needs high computing power	Scire et al. (2000); Yamartino et al. (1992)

Regardless of the type of model, necessary input data are: i) topographic and orographic data; ii) meteorological data (e.g. air temperature, relative humidity, air pressure, solar radiation, precipitations, wind speed, and wind direction); iii) emission data (identification and characterization of the odor source, quantification/estimation of the amount of pollutant emitted in unit of time) (Schauberger et al. 2012; Schulte et al. 2007; Sironi et al. 2010). The quality of the input data affects the goodness of the results other than the choice of the model (Capelli et al. 2013b).

Topographic and orographic data shall include the characteristics of the terrain roughness, heights of potential receptors near the emitting source, the planimetry of the plant and of the surrounding impact area (Schauberger et al. 2012; Sheridan et al. 2002).

Regarding meteorological input data, besides wind data (speed and direction), the main weather variables involved are temperature, solar radiation, atmospheric pressure, humidity and precipitations (Danuso et al. 2015; Piringer and Schauberger 2013). Furthermore, information on the stability of the atmosphere and the planetary boundary layer (the portion of the atmosphere where the generation, decay, transformation, and diffusion of most pollutants take place) is important, usually expressed as time series over at least one year (Piringer and Schauberger 2013). The classifications proposed by Pasquill (1961) and Gifford (1959) for atmospheric stability classes are usually adopted (ranging from class F “very stable”, to class A “very unstable”, according to the meteorological conditions). Meteorological data can be obtained from one or more near surface meteorological stations to define a representative station for the emission source considered (Capelli et al. 2013b; Wu et al. 2019).

In addition to topographic and meteorological data, emission data are necessary for the simulation of odor dispersion, since the evaluation of odor concentration alone is not sufficient, also the air flow associated with the monitored odor source has to be taken into account. OER (ouE s^{-1}) is calculated by taking the product of the odor concentration and flow rate from a source. So, the method adopted for OER estimation is strictly related to the characteristics of odor source (point or area). In case of point sources, OER is the product between the odor concentration and the emitted air flow. For active area sources, the average emitted odor concentration needs to be used (Capelli et al. 2013a). Finally, in case of passive area sources, the procedure is quite complex. Firstly, it is necessary to calculate the Specific Odor Emission Rate (SOER), which is the odor emitted from the area source per unit of time and surface (ouE m^2 s^{-1}), then OER is calculated by multiplying the SOER and the emitting surface of the considered source (Lucernoni et al. 2016).

To predict the odor impact, when data for a specific source are not available, odor emission factors (OEFs) are fundamental for estimating emissions since they enable the estimation of the OER. OEFs correlate the quantity of odor emitted with the activity associated with the emission of that odor. OEFs can be derived from experimental data or found in the literature (Capelli et al. 2014b). OEFs are usually expressed as OER divided by a specific activity index, expressed in terms of unit, such as animal, animal body weight, gross weight production, area, production place, the site surface or a time unit (Mielcarek and Rzeznik 2015).

5.3.1 Conclusions on air dispersion models

Different types of air dispersion models are available and can be applied to simulate odor dispersion into the atmosphere. They are considered a useful tool in assessing the odor impacts associated with existing and future or modified (with abatement systems) emission sources.

Regarding Gaussian models, they assume that dispersion in cross-wind direction and in the vertical direction has a Gaussian distribution, with the maximum concentration in the center of the plume. Their main advantage is that they require little computing power. However, they are less accurate due to their assumptions (no three-dimensional space characteristics; no memory of past conditions; no calm wind conditions).

Conversely, Lagrangian and Eulerian models need high computing power because, to achieve an adequate accuracy level, they simulate several trajectories of elementary particles and consider more inputs. However, having a more correct spatial-temporal representation, they are more accurate and represent a more advanced tool. Their limit is that turbulence is difficult to represent (Capelli et al. 2013b), indeed turbulence that characterizes the planetary boundary layer has mainly two origins: movement of air masses with viscosity effects on a rough surface (mechanical turbulence) and soil heating/cooling effects caused by daily solar radiation and nightly radiative cooling effects (Schiffman et al. 2005).

5.4 Major sources of odor nuisance

5.4.1 Waste from treatment plants

Landfill, compost and anaerobic digestion plants are among the main disposal technologies to treat municipal solid waste (MSW) (Cadena et al. 2018). During waste handling and biological decomposition steps, several gaseous compounds are emitted from the organic matrix (Sarkar and Hobbs 2002). VOCs, NH_3, and H_2S are responsible for the unpleasant odors (Rincón et al. 2019), contributing to the odor impact (Moreno et al. 2014). Among the organic compounds, it is possible to find: terpenes, alkanes, oxygenated compounds, aromatics, ketones, and other compounds such as sulfur compounds (Cheng et al. 2019; Fang et al. 2012). Sulfur compounds or VSCs consist of VOCs that contain sulfur, they are toxic and easily perceived at extremely low concentrations, they are produced by organic matter degradation during waste treatment and composting conditions (Rincón et al. 2019; Zhang et al. 2013).

The organic fraction of MSW (OFMSW) is usually treated with anaerobic digestion and composting (Font et al. 2011). Although the objective of these technologies, as well as other waste treatment technologies, is to enable sanitization of the waste by the elimination of pathogenic microorganisms and transforms the organic fraction in soil amendment (Renaud et al. 2017), they emit potential toxic compounds, such as VOCs, VSCs, NH_3 and nuisance odors (Moreno et al. 2014; Nie et al. 2019; Rincón et al. 2019). Degradation of the organic fraction by microorganisms is the main cause of odor production (Pierucci et al. 2005). Ammonia emissions depend almost on the C/N ratio of the waste, temperature, moisture content and pH (Moreno et al. 2014; Pagans et al. 2006), whereas VSCs emissions are correlated to temperature and O_2 concentration (Higgins et al. 2006; Moreno et al. 2014). Moreover, offensive odors generated by waste treatment depend on the type of raw material, the stage of the decomposition and the operating conditions at the site (Bruno et al. 2007; Toledo et al. 2018). As an example, Toledo et al. (2019) found that fresh organic waste such as sewage sludge and OFMSW were the most influential odorous substrates, due to their high concentration in biodegradable organic matter.

The presence of odors associated with VOC emissions has been investigated by a wide number of researchers (Bruno et al. 2007; Defoer et al. 2002; Fang et al. 2012; Mao et al. 2006; Pierucci et al. 2005), that focused on how the odors mixture change according to the type of waste, treatment technology and the sites within the treatment plant, as reported in Table 4.

According to the literature reported in Table 4, in landfill the main emissions sources can be identified in the MSW-related area, the leachate-related area, and the sludge-related area, in particular gas extraction wells, sludge discharge area, sludge disposal work place, leachate storage pool, wharf, dumping pool, lane, active working face, and gas vent (Cheng et al. 2019; Fang et al. 2012). Instead in composting plants, odorants are released by belt conveyor area, pile-turning workshop, stacking workshop other than by the different substrates (agricultural wastes, biowastes, green wastes, and kitchen waste) (Cheng et al. 2019; Rincón et al. 2019; Zhang et al. 2013).

In general, to characterize the odor and chemical emissions released from the different sources of waste, GC-MS and dynamic olfactometry have been used. In particular, GC-MS to determine the chemical and dynamic olfactometry the odor concentration.

As it is possible to see from Table 4, among VOCs, terpenes, hydrocarbons, and oxygenated compounds were the most emitted chemical families. While, among nitrogen and sulfide compounds, NH_3 and H_2S were the most abundant compounds emitted, respectively. Not in all studies results of the chemical analyses showed a correlation with odor concentration measured by dynamic olfactometry.

Table 4. Odor compounds from different sources of waste.

Waste	Technology	Source	Odor compounds	Reference
Solid wastes and digestate	Composting reactor[a]	Agricultural wastes	Nitrogen compounds (NH_3 and trimethylamine), sulfide compounds, terpenes, benzene, toluene, and methyl thiocyanate	Rincón et al. (2019)[1,2]
		Biowastes	Terpenes, and oxygenated compounds (ethanol, methanol, ethyl acetate, methyl acetate, and 2-butanone)	
		Green wastes	Terpenes (limonene and α-pinene), ketones (acetone and 2-butanone), and alcohols (methanol)	
		Sewage sludge	Nitrogen compounds (NH_3), terpenes (limonene), sulfide compounds (methanethiol and dimethyl sulfide), and acetic acid	
		Municipal solid waste	Terpenes (limonene and α-pinene), nitrogen compounds (NH_3), sulfide compounds (methanethiol), ketones, and alcohols (methanol)	
		Digestates	Nitrogen compounds (NH_3), terpenes (limonene, α-pinene, and myrcene), sulfide compounds, alcohols, aldehyde and ketones, acids, esters, and alkenes	
Solid waste	Landfilling[b]	Landfill at the moment the material was discharged and during a period of inactivity at the landfill	2-butanone, α-pinene, tetrachloroethylene, dimethyldisulfide, β-pinene, limonene, phenol, and benzoic acid	Bruno et al. (2007)[1]
MSW	Landfilling[b]	Wharf, dumping pool, lane, active working face and gas vent	Sulfides (H_2S and ethyl sulfide), aromatics, alkanes (heptane)	Cheng et al. (2019)[1,2]
MSW	Landfilling[b]	Landfill	Styrene, toluene, xylene, acetone, methanol, n-butanone, n-butylaldehyde, acetic acid, dimethyl sulfide, and ammonia	Fang et al. (2012)[1]
		Gas extraction wells	Acetaldehyde, ethyl benzene, xylene, methylamine, and dimethyl formamide	
		Sludge discharge area	Methyl mercaptan, valeric acid, and isovaleric acid	
		Sludge disposal work place	Carbon disulfide, acetone, 3-pentanone, methanol and trimethylamine	
		Leachate storage pool	Hydrogen sulfide, n-butylaldehyde, and acetic acid	
MSW	Landfilling[b]	Inside and away from the dumping area	Benzene, alkylbenzenes, terpenes (α-pinene, β-pinene, camphene, methyl-isopropylbenzene, and limonene), aromatic hydrocarbons (BTEX), chlorinated aromatics and hydrocarbons	Zou et al. (2003)[1]
MSW	Anaerobic digestion[b]	Working face	Ethyl alcohol, α-pinene, hydrogen sulfide, dimethyl sulfide, limonene, methyl mercaptan, dimethyl disulfide, and diethyl sulfide	Wenjing et al. (2015)[1,2]
		Mixed paper	Alkylated benzenes, alcohols and alkanes	
		Yard wastes	Terpenes (α-pinene and $_D$-limonene), aromatic hydrocarbons, ketones, and alkanes	
		Food wastes	Sulfides, acids, alcohols, and terpenes	
MSW	Anaerobic digester and composting[b]	Fresh compost fraction	Limonene, p-cymene, pinene	Moreno et al. (2014)[1]
		Rejected compost fraction	Limonene, ketones, pinene	
		Leachate pond	Limonene, hydrocarbons, cyclohexane, pinene, ketones	
		Landfill leaks	p-cymene, pinene, C4-C5 hydrocarbons, BTEX	
MSW	Composting[b]	Belt conveyor area, pile-turning workshop, and stacking workshop	NH_3, oxygenated compounds (ethyl acetate and 1-butanol), alkanes (n-hexane and heptane)	Cheng et al. (2019)[1,2]

MSW	Composting[a]	MSW	Hydrogen sulfide, methyl mercaptan, dimethyl sulfide, carbon bisulfide, and dimethyl disulfide	Zhang et al. (2013)[1]
		Kitchen waste		
		Kitchen waste mixed with dry cornstalks		
OFMSW	Composting[a]	All waste	Aromatic VOCs: toluene, ethylbenzene, 1,4-dichlorobenzene, p-isopropyl toluene, and naphthalene	Komilis et al. (2004)[1]
Green waste	Natural decay[b] and composting[a]	Feedstock	Terpenes: α-pinene, β-pinene, 3-carene, camphene, β-myrcene, and $_{D}$-limonene	Büyüksönmez and Evans (2007)[1]
MSW	Lab-scale reactor[a]	MSW headspace before processing	Alkanes, terpenes, benzene, and halogen-containing compounds	Pierucci et al. (2005)[1,2]
		Airflow stream exit from the reactor	Terpenes (α-pinene, β-myrcene, and D-limonene), monocyclic arenes (C2, C3, C4 benzenes), alkane, halogenated compounds and esters	
		Before and after the biofilter	Terpenes, toluene, monocyclic arenes (C2, C3, and C4 benzenes), halogen-containing compounds, alkane, and bicyclic arenes	
		Condensate and leachate of the reactor	Alkane, alcohols, ketones, aldehyde, acid, ester, terpenes, benzenes, phenols, heterocyclic, chlorinated, sulfurated, nitrogenated, and phosphorated	
		Airflow stream exit from the reactor	Terpenes (α-pinene, β-myrcene, and D-limonene), monocyclic arenes (C2, C3, C4 benzenes), alkane, halogenated compounds and esters	
MSW	Mechanical–biological waste treatment[b]	All emitting areas	Alkanes, aromatic hydrocarbons, terpenoids, esters, alcohols, ketones, halocarbons, aldehydes, acids, ethere, and furans	Gallego et al. (2012)[1]
		Rotating biostabilizers	Esters, acids, and aldehydes	
		Shipping warehouse	Esters, acids, and aldehydes	
		Composting tunnels	Ketones (methyl ethyl ketone), aromatic hydrocarbons (BTEX and styrene), esters, and acids	
		Digest centrifugal	Esters, aldehydes, acids, organosulfurs, and alcohols	
		Humid pre-treatment	Esters, aldehydes, acids, and organosulfur	

[a]Laboratory experiment.

[b]Common disposal methods.

[1]GC-MS or GC-FID.

[2]Dynamic olfactometry.

5.4.2 *Livestock farms*

Intensive livestock farming, especially swine operations (Trabue et al. 2011), extensively contributes to NH_3, VOCs and particulate matter (PM_{10} and $PM_{2.5}$) emissions (Bibbiani and Russo 2012). The agricultural sector is currently responsible for the vast majority of NH_3 emissions in the European Union (EEA 2018). NH_3 is an atmospheric pollutant causing soil acidification, nutrient-N enrichment of ecosystems, and eutrophication of terrestrial and aquatic ecosystems (Erisman et al. 2007). Moreover, in the atmosphere it reacts with other compounds to form ammonium sulfate and ammonium nitrate aerosols, leading to the formation of secondary inorganic aerosol ($PM_{2.5}$), a potential hazard (Kiesewetter and Amann 2014). Therefore, NH_3 affects human and animal health both as a gas and as particulate matter (Wagner et al. 2015). The particulate form of NH_3 has broader implications for the population, while the gaseous form is a localized issue for the health of animals and agricultural workers (Kafle and Chen 2014). Emissions of NH_3 mainly occur from feces and urine in housing and manure storage systems, from excreta of grazing animals voided on pastures and from agricultural land following application of manure and mineral N fertilizers (Velthof et al. 2014). The principal key categories for NH_3 emissions considered in EU are: i) animal manure applied to soils; ii) inorganic N-fertilizers; iii) non-dairy manure management; iv) dairy cattle manure management; v) swine manure management; they jointly make up 52 % of total NH_3 emissions (EEA 2018).

As for NH_3 emissions, manure and urine are the main constituents of odor from livestock operations (Barth et al. 1984; Kreis 1978; Lemay 1999). Particularly, storage, handling and land application of livestock manure are the main causes of odor annoyance (Hansen et al. 2006; Sheridan et al. 2002). Offensive odor is related to the incomplete anaerobic decomposition of animal wastes (slurry or manure) (Wheeler et al. 2011). However, odor emissions from animal production facilities are a function of many variables including species, housing types, feeding methods, management factors, manure storage, and handling methods (Guo et al. 2004; Jacobson et al. 2005). Instead, their impact on nearby communities depends on the amount of odor emitted from the site, the distance from the site, weather conditions, topography, other than odor sensitivity and tolerance of the neighborhood (Danuso et al. 2015). Finally, the extent of emissions depends on: the size of the settlement, phase of the rearing cycle, feeding operations, type of building, conditioning and ventilation, type of paving and manure removing and collection systems (Guo et al. 2006; Mielcarek and Rzeznik 2015).

Regarding species, for example in the north of Italy, intensive poultry and pig farming in are major contributors to ammonia, odor and particulate matter emissions (Bibbiani and Russo 2012). Similarly, in Denmark, odor annoyance is strictly related to pig farming, being Denmark a country with an intense production of pork meat (Cantuaria et al. 2017). For swine facilities, Ni et al. (2012) identified six major sources of VOCs: confined spaces, wastewater, air above wastewater surfaces, ambient air nearby, manure, dust inside and outside pig barns.

For swine facilities, different authors have investigated the relation between animal buildings and odors (Akdeniz et al. 2012; Blanes-Vidal et al. 2009; Miller et al. 2004; Ye et al. 2008). Akdeniz et al. (2012) reported that OER for pig finishing rooms were lower than OER for sow gestation barns although management characteristics of swine building were similar (slatted floor type, mechanical tunnel ventilation, deep pit manure storage removed twice a year) and they differed only for the average body weight of animals and for the feeding method. Miller et al. (2004) examined the OER differences between deep pit buildings and shallow pit systems, taking into account building characteristics and farming management (air cleanliness, barn cleanliness, manure depth, and pig density). In the end, deep pit was found to have lower OERs than shallow pit systems. Ye et al. (2008) focused only on NH_3. From the results of their study, NH_3 emission rate resulted to be influenced by ventilation rate, floor slat opening and the air headspace height in the slurry pit. Differently Schauberger et al. (2014a) focused on constant emission factors vs time-resolved emission models. Usually to calculate setback distance between livestock building and nearby residents OER is estimated as an annual constant value, obtained by multiplying live mass of the animals and OEF. However, this approach is inappropriate since the live mass increases and also the odor release is influenced by the indoor temperature, ventilation rate, animal activity and so on. Taking into account these variables using time-resolved OER allows to obtain reliable separation distances, avoiding bias due to the assumption of an annual mean value of OER.

In poultry production, Dunlop et al. (2016) identified litter as the primary source of odor caused by anaerobic and aerobic microbial activity. Also, Hayes et al. (2006) reported that litter is an important factor in the production of odor and NH_3 emissions for broiler units.

Regarding dairy farms, buildings and manure management (flooring system, removal system, and land application) are a relevant source of NH_3 and odors (Baldini et al. 2016; O'Neill and Phillips 1991). Odor is a result of the incomplete anaerobic degradation of manure (Barth et al. 1984). Baldini et al. (2016) identified higher NH_3 emission in dairy farms equipped with scrapers, and lower with perforated floor or flushing system for manure removal.

Approximately 330 different odorous compounds have been identified in swine production facilities (Schiffman et al. 2001), 110 in dairy facilities (Filipy et al. 2006), and more than 75 in animal manure (Barth et al. 1984). Regardless of livestock facilities, relevant odorants included many acids, alcohols, aldehydes, ketones, esters, ethers, aromatic hydrocarbons, halogenated hydrocarbons, terpenes, other hydrocarbons, amines, amides, nitriles, phenols, steroids, other nitrogen-containing compounds, and sulfur-containing compounds, and other compounds (Barth et al. 1984; Filipy et al. 2006; Schiffman et al. 2001).

5.5 Mitigation strategies and odor impact assessment

5.5.1 Mitigation Strategies against odor nuisance

The relevance of the odors on public health and the increasing interest of national and international authorities have led Authorities and Governments to tackle the problem. In Europe, according to the Directive 2008/98/CE, "Member States shall take the necessary measures to ensure that waste management is carried out without endangering human health, without harming the environment and, in particular: (b) without causing a nuisance through noise or odors". Moreover, Commission Implementing Decision (EU) 2018/1147 establishes best available techniques (BAT) conclusions for waste treatment and Commission implementing decision (EU) 2017/302 establishes BAT for the intensive rearing of poultry or pigs.

In the U.S. the Environmental Protection Agency (EPA) does not regulate odor, even if it is in force the Clean Air Act, a federal law designed to control air pollution on a national level. Nevertheless, EPA allows states to regulate odor directly. States that try to regulate odor, follow the principles of the Nuisance Laws, that means that they identified odor as a nuisance and established limits for odorous emissions as a nuisance phenomenon (Brancher et al. 2017; Nicell 2009).

In Japan, the Offensive Odor Control Law regulates offensive odors emitted from business activities in order to preserve the living environment and to protect people's health (Government of Japan - Ministry of the Environment 2019).

Moreover, some countries (e.g. Germany, The Netherlands, Switzerland, Austria, and Canada) have established guidelines based on minimum separation distances between livestock units and residential areas, for determination of odor-annoyance-free level (Ubeda et al. 2013). Setback distances can be determined by either by empirical models or by a combination of experience and calculations by dispersion models. Using this last technique, separation distances are calculated in a direction-dependent manner (Piringer et al. 2016). Dispersion models are able to predict time series of ambient odor concentrations with suitable meteorological data and source information (Capelli et al. 2013b). Combinations of tolerated exceedance probabilities and threshold odor concentrations are referred to as odor impact criteria (OIC) (Piringer et al. 2016). In the study of Sommer-Quabach et al. (2014) a wide variety of OIC used to determine separation distances is reported, thus avoiding odor nuisance and complaints by the residents. To establish appropriate OIC it is necessary to consider odor concentration, intensity and hedonic tone and their relationship (Huang and Guo 2018). These impact criteria are selected by the responsible authorities and vary by a quite extend (Sommer-Quabach et al. 2014). In this regard, for example, in Austria a peak-to-mean approach is used to assess the odor perception (Piringer et al. 2016). In particular, in Germany and Austria the separation distances are established with two empirical models based on dispersion model AUSTAL2000 (also included in German VDI guidelines) and AODM, respectively. These empirical models are based on equations that include one or more factors, such as type of the animal, indoor air temperature and meteorology (Wu et al. 2019).

Overall, in empirical models (experience-based) setback distances are established according to different scaling factors, determined by animal number and species, weather parameters, housing characteristics (e.g. ventilation, manure collection system, etc.) and abating technologies used. Subsequently, these factors have been used to adjust dispersion models to determine odor-annoyance-free intensity (Lim et al. 2000; Yu et al. 2010). For example, the OFFSET (Odor From Feedlots − Setback Estimation Tool) model was developed in Minnesota, taking in consideration numerous emission measurement, an air dispersion model (INPUFF-2) and historical weather data (Jacobson et al. 2005). In Canada Minimum Distance Separation guidelines (MDS-I and II) were developed by the Ontario Ministry of Agriculture, Food, and Rural Affairs in the 1990s (OMAFRA 1995a; b), with separate procedures for buildings and manure storage units (Guo et al. 2004). Instead, AODM (Austrian Odor Dispersion Model), estimates the odor emission by considering animal number and species, housing and ventilation systems, handling of manure inside the building, the feeding methods, land use, and topography (Guo et al. 2004; Schauberger et al. 2001). In conclusion, since the setback distance models consider different scaling factors, they generate different odor-annoyance-free level, ranging from 99% to 91% (Guo et al. 2004). However, these models, used to determine separation distance, need the same input data previously described in Section 3, namely topographic, meteorological and emissions data. Regarding this last input, OERs are usually estimated as an annual constant value (Hayes et al. 2006) obtained by multiplying the mean live mass of the animals by a constant OEF. This means that the increasing of the live mass of the animals, as well as variation in indoor air temperature and ventilation rate are usually not considered (Schauberger et al. 2013). Therefore, assuming an annual mean and a constant OEF to calculate OER is inappropriate, as pointed out by Schauberger et al. (2014a) and Schauberger et al. (2014b). In their studies, they suggest using an emission model which considers a time series of the OER, to obtain a more realistic description of the odor emission characteristics and to avoid overestimation (in winter) or underestimation (in summer).

In addition to separation distance, other strategies against odor nuisance can be applied. Generally, the odor control strategies involve different measures oriented to prevent, control dispersion, minimize the nuisance or remove the odorants from emission. Prevention of odorant formation at the source can be obtained by adequate process design and good operational practices. Establishing buffer zones or installation of odor covers are useful for the control of dispersion of the emissions (Guo et al. 2004; Hörnig et al. 1999). To minimize/remove the nuisance an alternative to the traditional treatment technologies (e.g. scrubber, incineration, and biofilter) is the use of chemical additives designed to mask, neutralize or minimize the perception of odorous emission (McCrory and Hobbs 2001). However, as reported by Bortone et al. (2012) these technologies are not suitable to treat extensive areas such as waste landfills.

Also composting can be applied to reduce odorous emissions (Hurst et al. 2005). The odor reduction efficacy of composting is strictly linked to well functioning composting plants, where the conditions (pH, temperature, and aeration rates) are maintained optimal throughout the whole process (Sundberg et al. 2013). The odor emission reduction can be furtherly enhanced by increasing compost bulk density, Hurst et al. (2005) obtained a reduction by up to 97% of odors, and sulfurous compounds by up to 100% from landfill sites.

To reduce VOCs emissions from composting facilities, composting process need to be conducted at neutral or alkaline pH values (Sundberg et al. 2013), alternatively biofilter can be used to remove VOCs and NH_3 from the exhaust gases during organic waste composting process (Li et al. 2013; Pagans et al. 2007).

Instead, regarding odor control strategies to reduce odors from concentrated animal feeding operations, livestock housing, manure storage facilities, and during land application, they have been studied since the end of the 1900s (Kreis 1978; Powers 1999). Indeed, a wide range of mitigation techniques is available, such as nutritional strategies, manure additives, building design, air filtration, manure covers, and treatment systems (Bibbiani and Russo 2012; Ubeda et al. 2013).

Modified animal feeding can decrease odors. Since proteins and fermentable carbohydrates are the main precursors of odor formation, it has been demonstrated that altering their level can affect odor emissions (Le et al. 2005). In their study, Hayes et al. (2004) found a 30% reduction of odor emissions in case of 160 and 130 g/kg crude protein diets. Moreover, reducing crude protein can effectively reduce excreted nitrogen, which is associated with lower NH_3 emissions (Ubeda et al. 2013). Similar results can also be obtained increasing the fermentable carbohydrates in diets. Indeed, they affect bacterial proliferation in both the gastrointestinal tract and in the manure modifying N excretion mechanism thus reducing NH_3 and odor emissions (Le et al. 2005).

As regards manure additives, the most common are: digestive additives, disinfecting additives, oxidizing agents, adsorbents, and masking agents (McCrory and Hobbs 2001). However, at the moment, the main disadvantages of using additives are that they provide a short-term efficacy and they require frequent reapplication (McCrory and Hobbs 2001; Wheeler et al. 2011). Alternatively, covering stored manure provides a physical barrier to reduce both NH_3 and odors emissions, by constructing rooftops or covering the surface with different materials to reduce the free surface of the slurry (Hörnig et al. 1999). Other manure treatments are based on solid-liquid separation alone (Zhang and Westerman 1997) or coupled with anaerobic digestion (Hansen et al. 2006; Hjorth et al. 2009). By reducing the dry matter content, anaerobic digestion degrades also some VOCs, particularly volatile fatty acids, which can be reduced by between 79% and 97% (Hansen et al. 2006).

Regarding air filtration, bioscrubbers and biofilters can be used for reducing the emission of odors and VOCs to the atmosphere, thanks to the action of microorganisms that degrade gaseous contaminants (Sheridan et al. 2002). In short, the exhaust air is injected in these devices, pass through a biologically enriched layer where microorganisms use the organic matter as feed, thus letting clean air out (Pagans et al. 2005). Another straightforward way to reduce emissions is to install air scrubbers at the ventilation outlets of livestock buildings. Scrubbers consist mainly of three parts, at the bottom is located the buffer tank, in the middle the packing material and on the top, there are spray nozzles and air outlet nozzle. NH_3, odorous compounds, and dust are trapped by the packing material (Van der Heyden et al. 2015).

In conclusion, odor emission reduction can be obtained thanks to the application of one or more of the strategies reported above. Although these strategies have been longer investigated and are well known, further research is needed to explore efficient and cost-effective management systems other than to ensure effective implementation at the farm level.

5.5.2 Odor impact in Life Cycle Assessment

The Life Cycle Assessment (LCA) is holistic approach to evaluate the potential environmental impact of production processes. Although originally developed for industrial processes, in the last years, it is more and more applied also to agricultural systems. Nowadays, LCA is internationally recognized as a viable and consistent approach for the environmental impact assessment. Nevertheless, among the different environmental impacts that can be quantify with LCA the odor impact is missing. Recently some preliminary approaches to include odor impact in the LCA framework were proposed (Cadena et al., 2018; Peters et al., 2014).

In particular, Cadena et al. (2018) presented an indicator for odor impact potential (named Odor Impact Potential, OIP) that can be applied to different activities and processes. In the study, OIP is applied to an anaerobic digestion plant considering that anaerobic digestion has been recognized as an effective solution to reduced odor impact from animal slurry storage (Lim et al., 2003; Fusi et al., 2016). OIP aims to include odor-derived impact in LCA studies by combining (and not replacing) parameters such as odor emission rates, odor concentration, or odor emission factors. OIP expresses the amount of air needed to dilute the odor emission below a concentration not detectable by human panelists in olfactometry. According to the LCA approach, OIP should be referred to the functional unit (i.e., the reference to which the inputs and outputs and the potential environmental impact can be related).

Based on the previous experience of OIP proposed by Cadena et al (2018), odor could be introduced in the LCA framework with an additional impact category able to quantify the potential odor impact. More in details, using olfactometry the different odor sources could be quantified and, at the end, summed up to calculate the potential olfactometryc unit (OU) related to a specific production process. Even if OU is quantified in laboratory and doesn't account the dispersion models, the developed odor impact category could be useful to compare different production processes that provide the same function and are responsible of odor annoyance. As for other impact categories such as Climate Change, Ozone depletion, Human toxicity, the odor impact category will consider (and sum up) emissions geographically located in different areas; this issue should be considered in the last step of LCA "Results interpretation". From a practical point of view, the first step to set up the odor impact category is the building, using olfactometry, of a database with the different odor emission sources.

5.6 Conclusions

The release of odorous components, from waste treatment plant and husbandry practices, is a concern for the people living near these facilities, other than for the health of workers and animals. A literature review of odor measurements and of a major source of complaints was carried out to point out the state of the art of this concern with regard to the odor from waste management and from livestock activity.

The literature reviewed highlighted that, over the last century, wide attention has been paid to "odor issue" in different countries. If in Asia, and in China in particular, it is mainly focused on odor from waste management plants in other countries some researchers investigated the issue from a livestock point of view. Considering that the impact of odors on the surrounding communities depends on several factors (from climatic conditions to technological aspects) some general conclusion can be drawn regarding the evaluation of odors as well as about possible mitigation solutions and future research activities.

Sensorial or analytical techniques can be used, in alternative or in combination, for quantitative and qualitative characterization of odors. The main advantage of sensorial techniques is the higher sensitivity of human nose compared to electronic instruments, while the main disadvantage is that the panel should be composed of qualified assessors to ensure reliable and repeatable results. Instead, analytical methodologies are not subjected to human errors, but are less sensitive, non-reliable in case of mixture of many odorants at very low odor concentration and they are not able to detect the interaction between odorants. The goal of both techniques is the evaluation of odor concentration.

However, to simulate odor dispersion, the evaluation of odor concentration alone is not sufficient, as also the air flow associated with the monitored odor source have to be taken into account. For determining odor plume extents, and therefore evaluating odor exposure at receptors, different air dispersion models, that applied different simulation procedures, can be used. Models follow the Gaussian, Lagrangian or Eulerian approaches. To be performed they require topographic/orographic, meteorological and emission data, which goodness affect the quality of the results. Air dispersion models are a useful tool when evaluating technologies to reduce the odor release at existing operations or when planning new operations, thus having not only a descriptive nature but also predictive. Based on the model applied and underestimation or an overestimation can be obtained. So, it could be useful combining with sensorial and analytical techniques. Concerning the livestock sector, specific odor dispersion models should be developed to include all the specific features of livestock odor dispersion (e.g. short distance of transportation, multiple sources, animal mass, and number). OERs are usually estimated as a yearly constant value obtained by multiplying the average live mass of the animals by a constant OEF. However, in this way, some factors (e.g. live mass, variation of indoor air temperature, ventilation rate) that influence the odor release are not considered. So, using a model that takes into account a time series of OERs allows obtaining a better description of OEF and consequently a more realistic emission scenario. This scenario will be useful to improve the reliability of the calculation of setback distances.

Separation distances are just one possibility to protect people from odor annoyance, but a wide range of mitigation techniques are available and applied. Some solutions have been longer investigated and are well known (e.g. scrubber, additives, manure covers, etc.), while for others (e.g. activated carbon adsorption and activated sludge diffusion) further research is needed to explore efficient and cost-effective management systems other than to ensure effective implementation. To minimize the nuisance of waste treatment plants a strategy is represented by composting which need to be conducted at neutral or alkaline pH values. Regarding intensive livestock farming, the common mitigation techniques adopted are: nutritional strategies, manure additives, building design, air filtration, manure covers, and treatment systems, whose efficacy increases by combining them.

In conclusion, since odors have such a big impact on the surrounding environment, it would be useful to quantify the nuisance also from an environmental point of view. Thus, it is reasonable to integrate odors as an indicator to be used in a life cycle assessment framework, and consequently, to develop a specific impact category quantifying the odorous impact related to the life cycle of a product or a process. In fact, up to now, although the different compounds responsible of the odor (e.g. NH_3, VOCs) affect some impact categories (e.g. eutrophication, acidification, and particular matter formation) an impact category specifically referred to the odor is missing.

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CHAPTER 5

This chapter consists of three published peer reviewed articles (*Paper 4, 5 and 6*). The aim of this chapter is to describe abatement efficiency and environmental benefits achievable by adopting mitigation strategies. The LCA method was used to assess the environmental impact. In particular, the first two articles focus on air treatment technologies installed in pig barns, whereas the third one on the application of an additive made of gypsum during cattle slurry storage. Each solution presents advantages and disadvantages, fully described in the following papers.

6. Evaluation of a Wet Acid Scrubber and Dry Filter Abatement Technologies in Pig Barns by Dynamic Olfactometry

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Abstract

Livestock activities, in particular swine farms, are sources of odorant compounds that cause conflicts with the neighboring population. Beside the effects on the neighborhood, excessive odor emission can cause discomfort to farm workers. In this context the APPROAch project, aims to test the application of two different air cleaning technologies (a wet acid scrubber and a dry filter) to reduce dust, ammonia and odors, in naturally ventilated pig facilities. The aim of the present study is to evaluate, in a pig farm, the odor removal efficiency of the two tested abatement technologies, based on air samples analyzed by dynamic olfactometry. Odor sampling was carried out at a pig facility involved in the project and brought to the lab within 30 h from sampling, as established by the European Standard EN 13725:2004. Odor concentration was evaluated by dynamic olfactometry using an Olfaktomat-n 6 (PRA-Odournet B.V.—Amsterdam, The Netherlands). The results show that the wet acid scrubber prototype presents an average odor removal efficiency of 16%, whereas dry filter has from limited to no effect. This efficiency could be considered as a good result for a prototype even if further analysis, with longer sampling periods are needed.

Keywords: Odor; Pig farming; Wet acid scrubber; Dry filter; Dynamic olfactometry

6.1 Introduction

The expansion of residential centers has led to some problems of coexistence between the population and the production activities located in the same area [1]. Livestock activities or husbandries, in particular swine, dairy and poultry farms, are sources of odorant compounds that cause conflicts with the neighboring population [2,3]. The most common problem caused by odor issues is nuisance [4]. Moreover, extended exposure to malodors leads to a reduced quality of life and health [5]. In the agricultural sector, offensive odors are mainly generated by the incomplete anaerobic decomposition of manure and by manure management (storage, handling and land application) [6]. Ammonia (NH_3), hydrogen sulphide (H_2S) and volatile organic compounds (VOCs) are the principal odorous substances emitted by pig farms [7].

The compounds emitted from fattening swine houses vary, are complex and may result in odors nuisance. Representative VOCs emitted from swine facilities are mainly amines, sulfuric compounds, volatile fatty acids, phenolic compounds, carboxylic acids, esters, ketones, terpenes and aromatic compounds [8]. Few specific

VOCs have been identified to be the dominant odorants on swine farms such as sulfuric compounds and ammonia, which are often identified as the predominant components of all odor compounds [9,10]. Beside the effects on the neighborhood, excessive odor from swine facilities can cause discomfort to workers, leading to low labor efficiency [11] and reduced well-being [12].

In air, odors are usually transported by dust particles [13], so they can travel for long distances before being deposited to the ground surface. The numerous complaints from people working and living nearby farms made it necessary to adopt strategies to reduce odor nuisance. As an example, Germany, The Netherlands, Austria and other countries have established guidelines based on separation distances between livestock units and residential areas, for determination of odor-annoyance-free level [14,15]. Alternatively, strategies for reducing odor emissions from livestock activities can be implemented in farms and can be divided into two main lines of action: (i) “upstream”, aimed at reducing emissions before they are emitted; and (ii) “downstream”, aimed at containing emissions, once produced. As an example, nutritional strategies [16] and building design [17,18] fall into “upstream” category, whereas the application of covering systems [19], manure treatments [20,21] or air treatment technologies [22] fall under “downstream”. In particular, for what concerns air cleaning technologies, acid scrubbers, bio-scrubbers and biotrickling filters are considered effective techniques for odor removal in pig farms [23–25]. Biofilters, biotrickling filters, acid scrubbers and multi-stage air cleaning systems are also reported by Santonja et al. [13] in the JRC report entitled “Best Available Techniques (BAT) Reference Document for the Intensive Rearing of Poultry or Pigs”. In particular, for pig farms with a forced ventilation system, odor removal is reported to be, on average, 30% for acid scrubbers, 45% for biotrickling filters and 60% for bioscrubber.

The adoption of measures for the reduction and mitigation of odor emissions not only allows conflicts between the residents of the neighboring areas and farmers to be reduced [6], but it also allows the air quality inside sheds to be improved, with positive effects on animal and workers [11].

The chemical-analytical techniques allow the identification and quantification of the odorous compounds, but to quantify the response (the perception of odor), sensory techniques are required [26]. Dynamic olfactometry, a sensorial technique, is the only internationally accepted method for measuring odor concentration [27]. It is regulated by the European standard EN 13725 [28] and it is based on the use of human panelists and an olfactometer. It is commonly used to assess the abatement efficiency of air treatment technologies [29,30]. The main advantage of this technique is the high sensitivity of the human nose [26]; moreover, it provides results that can be used as input data for dispersion modelling [31].

The APPROAch project, financed by Regione Lombardia through the European Agricultural Fund for Rural Development (EAFRD), aims to test the application of two different abatement technologies, a wet acid scrubber prototype using a citric acid solution and a commercial dry filter used in dusty industrial processes. The goal of the project is to reduce NH_3, dust and odor emissions in pig fattening barns, the buildings mostly affected by poor air quality.

Therefore, the aim of the present study is to evaluate, in a pig farm, the odor abatement efficiency of two different abatement technologies (a dry filter and a wet acid scrubber) based on air samples analyzed by dynamic olfactometry.

6.2 Materials and Methods

6.2.1 Animal Housing and Conditions

The measurements about the odor abatement efficiency of a dry filter and a wet acid scrubber were carried out in an intensive swine facility located in the province of Brescia (Italy) producing heavy pigs (> 160 kg) for traditional dry-cured hams.

More specifically, the pig barns were located in the Po Valley. The orientation of the farm is reported in Figure 1.

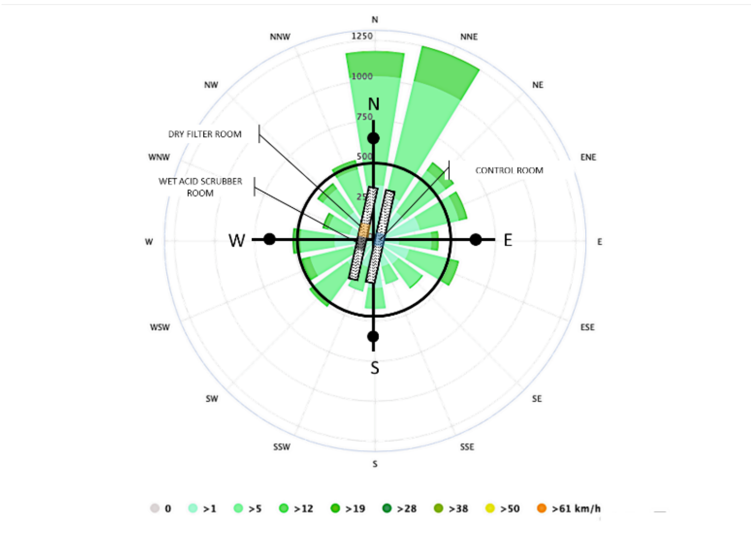

Figure 1. Orientation of the barns and prevailing winds.

The barns were naturally ventilated. Animals were fed twice a day with liquid feed (morning and late afternoon) and had unlimited access to water. The trial was performed in three rooms (42.5 × 17 m) with the capacity of 640 pigs each (from 30 to 110 kg). Two different abatement systems were tested, in the first room a dry filter (DF), whereas in the second one a wet acid scrubber (WAS) was installed and both were compared to a control room (CR). The rooms were divided into 32 boxes (8.5 × 2.5 m each) and animals housed on a fully slatted floor. The manure was collected under the flooring surface and in a pit equipped with the vacuum system.

The rooms equipped with the two different abatement systems presented the following characteristics: 32 windows, 2 doors and the inner volume of each room was around 3307.5 m^3.

During odor bags sampling collection, the ambient conditions (temperature and humidity) in the three experimental rooms were recorded.

6.2.2 Abatement Technologies

A dry filter system was provided by Zehnder Group USA (Buffalo, NY, USA), a company specialized in air treatment technologies and it was installed in the first room. It is a technology already used for the air treatment of industrial environments, such as bakery. The air is conveyed by a ventilation system through a series of filters that retain dust of different particle sizes. The filters are made of polypropylene fibers that use electrostatic charges to capture dust particles. The clean air is then returned to the barn by a blower. The operating principle of the dry filter is based on the interposition of serial filtering panels between the dusty zone and the clean zone. This arrangement, in addition to ensuring a remarkable separation capacity, allows filters to retain large amounts of dust. Usually, the dirty area is that in which the effluent enters the filter, while the clean one is downstream of the cells.

The maximum airflow rate was 6000 m^3 h^{-1} and the operating voltage was 320 W. The dry filter was sized 800 × 1390 × 1084 mm and it was installed in the middle of the barn, hanging from the ceiling 4 m above the ground.

Wet acid scrubber is a technology already tested in pig farms with forced ventilation [32]. The scrubber involved in this study is a prototype. It presents two tanks of 50 L capacity each, the first one is filled with water, the second one with a citric acid solution (15% of citric acid). Passing through the tanks, odorous compounds, dust and NH_3 are trapped by the packing materials. The intensive contact between the air and sprayed liquid enables soluble pollutants to pass from gas to the liquid phase. Thus, the air gets withdrawn from the pigsty, it gets washed thanks to the passage through the two tanks and it is finally, returned to the shelter. With respect to sulfuric acid, commonly used in air scrubber systems, citric acid presents the advantage of being safer to handle and harmless for pigs and workers [33]. The prototype was installed in the middle of the room at 4.8 m above the ground, to fulfill the idea to collect and release the cleaned air directly inside the barn, differently from what normally happens in forced ventilation system. In this latter case, treated air is released outside the piggeries, to reduce odor and NH_3 emissions in the environment and avoid problems with residents located in the neighboring areas [23]. The maximum air flow rate of the prototype was 6700 m^3 h^{-1}, with a 4 Kw fan and two pumps with the capacity of 0.75 Kw.

6.2.3 Odor Sampling and Dynamic Olfactometry Analysis

Odor sampling was carried out at the pig facility. For each of the 5 sampling trials, in DF, WAS and CR rooms, 5 air samples were collected in the middle of the CR or in the proximity of air cleaning systems to test their abatement efficiency. To perform the analysis, air samples were collected in 30 L Nalophan® bags equipped with a Teflon™ inlet tube by means of a vacuum pump. Each sampling lasted 3 min. In Figure 2 is provided a photo of the sampling system.

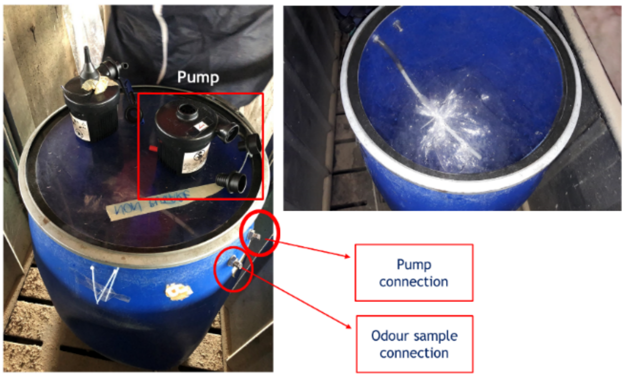

Figure 2. Sampling system.

The sampling period lasted for three months, from September to November 2020, details of performed activities are reported in Table 1.

Table 1. Details of performed activities.

Trial	Date	Description of Activities
1	7 September	Sampling on farm in WAS and CR
	8 September	Dynamic olfactometry analysis
	9 September	Sampling on farm DF and CR
	10 September	Dynamic olfactometry analysis
2	15 September	Sampling on farm in WAS and CR
	16 September	Dynamic olfactometry analysis
	17 September	Sampling on farm DF and CR
	17 September afternoon	Dynamic olfactometry analysis
3	17 September	Sampling on farm in WAS and CR
	18 September	Dynamic olfactometry analysis
	21 September	Sampling on farm DF and CR
	22 September	Dynamic olfactometry analysis
4	29 October	Sampling on farm in WAS and CR
	30 October	Dynamic olfactometry analysis
	2 November	Sampling on farm DF and CR
	2 November afternoon	Dynamic olfactometry analysis
5	24 November	Sampling on farm WAS, DF and CR
	24 November afternoon	Dynamic olfactometry analysis
	25 November	Dynamic olfactometry analysis

Odor samples from treatments and control rooms were collected in different dates, to avoid an excessive number of odor samples need to be analyzed in dynamic olfactometry laboratory.

After sampling, the bags were protected from light and analyzed within the 30 h maximum storage time, to reduce the risk of sample modification, as established by the European Standard EN 13725:2004. A total of 100 bags of air were analyzed out by dynamic olfactometry to measure the odor concentration. In the laboratory, samples bags were connected to the olfactometer, an instrument which dilutes the samples according to given ratios with reference air, which is made odor- and humidity-free through filtration with silica gel. Then, it presents odor, at the different concentration levels, to a group of evaluators. According to the European Standard EN 13725:2004, four qualified panelists were selected for the olfactometric analysis. The outcome is the odor concentration of the sample, which is expressed in European odor units per cubic meter (ouE m^{-3}). This represents the number of dilutions with neutral (odorless) air needed for the sample to reach its odor detection threshold concentration. The odor detection threshold corresponds to the level at which the odor is perceived by 50% of the panelists. Only people with an average sensitivity to *n*-butanol within the range of 20–80 ppb can be selected as panelists [28]. In order to ensure reliable and repeatable results, the EN 13725:2004 fixes precise criteria for the standard deviation of the individual's responses, which should be verified periodically. For this case study, the panel was tested with *n*-butanol at the beginning of each session, to fulfill the ISO EN 13725:2004 requirements of precision under repeatability conditions (r) and accuracy (A_{od}). The assessors were the same for all dynamic olfactometry sessions to avoid bias linked to variations in panel members composition.

In total analyses were performed in 5 trials. The subdivision in 10 olfactometric sessions was foreseen to solve the problem linked to the maximum sample limit of the dynamic olfactometry lab. Indeed, due anti-COVID procedures, panelists and the permanence in the lab were reduced to the minimum. Thus, sampling and analysis were performed according to the following scheme:

- Sampling on farm in WAS and CR;
- Dynamic olfactometry analysis;
- Sampling on farm DF and CR;
- Dynamic olfactometry analysis.

The olfactometer applied for the laboratory analysis was an Olfaktomat-n 6 (PRA-Odournet B.V.—Amsterdam, The Netherlands). The operative method used was the forced choice method; the olfactometer presents more active ports and the odor goes out only from one of them. Role of the panelists was to smell the air and to identify the port containing an odorant and to indicate whether their choice was a guess, inkling or certain on a tablet. Panelist choices were then collected to the Olfaktomat-n 6 software and presentations of dilutions as well as odor concentrations calculated according to EN 13725.

Olfactometric analysis consisted in two repetitions for each sample. This means that for each bag two measurements were executed. So, each odor concentration value was calculated as the geometric average of 2 repeated measures. The final total number of valid samples was 84 (removing outliers). The reduction of the number of samples was linked to damages occurred to Nalophan® bags during transport and to invalid measures. A measure was considered invalid when an assessor recognized the odor earlier or later than the other panelists.

6.3 Results and Discussion

6.3.1 Environmental Parameters

Table 2 shows the outdoors weather conditions during the sample collection trials, in particular referring to temperature (T), humidity (RH), wind speed and direction. These data were retrieved from the regional environmental protection agency's database (ARPA Lombardia, 2020).

Table 2. Mean meteorological data.

Sampling Date	T, °C	RH, %	Wind Speed, m/s	Wind Direction
7 September	22	84	1.78	SSW
9 September	23	80	1.43	E
15 September	24	72	1.61	E
17 September	24	70	1.26	S
21 September	19	96	1.55	SSE
29 October	11	95	1.12	WSW
2 November	11	100	0.70	S
24 November	4	89	1.21	WSW

Table 3 summarizes the average temperature (T), relative humidity (RH) and windows conditions in the three experimental rooms during sampling.

Table 3. Average environmental conditions inside the barns.

Sampling Date	T, °C	RH, %	Windows
7 September	20.1	81.1	Open 70%
9 September	20.6	77.2	Open 90%
15 September	21.2	73.0	Open 80%
17 September	21.1	72.8	Open 60%
21 September	18.7	84.6	Open 80%
29 October	17.2	84.1	Open 10%
2 November	19.3	81.0	Open 5%
24 November	10.7	83.2	Open 50%

In the farm considered, windows opening was automatized as they started opening when inside temperature and/or relative humidity (RH) was higher than 20°C and 80%, respectively.

Despite a large variation of outdoor temperature, the indoor temperature remained in a narrow interval, ranging from 21.2°C to 16.5°C, except for the 24th November, when slurry pits were emptied and windows were manually opened, despite the outside temperature (+4°C), to preserve animals and operators welfare.

6.3.2 Olfactometric Measurements

In Table 4 the average odor concentrations (ouE m^{-3}) ± standard deviation, for each session, measured by olfactometric analysis for DF, WAS and CR, are presented. In order to satisfy the requirements of EN 13725, the panel was tested using *n*-butanol for accuracy and repeatability before each olfactometric session.

Accuracy (A_{od}) ranged from 0.098 to 0.160 and repeatability (r) ranged from 0.268 to 0.322, with a 95% confidence interval, confirming the panel selection criteria.

Table 4. Average ± standard deviation odor concentrations (ouE m^{-3}) for each abatement system and for the control room.

Session	Wet Acid Scrubber	Dry Filter	Control
1 (7–10 September 2020)	1102 ± 175	2285 ± 446	3260 ±1168
2 (15–17 September 2020)	2617 ± 562	11,220 ± 1490	10,131 ± 1572
3 (17–22 September 2020)	8287 ± 2064	10,001 ± 567	10,131 ± 1572
4 (29 October–2 November 2020)	34,123 ± 4317	38,339 ± 10,078	34,112 ± 3860
5 (24–25 November 2020)	104,329 ± 9112	163,478 ± 36,340	57,911 ± 23,101

As expected, odor concentration increases during time in correspondence of an increasing animal live weight. Pig weight at the beginning of the cycle was 28 kg and the average daily gain was 0.93 kg. November sessions odor concentration measured by dynamic olfactometry was higher than September ones.

Generally, WAS registered lowest values compared to DF and CR, except for the last session. The 24th of November, slurry pits in WAS and DF rooms were emptied during collection of air samples, substantially increasing the ouE m^{-3} measured. Nevertheless, WAS always presents lower odor concentration values than DF room and, excluding session 5, also lower than CR.

Results show a detected odor concentration for all analyzed samples between 1102–163,478 ouE m^{-3}. Environmental conditions inside and outside the rooms could have influenced the final result. Although samples were collected always in the morning around 10 a.m., away from meals, to have repeatable test conditions, external variables, such as windows opening and pits emptying, certainly affected the results. In laboratory, uncertainties associated with odor quantification, were reduced using the same olfactometer and the same panel. Assessors were selected only using *n*-butanol as a standard reference odorant; although, as underlined by Hove et al. [34], this gas does not reflect the characteristics and intensities of odors associated with pig farming. Consequently, knowing the major odorous compounds associated to swine facilities, is crucial for developing odor control strategies. For this purpose, gas chromatography coupled to mass spectrometry (GC-MS) is frequently used to identify and quantify key odorous compounds. As suggested by Hove et al. and by Gralapp et al. [34,35], panelists should be trained also to recognize "pig odor". Although, in this case study, assessors were not trained to recognize the "pig odor", they demonstrated consistency in their responses as, during olfactometric sessions, they recognize it at the same dilution. Moreover, it can also be considered that each session was a training for the following one as panelists and odor were always the same for all the experimental periods.

Each room was compared to the others to evaluate the odor abatement efficiency (WAS Vs. DF, WAS Vs. CR and DF Vs. CR). In Table 5, the percentage reduction for each comparison is reported.

Table 5. Odor abatement efficiency comparison among the three rooms (WAS, DF and CR). WAS = Wet acid scrubber room; DF = dry filter room; CR = control room.

Session	WAS vs. DF	WAS vs. CR	DF vs. CR
1	−52%	−66%	−30%
2	−77%	−74%	+11%
3	−17%	−18%	−1%
4	−11%	0%	+12%
5	−36%	+80%[1]	+182%[1]

[1]pit evacuated.

The WAS system presents, for all session, higher odor abatement efficiency compared to DF system, ranging from −11% to −77%, with an average of −39%. In addition, compared to CR, WAS technology reduces the odor concentration by an average of about 16%, while excluding the session 5, when pits were emptied, this value increases to 53%. DF abatement system resulted to be not an effective strategy to reduce odor concentration, even if dust is a carrier of odorous compounds. Compared to CR it presents the highest removal efficiency of 30% in the first session and lower in the following ones. This can be explained by the fact that the propylene filters of the DF system were replaced in July and, therefore, their dust removal efficiency could be affected. Even if biofilters have been demonstrated to be the most effective end-of-pipe technique to reduce odors [9], the use of a DF technology could be an advantage because of the high maintenance requirements of biofilters. Biofilters need an accurate control of the medium moisture of the packing material is required for a correct functionality [36,37], other than of temperature and nutrients to guarantee the survival of the bacterial population [13]. Moreover, they have the disadvantage of producing N_2O emissions if nitrifying bacteria are present [22].

The concentration of odor was reduced by WAS technology. The removal of odor-carrying particles might be explained by capture of soluble compounds in the two tanks, such as N-containing compounds (e.g., NH_3). It is well known that wet and acid scrubber represent an effective strategy to reduce NH_3 emissions, thanks to the intensive contact between the air and liquid phase, NH_3 is trapped by the acid solution leading to the production of ammonium salt [13]. Considering that the trial was conducted in a naturally ventilated pig facility, an average of 16% odor removal efficiency could be considered a good result. Wet and acid scrubber are normally used in facilities provided with forced ventilation and removal efficiencies are calculated considering the difference in the odor concentration between the inlet and outlet air from the scrubber. An average odor removal of 27% for acid scrubbers in forced ventilation was reported by Melse and Ogink [25] and, Van der Heyden et al. [22], in their review, indicated removal efficiencies ranging from negative values up to 80%, for all types of air scrubbers. Variations are principally linked to the type of air scrubber and the method chosen for the analyses. In addition, Melse and Mol [23] highlighted that odor removal efficiency presents a large variation attributable to variations in the composition of the air not completely reflected by the olfactometric measurement, by the functioning of the filter itself or by the methodology used. Moreover, the use of citric acid instead of sulfuric acid could have affect the results. Further studies on citric acid abatement efficiency are necessary, as its application is poorly studied [21]. Finally, it should be considered that acid scrubbers are more effective at removing NH_3 than odors [38].

6.4 Conclusions

The odor abatement efficiency using two different abatement technologies (a dry filter and a wet acid scrubber) was evaluated with dynamic olfactometry methodology. The results show that the wet acid scrubber prototype presents an average odor removal efficiency of 16%, whereas dry filter has from limited to no effect, representing a promising achievement for the prototype and for its installation in naturally ventilated pig farms. The above-mentioned odor abatement efficiency could be considered as a good result for a prototype, even if further analysis with longer sampling periods is needed.

The aim of the present study was to determine the odor removal efficiency inside the barns, but in the future, it could be certainly interesting to evaluate the effects of abatement systems also in neighboring areas through the use of air dispersion models, in particular considering the well-known problem of conflicts with citizens living nearby livestock activities. In addition to air dispersion models, also field inspection could be applied to provide the characterization of odor exposure in a defined assessment area.

Since dynamic olfactometry is time-consuming, a discontinuous measurement method and it does not provide any information on quality, origin, offensiveness or intensity of a smell, a combined analytical-olfactory approach could be useful. Indeed, both gas chromatography and electronic nose could be applied, for this purpose, to identify which odorous compounds characterize pig facilities and how the abatement technologies can affect them.

In conclusion, there is not one best method for measuring odors. Each situation should be individually evaluated by considering the applicability and limitations of each method and the aim that needs to be reached. In most cases, an integrated approach combining different methods and techniques is the best solution to depict exhaustively the problem and define a good strategy for its management.

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7. Environmental impact of pig production affected by wet acid scrubber as mitigation technology

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Abstract

Ammonia (NH_3) is the most common air pollutant in pig farms, affecting animals and workers' health, and causing damages to ecosystems. Hence, there is a need to reduce NH_3 emissions. Many mitigation strategies can be applied to limit gaseous emissions, such as the application of air treatment technologies. In this study, the environmental impact of a typical Italian pig farm, adopting a wet acid scrubber to abate NH_3 emissions, was evaluated using the Life Cycle Assessment approach. 1 kg of live weight (LW) was selected as Functional Unit. Two scenarios were considered. The baseline scenario (BS) represents the situation as it is, while the alternative scenario (AS) a wet scrubber prototype (with 70% NH_3 removal efficiency) was adopted. For 8 of the 12 evaluated impact categories, AS shows the highest environmental impact, due to the scrubber construction and maintenance. However, it was the best for those impact categories most affected by NH_3. Observed reduction ranged from 10% (for acidification, TA, and terrestrial eutrophication, TE) to 0.4% (for marine eutrophication, ME). The climate change impact was 3.55 kg CO_2 eq kg^{-1} LW and 3.65 kg CO_2 eq kg^{-1} LW for BS and AS, respectively. For almost all impact categories, the consumable materials for wet scrubber operation represented around 85% of the total impact of the scrubber. The results of the sensitivity analysis showed that variation in NH_3 removal efficiency had the greatest effect on particulate matter formation, TA, and TE. The achieved results provide a first quantitative indication of the environmental benefits that can be achieved using wet acid scrubber in naturally ventilated pig facilities.

Keywords: Pig farm; Air quality; Ammonia; Wet acid scrubber; Environmental impact; Life cycle assessment

7.1 Introduction

Western Europe is characterised by medium to large-scale intensive pig farms. The European main pork meat producer countries are Germany (5.34 million t product mass), Spain (4.53 mil- lion t), France (2.18 million t), Poland (2.08 million t), Denmark (1.58 million t), the Netherlands (1.54 million t), Italy (1.47 million t) and Belgium (1.07 million t) (Pigmeat, 2020). Italy houses 8.4 million pigs on approximately 24,950 farms. The pig population is mainly concentrated in the northern part of the country, in particular in Lombardy (52%), Piedmont (14%), Emilia-Romagna (13%), and Veneto (9%) regions (ISMEA, 2019). The Italian pig sector presents a high degree of specialisation in favour of heavy pigs (over 110 kg, with a minimum slaughter weight of 160 kg) used for the traditional production of dry-cured hams (Bava et al., 2017). Currently, in Italy, there are 21 Protected Designations of Origin (PDO) for dry-cured hams and 18 Protected Geographical Indications (GPI) (e.g. Parma ham, San Daniele ham, speck, mortadella) (ISMEA, 2019).

Ammonia (NH_3), odours (VOCs, volatile organic compounds) (Schauberger et al., 2018), particulate matter (PM) (EEA, 2019a), and greenhouse gases (GHG), such as methane (CH_4) (Marszałek et al., 2018), are the most abundant pollutants emitted by pig farms. In the European Union, the agricultural sector is responsible for 92% of NH_3 emissions (EEA, 2018), mainly generated by animal manure (McIlroy et al., 2019) and inorganic N-fertilisers (Schauberger et al., 2018). In 2018, NH_3 emissions from the agriculture sector in Italy were 345 Gg (94.2%), of which 9.1% belonged to the pig sector category (ISPRA, 2020). NH_3 contributes to indirect emissions of nitrous oxide (N_2O) as well as to acid deposition and eutrophication, causing changes in biodiversity and ecosystem functioning (Kebreab et al., 2016, Schauberger et al., 2018). Moreover, NH_3 plays a significant role in the formation of particulate aerosols in the atmosphere. Secondary aerosols, which have diameters of less than 10 (PM_{10}) and 2.5 microns ($PM_{2.5}$), are formed in the atmosphere from chemical reactions involving mainly NH_3, nitrogen oxides (NOx), and sulphur dioxide (SO_2) (Behera et al., 2013; Hristov, 2011). This is a concern because fine PM is capable of penetrating deep into the alveolar region and entering the bloodstream, increasing the risk of cardiovascular and respiratory diseases, and thus having an adverse impact on human health (Dominici et al., 2006). In 2016, $PM_{2.5}$ concentration in Europe was estimated as contributing to more than 400,000 premature deaths (EEA, 2019b), whereas in the Po valley, it has been estimated that high PM levels lead to a reduction in life expectancy of about 36 months (Kiesewetter et al., 2015).

Regarding GHG, globally the pig sector contributes 9% to total emissions from the livestock sector. Among the main emission sources, feed production is the largest (48%), followed by manure storage and processing (27.4%), post-farm emissions from post-farm activities and transport (5.7%), and finally on-farm energy consumption (3.5%) (Gerber et al., 2013). N_2O is mainly emitted from manure management, whereas CH_4 comes from both the enteric fermentation and manure management (IPCC, 2019).

Finally, odours, besides being responsible for annoyance to nearby residents, can cause airway irritation (Conti et al., 2020) and respiratory diseases in farmers and agricultural workers (Maesano et al., 2019).

As pollutants from the livestock sector lead to many environmental problems, affecting the atmosphere, the neighbourhood, the health of both public and pig workers, as well as pig welfare, many mitigation strategies can be applied to limit gaseous emissions (Calvet et al., 2017; Burchill et al., 2019). GHG, NH_3, and VOCs reduction can be achieved by various methods, such as nutritional strategies (Andretta et al., 2018), frequent slurry removal (Hoffet al., 2006), use of closed slurry tanks or pits (Guarino et al., 2006), slurry injection techniques that permit to spread slurry into the soil through anchors and avoiding the superficial spreading with plates (Bacenetti et al., 2016), use of treatment systems (e.g. acidification, anaerobic digestion, solid-liquid separation, nitro-denitro, etc.) or use of additives during storage (Fangueiro et al., 2009; Finzi et al., 2019), and, finally, the application of air treatment technologies (Van der Heyden et al., 2015). Concerning air cleaning systems, they are mentioned in the Best Available Techniques (BAT) reference document for the intensive rearing of poultry or pigs (Santonja et al., 2017). According to Decision (EU) 2017/302 among BAT available for the intensive rearing of poultry or pigs, BAT 30, point c, refers specifically to air cleaning systems, such as biofilter, bioscrubber (or biotrickling filter), dry filter, two-stage or three-stage air cleaning system, water scrubber, water trap and wet acid scrubber (EU, 2017). In Northern European piggeries, the wet acid scrubber technology is the most widely applied (Costantini et al., 2020), in order to comply with regulations, such as the National Emission Ceilings and the EU Clean Air Pol- icy Package that limit the emission of NH_3, GHG and other pollutants (Jacobsen et al., 2019). The wet acid scrubber is an end-of-pipe technique, used in forced ventilated animal house, for removing pollutants from the exhaust air. The air gets withdrawn from the pigsties, washed, thanks to the passage through an inert packing material sprayed with an acid solution (usually sulphuric acid), and finally returned to the barns. The intensive contact between the air and liquid enables soluble pollutants to pass from gas to the liquid phase. In this way, NH_3 is captured by the acid solution, leading to the production of ammonium salt (Santonja et al., 2017). In the acid scrubber, most of the trickling water is recirculated, the other part is discharged and replaced by fresh water (Melse and Ogink, 2005). Compared to biotrickling filters, acid and wet scrubbers present higher NH_3 removal efficiency, less discharge water and higher nitrogen concentration into the water discharged (Costantini et al., 2020; De Vries and Melse, 2017). NH_3 removal efficiencies ranging from 70 to 99% are reported for wet acid scrubber using sulphuric acid solution, in mechanically ventilated pig housing facilities (Costantini et al., 2020; Van der Heyden et al., 2015). NH_3 removal efficiency depends on the pig barn structure, ventilation (e.g., natural or mechanical), and type of acid used. The removal efficiency is usually calculated considering the difference in the NH_3 concentration between the inlet and outlet air from the scrubber. In particular, higher NH_3 removal efficiency (>90%) is reported for wet acid scrubber using sulphuric acid solution and installed in mechanically ventilated facilities (Melse and Ogink, 2005; De Vries and Melse, 2017; Dumont, 2018) while lower efficiencies are recorded for devices using acids less strong than the sulphuric one (e.g., citric acid) and installed in naturally ventilated facilities (Starmans and Melse, 2011). When citric acid is used instead of sulfuric one, despite lower removal efficiency (around 70%), the management of the scrubber is easier being the citric acid safer to handle (Jamaludin et al., 2018).

For the other air cleaning technologies, installed in mechanically ventilated pig housing facilities, Melse and Ogink (2005) reported 70% NH_3 removal efficiency for biotrickling filters, Van der Heyden et al. (2015) an average of 64% for biological air scrubbers, Van der Heyden et al. (2016) 86% for biological air scrubbers with nitrification tank, and Dumont (2018) 70% for bioscrubbers.

The environmental impact of pig production systems using air treatment technologies remains poorly documented in the literature. De Vries and Melse (2017) compared the environmental impact of three different types of air scrubbers in piggeries (acid scrubber, biotrickling filter nitrification only, and biotrickling filter with nitrification and denitrification). They concluded that the scenario with acid scrubbers showed the lowest environmental impact in all impact categories and in particular had greatest effects on NH_3-related impact categories (i.e. acidification, particulate matter formation, and marine eutrophication). So, acid scrubbers have been suggested as the most appropriate technology for NH_3 emissions abatement at pig farms.

Scrubbers usage in pig farms located in North Europe countries, such as Belgium and Netherlands, is consolidated (Zhuang et al., 2019), and it represent a way to improve the environmental sustainability of pig farming, as they reduce NH_3 emissions and their related impact (De Vries and Melse, 2017). The environmental problems associated to NH_3 emissions are well known and described above. As it is mentioned that agriculture is the largest contributor to NH_3 emissions. Therefore, the treatment of NH_3 from intensive pig farms represents a crucial issue to ensure sustainability both in meat production and environmental protection. With regards to this last aspect, Costantini et al. (2020) explored the effect that the large-scale implementation of wet acid scrubbers in pig housing facilities could have in the European Union. They concluded that the abatement of NH_3, obtained by the application of wet acid scrubber, can reduce both the human health impact and environmental costs.

Even if the construction and maintenance of scrubbers involves the consumption of acid, energy, and materials, optimising their design and operation can facilitate the simultaneous reduction of other pollutants, such as odour, GHG and PM in an efficient and cost-effective manner (Van der Heyden et al., 2015), improving the environmental sustainability. Moreover, the ammonium citrate or ammonium sulphate formed can be used as a fertiliser for crop production (De Vries and Melse, 2017; Jamaludin et al., 2018) constituting an environmental credit by means of mineral fertilizer replacement and thus the avoidance of its production.

Finally, consumer behaviour towards a more sustainable pig production is increasing. The use of air cleaning technologies in pig farms can move to this direction.

Despite the numerous current and future benefits offered by air scrubbers in piggeries, they do not represent a consolidated method to reduce emissions and related impacts in southern European regions, such as Italy and Spain. In these countries, pigs, during the fattening phase, are usually housed in naturally ventilated buildings thanks to the warm climate (Aguilar et al., 2010; Estellés et al., 2009). However, in order to comply with current and future regulations, the implementation of air scrubbers is expected to expand in intensive livestock production areas across Europe.

This study aims to evaluate the environmental performance of a typical Italian pig rearing systems paying particular attention to the environmental effectiveness of a wet acid scrubber as a mitigation solution to reduce NH_3 emissions from livestock housing and the related environmental impacts. For this purpose, a pig farm in the Po valley area (province of Brescia), specialised in the production of heavy pigs, was evaluated considering two scenarios: with (Alternative) and without (Baseline) the adoption of a wet acid scrubber. The novelty of this study is to quantify the impact variation related to the implementation of an air treatment solution. For this purpose, the Life Cycle Assessment (LCA) approach was applied. Besides, this LCA study aims to identify the environmental processes with the greatest impact, how the impact varies between farms using or not using a wet acid scrubber, and margins for improvement.

To the authors' knowledge, this is the first LCA study considering the application of air scrubber for NH_3 abatement at Italian pig houses. Indeed, De Vries and Melse (2017) assess and compare the environmental impact of three types of air scrubbers in conventional Dutch housing system. Typical Dutch farms are mechanically ventilated and bring pigs to a slaughter weight around 105-110 kg. Whereas, Bava et al. (2017) and Pirlo et al. (2016) assessed the environmental impact of Italian heavy pig production without considering the implementation of air treatment technologies to abate NH_3 emissions.

7.2 Material and methods

This LCA study was carried out following the ISO Standards 14040 and 14044 (ISO, 2006; ISO, 2018).

7.2.1 Goal and scope definition

The goal of this study is to evaluate, for an intensive pig farm specialising in the production of heavy pigs and located in the province of Brescia (Po valley), the potential reduction in environmental impact linked to the installation of a wet acid scrubber.

The Po valley is an area, located in Northern Italy, characterized by the high concentration of agricultural and livestock activities. Due to the concurrent high density of anthropogenic sources and its orographic and meteorological characteristics it is characterised by the frequent occurrence of stagnant meteorological conditions, particularly unfavourable for pollutant dispersion.

The evaluated farm is representative of the intensive pig farming system characterising northern Italy (housing facilities with natural ventilation; low self-sufficiency for feed; production of heavy pigs) (Bava et al. 2017). Regarding the house facilities and in particular their ventilation, the conditions are similar to the ones characterising the pig rearing systems in other European countries such as Germany and Denmark (26% and 13% of the EU pig herd, respectively).

The outcomes of this study could be useful for farmers and their associations to understand the actual environmental impacts of the pig production process and the consequences and benefits arising from the reduction of NH_3 emissions. Moreover, they can be useful for companies and technicians working with farmers to offer innovative solutions. Finally, these preliminary results can provide useful information to those organisations directly involved in the development of policies and technical reference documents, in addition to being transferred to stakeholders interested in the topic.

7.2.2 Farm description

The analysed farm is in Orzinuovi (Brescia) (45°25'44" N 9°58'04" E) Lombardy, Italy. It is an intensive farrowing to finishing farm, which means that it produces piglets and raises them to market weight. In this case, heavy pigs for PDO dry-cured ham consortia are produced. The Utilised Agricultural Area (UAA) for cultivation is 100 ha, dedicated to maize cultivation and entirely utilised as feed for animals.

The animals are housed in an indoor system, with different specific conditions depending on their growth stage. During lactation, sows are kept in farrowing crates where they are confined between bars to reduce the risk of the sow crushing her new-born piglets. After 3 weeks, piglets are weaned and placed in a nursery, and the sow is returned to the gestation barn. Here, all females are artificially inseminated and remain housed in the gestating housing section for 100 days, the gestation period. After the piglets reach approximately 25-35 kg, they are placed in a growing-finishing barn where they remain until they reach 160 kg, which corresponds to 9 months (minimum live weight and age required by PDO regulation). Boars are used to collect semen for artificial insemination.

The pigs are housed in pens, in closed mechanically ventilated buildings during the farrowing phase, and in closed naturally ventilated buildings during the fattening one. Electrical heaters are used in farrowing and nursery houses for new-born piglets. All the pig production systems involve electricity consumption for illumination, feeding processes, and manure management. The feeding process also requires diesel fuel combustion for grinding, mixing, and pelleting operations.

Pig excreta are handled as slurry, mainly removed using a vacuum system, and then stored in ponds located outside the pig buildings where natural crust formation takes place. The slurry is spread with an umbilical system in accord with the Nitrate Directive (EU, 1991). Only maize grain is partially produced on-farm (1300 t $year^{-1}$ at commercial moisture of 14%) whereas all other feed ingredients are purchased, showing very low feed self-sufficiency (4% of total feed consumption, as fed). Regarding maize, the cultivation is carried out in irrigated fields and following the indications for integrated production (Negri et al., 2014). Water for drinking and cleaning water is taken from a well.

Two different scenarios were considered, Baseline (BS) and Alternative (AS). The BS represents the situation as it was recorded and described above. The AS represents the same situation but envisages the implementation of a wet acid scrubber technique in the fattening barns, which are the most affected by poor air quality (Dumont et al., 2014). Consequently, the zootechnical parameters were the same for both scenarios but in AS data related to wet acid scrubber construction and consumption were included. Regarding AS, the wet acid scrubber is installed inside the barn. It has two air treatment towers, the first tower, connected to the air inlet, is filled with water, while the second tower is filled with a citric acid solution and it is connected to the air outlet. Thus, the air gets withdrawn from the pigsty, it gets washed thanks to the passage through the two tanks, and it is finally returned to the shelter. Even if it is still at prototype stage, an investment cost of 22,000€ can be taken into account for the wet acid scrubber (excluding the installation costs).

The comparison between the environmental impact of the two scenarios considered highlights the effectiveness of wet acid scrubber as a mitigation solution in intensive pig housing facilities. The environmental trade-off among the different impact categories could occur; in fact, despite the reduction of NH_3 emissions, the adoption of wet acid scrubber technology involves the consumption of acid and energy for its functioning as well as of materials for its manufacturing and maintenance.

7.2.2.1 Functional unit and system boundary

In developing an LCA according to ISO 14044 (ISO, 2018), the Functional Unit (FU) must be clearly defined and measurable. FU describes the quantified performance of the function of the studied system and it provides a reference to which the inputs and outputs can be related, enabling comparison among different studies. In this study, the selected FU was 1 kg of pig mass, referred to as live weight (LW) at the farm gate, in accordance with the Food and Agriculture Organization (FAO) guidelines "Environmental performance of pig supply chain" (FAO, 2018).

Regarding the system boundary, considering that all the steps of production system subsequent to the fattening phase are not affected by the wet acid scrubber, a "*cradle to farm gate*" approach was adopted. Consequently, all on-farm activities related to crop cultivation and animal and slurry management were included, as well as all upstream off-farm activities, starting from raw material extraction, related to the production and supply of the inputs consumed. For crop intended for feed production, the following activities were considered: raw material extraction (e.g. fossil fuels and minerals), manufacture (e.g. seeds, fertilisers, and agricultural machines), use (e.g. diesel fuel consumption, engine exhaust gas emissions, and fertiliser-related emissions), maintenance and final disposal of machines. Feed additives, such as minerals, vitamins, and amino acids were included in the assessment. On-farm emissions of NH_3, N_2O, and CH_4 from enteric fermentation, slurry management (e.g. housing and storage), and field spreading were included within the system boundaries.

Conversely, the following aspects were excluded from the analysis:

- the production and maintenance of farm infrastructure (e.g. pig housing, slurry and manure storage, silos) because their lifespan is higher than 3-years and their impact was highlighted as negligible in a life cycle perspective (Lovarelli and Bacenetti, 2017),
- veterinary medicines and other farm chemicals (e.g. cleaning and disinfection), similar to Anestis et al. (2020) and Monteiro et al. (2019).

The soil organic carbon was considered to be in a steady-state for all arable land destined for annual crop production, except for the soybean share imported from South America, for which emissions related to direct land use changes (LUC) have been included. Since soybean is totally imported, LUC accounts for a large amount of CO_2 emissions in animal feed supply chain. For this reason, it is important to include this parameter in the evaluation, as reported by Bava et al. (2017).

Fig. 1 shows the system boundary for the two scenarios considered. No allocation procedure was applied because the farm sells only finished heavy pigs.

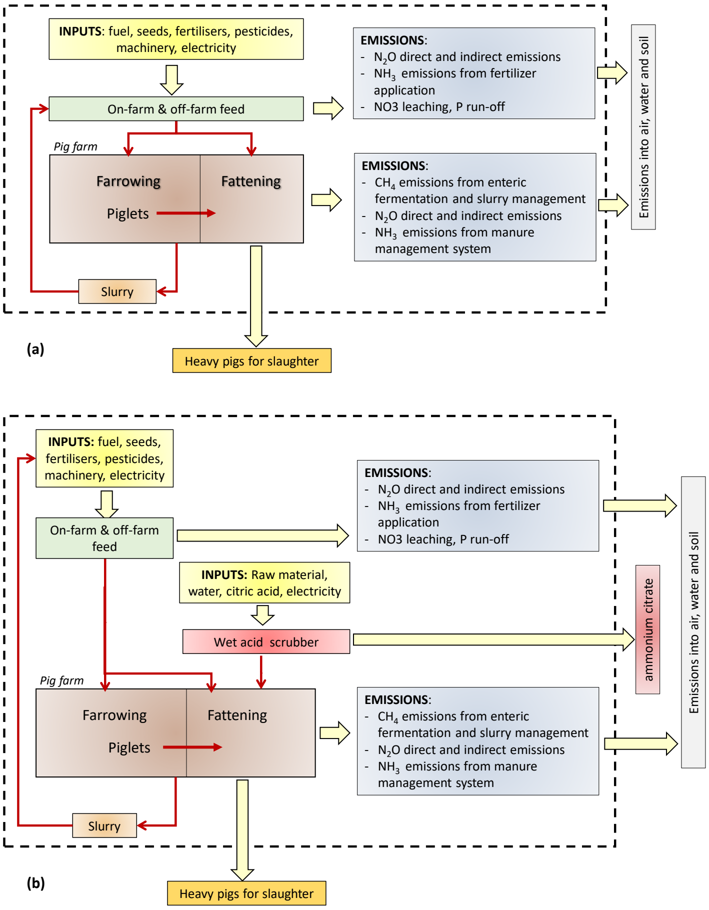

Fig 1. System boundaries for Baseline Scenario (a) and Alternative Scenario (b)

7.2.3 Life cycle inventory

Primary data concerning farm activities were collected from a questionnaire compiled by the farmer and through personal interviews with the farmer, including the following items: number of animals for each category, housing and ventilation system, slurry management, feed and diets, length and mass of animals in each sub-phase, electricity and fuel consumption. Specifically, the farmer provided all information regarding herd composition, crop production, animal diets, excreta management, cultivation practice and field operations, fertiliser, fuel, and electricity consumption. Concerning the diet supplied by the farmer, the formulation is mainly based on maize, wheat bran, soybean meal, soybean oil, fish meal, and mineral-amino acid-vitamin additive. However, feed material inclusion rates are confidential and therefore not shown. In AS, taking into consideration the adoption of wet acid scrubber using citric acid, a 70% reduction of NH_3 emissions was considered during animal housing, being the scrubber installed inside the barns. Moreover, according to Santonja et al. (2017) in some Western-European countries and regions, such as the Netherlands, Flanders, Germany, and Denmark this is the required minimum removal efficiency in pig houses. The inventory data related to scrubber energy, water, and acid consumption as well as raw materials and energy needed for the construction of the machinery were taken from the literature (Simpson et al., 2012; Van der Heyden et al., 2015; De Vries and Melse, 2017). According to Melse and Ogink (2005), it was assumed that the washing water is partially recirculated to reduce water consumption. Table 1 reports the main inventory data related to productive parameters adopted in the analysis of BS and AS. Table 2 reports inventory data related to wet acid scrubber used only for the AS. For wet acid scrubber a mass of 500 kg and 8 years of lifespan were considered.

Table 1. Average zootechnical data for the farm

	Piglets	Fatteners 1	Fatteners 2	Sows lactating	Sows nursery and dry period	Replacement males and females
LW at entering the stage, kg	7	31	80	180	160	40
LW at leaving the stage, kg	30	80	167	200	200	120
Duration of the stage, days	65	72	125	28	129	120
Feed intake, kg animal^{-1} day^{-1}	0.6	1.4	2.4	4	2.2	3.1
Diet CP content, % of dry matter	16.1	14.3	13.3	14.3	11.3	14.4
Daily weight gain, kg animal^{-1} day^{-1}	0.27-0.43	0.69-0.71	0.70	-	-	0.67
Feed consumption, kg year^{-1}	647,145	1,300,860	2,167,215	413,545	313,900	226,313
Feed Conversion Ratio, kg feed kg LW gain^{-1}	1.46	1.74	2.94	n.a.	n.a.	n.a.

LW = Live Weight; n.a. = not available

Table 2. Wet acid scrubber inventory data (expressed per kg of NH_3 removed)

Consumable	Unit	Amount
Water	dm^3	132
Citric acid	kg	5.67
Electricity	kWh	6.25

Regarding the emissions, the two main emission sources considered were:

- emissions related to maize cultivation including the nitrogen (N) and phosphorous (P) compounds released into water and air due to crop fertilisation (NH_3 volatilisation, nitrate leaching, denitrification, and phosphorous run-off) and the pollutants (dust, VOC, NMVOC, hydrocarbons, nitrogen oxides, etc.) in the exhaust gas emitted by tractor engines and due to diesel combustion;
- emissions related to livestock activities, including CH_4 emissions from enteric fermentation and slurry management, direct and indirect N_2O emissions, and NH_3 emissions from manure management system.

Field emissions of N and P compounds into the air, water, and soil were evaluated using the model EFE-So, (Estimation of Fertilisers Emissions-Software, available at http://www. sustainable-systems.org.uk/tools.php) (Fusi and Bacenetti, 2014), which assesses the NH_3, N_2O, nitrates (NO_3), and phosphate (PO_4) emissions taking into account soil type, climatic conditions and agricultural management operations, similarly to Brentrup et al. (2000) and Bacenetti et al. (2016). Volatilisation of NH_3 from the slurry application was assessed considering (i) air temperature, (ii) time between the application and rainfall or incorporation in the soil; (iii) infiltration rate, according to the fertiliser application circumstances (e.g. presence of crop residues on the soil). NH_3 emissions from mineral fertiliser applications were evaluated taking into account the type of fertiliser, climatic conditions, and soil properties (e.g. pH, texture). Finally, N_2O emissions were computed considering the emission factor proposed by the Intergovernmental Panel on Climate Change (IPCC) (IPCC, 2006). Pollutant emissions due to fuel combustion in the tractor engines were estimated according to Lovarelli and Bacenetti (2017), taking into account working time, the field shape, age of tractors, and related emissions stages. For the emissions related to livestock activities, CH_4 emissions from enteric fermentation and slurry management were estimated following the Tier 1 approach equations, as suggested by IPCC (2019). The N_2O direct and indirect emissions from manure management that occur during animal housing and slurry storages were estimated following the Tier 2 method proposed by IPCC (2019). The excreted nitrogen (N) was estimated by calculating N retention and N intake. For N intake, data about Dry Matter Intake (DMI) and Crude Protein (CP) percentage were provided by the farmer. The NH_3 emissions from the manure management system (housing and storage) were estimated using the European Environment Agency (EEA) Tier 2 approach (EEA, 2019a), on the basis of the total amount of N excreted by the animals. Further information on air emissions estimation can be found in Table 3.

Table 3. Inventory data used for the estimation of emissions from animal housing and manure management

Item	Value	Source
Pigs population	9760	Primary data
Diet DM content (average)	87%	Primary data
Average LW, piglets	23.5 kg	Primary data
Average LW, fatteners 1	40 kg	Primary data
Average LW, fatteners 2	103 kg	Primary data
Average LW, sows lactating	200 kg	Primary data
Average LW, sows nursery and dry period	180 kg	Primary data
Average LW, replacement males and females	80 kg	Primary data
CH_4 Enteric fermentation emission factor	1.5 kg [CH_4] head^{-1} yr^{-1}	Tier 1 IPCC, 2019
$VS_{rate(T,P)}$ – finishing swine	5.3 kg [VS] t^{-1} [live weight] day^{-1}	Tier 1 IPCC, 2019
$VS_{rate(T,P)}$ – breeding swine	2.4 kg [VS] t^{-1} [live weight] day^{-1}	Tier 1 IPCC, 2019
$EF_{T,S,P}$ emission factor for direct CH_4 emissions from manure management	111.6 g [CH_4] kg^{-1} [VS]	Tier 1 IPCC, 2019

Emission factor for direct N_2O emissions from manure management	0.005 kg [N_2O-N] kg^{-1} [Nitrogen excreted]	Tier 2 IPCC, 2019
Emission factor for indirect soil N_2O emissions due to nitrogen leaching and runoff from manure management	0.01 kg [N_2O-N] kg^{-1} [NH_3-N + NOx-N volatilised]	Tier 2 IPCC, 2019
EF_{hous_slurry} – finishing pigs	0.27 kg [NH_3-N] kg^{-1} [TAN excreted] $head^{-1}$	Tier 2, EEA 2019
EF_{hous_slurry} – sows and piglets	0.35 kg [NH_3-N] kg^{-1} [TAN excreted] $head^{-1}$	Tier 2, EEA 2019
$EF_{storage\_slurry\_NH3}$ – all animal category	0.11 kg [NH_3-N] kg^{-1} [TAN in storages] $head^{-1}$	Tier 2, EEA 2019

Background data regarding the production and supply of the inputs (i.e. feed additives and off-farm feeds including soybean meal and related LUC, diesel fuel, electricity, seeds, fertilisers, pesticides, and agricultural machinery) were obtained from the Ecoinvent Database v.3 (Weidema et al., 2013).

7.2.4 Life cycle impact assessment (LCIA)

The inventory data were transformed into potential environmental impacts using the characterisation factors defined by ILCD (International Reference Life Cycle Data System) midpoint method (ILCD, 2011). This method has been endorsed by the European Commission. For this study, 12 impact categories were evaluated:

- Climate Change (CC, kg CO_2 eq),
- Ozone Depletion (OD, kg CFC-11 eq),
- Particulate Matter Formation (PM, kg $PM_{2.5}$ eq),
- Human Toxicity–No Cancer Effect (HTnoc, CTUh),
- Human Toxicity–Cancer Effect (HTc, CTUh),
- Photochemical Ozone Formation (POF, kg NMVOC eq),
- Acidification (TA, mol H+ eq),
- Terrestrial Eutrophication (TE, mol N eq),
- Freshwater Eutrophication (FE, kg P eq),
- Marine Eutrophication (ME, kg N eq),
- Freshwater Ecotoxicity (FEx, CTUe),
- Mineral, Fossil and Renewable Resource Depletion (MFRD, kg Sb eq).

Besides these impact categories, also the Cumulative Energy Demand (CED, MJ) was evaluated to better explore the impact of the wet acid scrubber operationing on the energetic performance of the fattening system. Method to calculate Cumulative Energy Demand (CED), is based on higher heating values (HHV).

7.2.5 Sensitivity analysis

To explore the robustness of the environmental results achieved, a sensitivity analysis was carried out to investigate the influence of the wet scrubber abatement efficiency as well as a system expansion regarding the ammonium citrate produced by the ammonia abatement in the scrubber (due to the reaction with citric acid).

Variation in emission abatement was considered since, in naturally ventilated buildings, emissions are affected by ventilation differences. In cold seasons, windows are kept closed most of the time, which is not the case in warm seasons. In addition, it is known that temperature is positively correlated with NH_3 emissions (Vilarrasa-Noguéet al., 2020), thus in warm seasons NH_3 volatilisation is higher and it is more difficult to capture this gas in naturally ventilated buildings. Consequently, NH_3 removal efficiency of 80% has been assumed for cold seasons and 60% for warm seasons. Moreover, fluctuations in NH_3 removal efficiencies for acid scrubbers have been reported also by Melse and Ogink (2005) and Van der Heyden et al. (2015). The consumables used for the wet acid scrubber operation were varied accordingly.

Regarding the ammonium citrate produced by the NH_3 emission reduction process in the alternative scenario, no allocation and no system expansion were applied to consider this additional product. This was because, on this farm, the N requirements of the different crops are supplied by the pig slurry and because getting value from it as a fertiliser would involve storage and transport out of the farm. However, ammonium citrate can be considered as a mineral fertiliser, and taking into account that the nitrogen contained is in ammoniacal form, an efficiency of 100% can be assumed (1 kg of N in the ammonium citrate substitute 1 kg of N from mineral fertiliser) (Bacenetti et al., 2016b). To quantify the potential benefits related to the fertiliser value of this co-product, a systems expansion was applied, with the N contained in the ammonium citrate being assumed to substitute for an equal mass of N from mineral fertiliser.

7.2.6 Uncertainty analysis

Monte Carlo analysis is an important tool used in many LCA assessments to test the reliability and robustness of systems, structures or solutions (Lesage et al., 2018; Pexas et al., 2020; Bacenetti, 2019). This tool simulates a probable range of outcomes given a set of variable conditions and can be applied within a Life Cycle Inventory framework to capture parameter variability (Guo and Murphy, 2012). Thus, it is a technique employed to quantify variability and uncertainty using probability distributions.

In this study, a Monte Carlo approach, considering 1,000 iterations and a confidence interval of 95%, was applied for the quantification of potential uncertainties associated with data inputs in the model.

7.3 Results

The potentials environmental impacts of 1 kg of pig live weight at the farm gate for both scenarios are reported in Table 4. For 8 of the 12 evaluated impact categories, AS shows a higher impact than BS, due to the impact associated with the use of the wet scrubber. As a result of the reduction of NH_3 emissions in fattening barns (70%), AS has a lower impact for TE, TA, PM, and ME, which are the impact categories influenced by NH_3. This confirms the relevance of NH_3 emissions as an important source of acidification and eutrophication. For these impact categories, the impact for 1 kg of pig LW is lower in AS because the environmental benefits related to the reduction of ammonia emissions offsets the impact increase due to construction, maintenance, and operation of the scrubber. For the remaining impact categories (OD, HTnoc, HTc, POF, FE, FEx, and MFRD), AS is associated with an increase in the impact ranging from 0.9% (FEx) to 102% (MFRD). Indeed, the higher energy and resource consumption related to the construction, maintenance, and operation of the scrubber worsens the results for these impact categories. Although the reduction of NH_3 emissions slightly affects the formation of indirect N_2O, the construction, maintenance, and operation of the scrubber involve GHG emissions that offset the impact reduction related to lower N_2O emissions.

Table 4. Absolute environmental impact for the two scenarios (FU = 1 kg of pig LW; Δ = impact variation of AS compared to BS).

Impact category	Acronym	Unit	BS	AS	Δ
Climate change	CC	kg CO_2 eq	3.55	3.65	2.85%
Ozone depletion	OD	kg CFC-11 eq$\cdot 10^{-7}$	3.12	3.32	6.53%
Human toxicity, non-cancer effects	HTnoc	CTUh$\cdot 10^{-7}$	7.08	7.29	3.00%
Human toxicity, cancer effects	HTc	CTUh$\cdot 10^{-8}$	1.90	2.24	17.45%
Particulate matter	PM	g $PM_{2.5}$ eq	3.28	3.16	-3.62%
Photochemical ozone formation	POF	kg NMVOC eq$\cdot 10^{-2}$	1.08	1.13	4.66%
Acidification	TA	mol H+ eq	0.12	0.10	-10.16%
Terrestrial eutrophication	TE	mol N eq	0.51	0.46	-10.98%
Freshwater eutrophication	FE	kg P eq$\cdot 10^{-4}$	4.49	4.65	3.57%
Marine eutrophication	ME	kg N eq$\cdot 10^{-2}$	1.93	1.92	-0.36%
Freshwater ecotoxicity	FEx	CTUe	23.74	23.95	0.91%
Mineral, fossil & renewable resource depletion	MFRD	kg Sb eq$\cdot 10^{-5}$	2.42	4.88	101.84%

Fig. 2 shows the relative contributions to the overall environmental impact of the production factors and of the emissions sources for BS and AS, respectively.

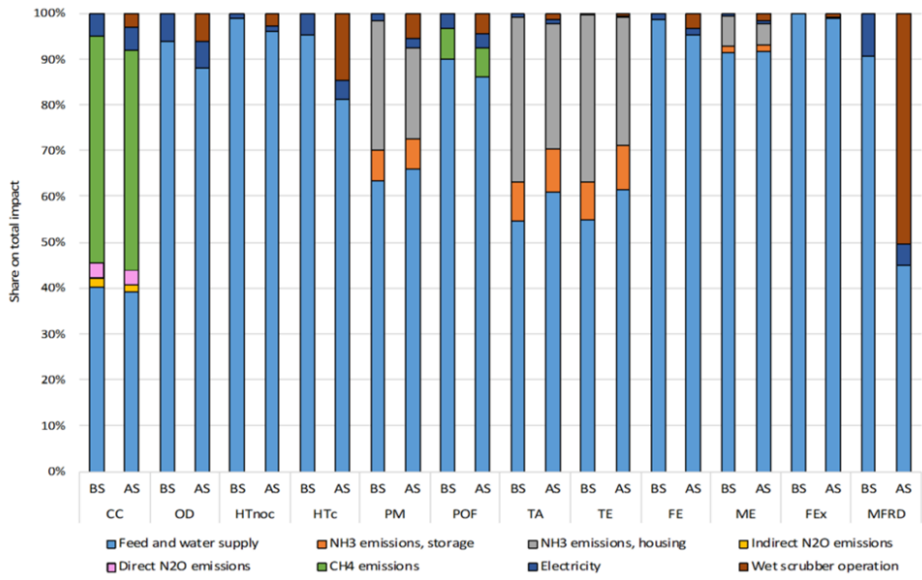

Fig. 2. Contribution of different inputs and outputs to environmental impact categories in BS and AS. BS: baseline scenario; AS: alternative scenario.

According to the results, for all the evaluated impact categories, feed is the most important contributor to the environmental impact of pigs: in BS its contribution ranges from 99% for HTnoc to 40% for CC. In particular, for CC, CH_4 emissions and feed purchase (due to the use of inputs such as fuel, machinery, fertiliser, pesticides, and transport) are the most significant processes.

In particular, CH_4 emissions account for 49.6% and 48.2%, and feed accounts for 40.3% and 39.1%, in BS and AS, respectively. Among feed ingredients, soybean and maize have been identified as the most significant environmental processes for CC, accounting for 50 and 40%, respectively. About 10% is attributable to electricity, and to direct and indirect N_2O emissions. In particular, electricity is responsible for about 5.0% and 4.8%, and N_2O emissions for 5.1% and 4.6%, in BS and AS, respectively.

Excluding feed, NH_3 emissions from storage and housing are mainly responsible for TA and TE. For TA, they range from 44% to 36%, in BS and AS, respectively; for TE, they account for 45% and 37%, in BS and AS, respectively. Finally, NH_3 emissions are also important for PM and ME. In particular, for PM they contribute about 34% and 26%, in BS and AS, respectively; whereas for ME they account for 8.1% and 6.1%, in BS and AS, respectively.

The abatement efficiency of the wet acid scrubber in AS leads to a reduction of impacts related to NH_3 emissions. Also, N_2O emissions show a slight reduction and result in a small decrease in CC impact (-0.01%), since indirect N_2O emissions are reduced. The contribution of the wet scrubber in AS to the environmental impact of 1 kg of pig LW at the farm gate registers the highest relative contribution for MFRD (50%) and the lowest for TE (0.6%), as shown in Fig. 2. For all impact categories, the consumable materials for scrubber operation (energy, citric acid, and water) represent around 93% of the total impact of the scrubber operation, except for HTc, in which it corresponds only to 57% with the remaining 43% of the impact related to its construction and electricity consumption, as shown in Fig. 3. Therefore, for all the evaluated impact categories, the production of citric acid is by far the main contributor followed by electricity and water supply. The latter has a negligible impact (<0.1%) for all the impact categories except for the human toxicity-related impact categories (about 0.5%).

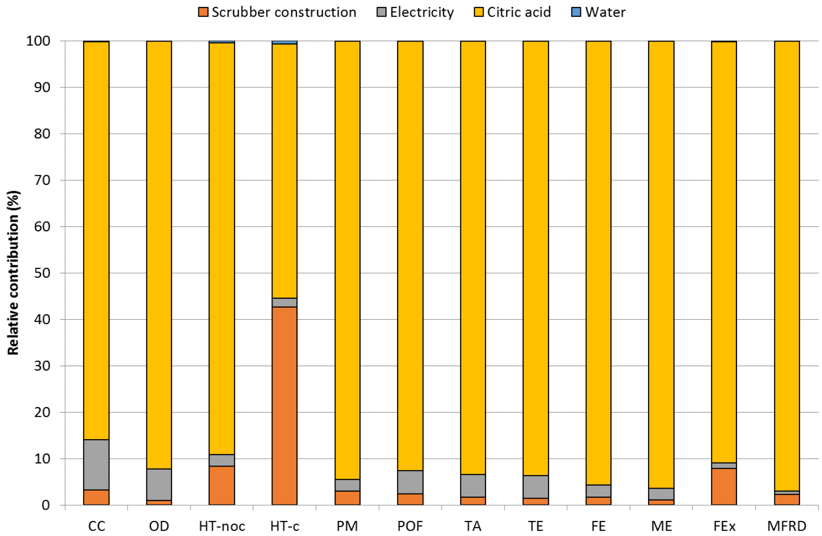

Fig. 3. Relative contribution for the wet acid scrubber operation.

Table 5 reports the results for CED. The comparison between the two scenarios shows that AS presents higher values compared to BS except for the impact category "Non-renewable, biomass". The differences, ranging from +5% to +17% are (as expected) related to the consumption of electricity.

Table 5. CED: results Comparison between BS and AS.

Impact category	Unit	BS	AS
Non-renewable, fossil	MJ	17.158	18.878
Non-renewable, nuclear	MJ	1.714	1.826
Non-renewable, biomass	MJ	2.673	2.673
Renewable, biomass	MJ	16.657	17.532
Renewable, wind, solar, geothe	MJ	0.187	0.213
Renewable, water	MJ	0.662	0.777

7.3.1 Sensitivity analysis results

Table 6 shows the impact variation for 1 kg of pig LW at the farm gate in the alternative scenario considering possible different levels of NH_3 emission abatement efficiency. As expected, the results show that, as NH_3 abatement efficiency increases, the impact for the categories related to its emissions (i.e. PM, TA, TE, and ME) is reduced, and vice versa. The variations for these impact categories reach a maximum of ±2.5% for TE. The remaining impact categories, on the other hand, show an opposite trend, due to the consumables for the scrubber operation which are greater as the abatement efficiency increases. Among these, the impact category showing the greatest variability is MFRD, which varied by ±4.0%.

Table 6. Sensitivity analysis results, expressed as percentage change in the impacts respect to the alternative scenario, in which 70% NH_3 abatement for the wet acid scrubber was considered.

	Ammonia abatement efficiency	
Impact category	**60%**	**80%**
Climate change	-0.31%	+0.31%
Ozone depletion	-0.66%	+0.66%
Human toxicity, non-cancer effects	-0.07%	+0.07%
Human toxicity, cancer effects	-1.59%	+1.59%
Particulate matter	+0.53%	-0.53%
Photochemical ozone formation	-0.57%	+0.57%
Acidification	+2.25%	-2.25%
Terrestrial eutrophication	+2.51%	-2.51%
Freshwater eutrophication	-1.13%	+1.13%
Marine eutrophication	+0.02%	-0.02%
Freshwater ecotoxicity	-1.20%	+1.20%
Mineral, fossil & renewable resource depletion	-4.04%	+4.04%

Regarding the system expansion applied to utilise the value of ammonium citrate as a mineral fertiliser, the sensitivity analysis highlighted a small impact variation. When the N in the ammonium citrate replaces the same amount of N fertiliser, the impact reduction for the alternative scenario ranges from 0.31% for TE to 4.05% for MFRD (with CC, PM, TA, FE, ME, and TE reduced by less of 1% and only HT-noc, FEx and MFRD by more than 2%).

7.3.2 Uncertainty analysis results

To test the robustness of the achieved results while comparing the two scenarios, a quantitative uncertainty analysis was carried out by using the Monte Carlo technique (1,000 iterations and a confidence interval of 95%) as a sampling method. The results are reported in Fig. 4.

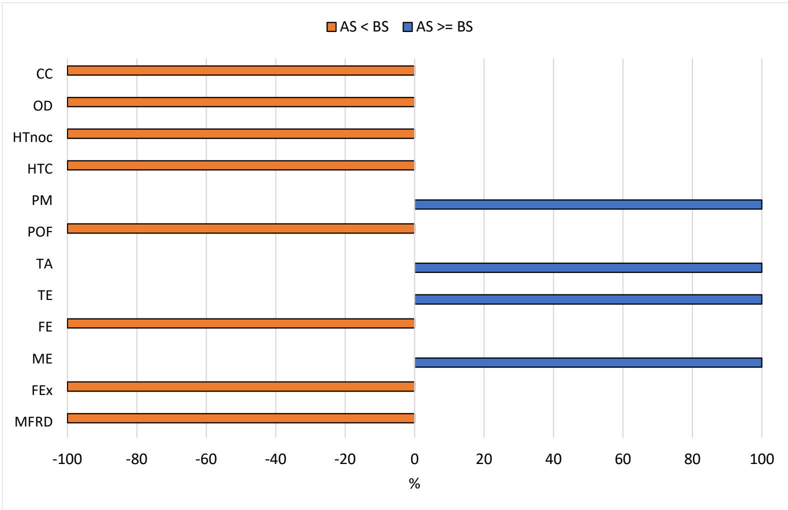

Fig 4. Uncertainty analysis results regarding the comparison between Baseline Scenario and Alternative Scenario.

The bars represent the probability that the environmental impact of BS is higher than or equal to the AS one, while those on the left represent the opposite probability. The uncertainty due to the selection of the data from databases, partial model adequacy, and variability of data does not significantly affect the comparison between baseline and alternative scenarios for all the evaluated impact categories.

7.4 Discussion

Previous LCA studies, carried out in different European countries, have found the CC indicator to range from 2.25 to 9.35 kg CO_2 eq kg^{-1} LW (Dourmad et al., 2014; Monteiro et al., 2019; Pirlo et al., 2016). In this study, CC was 3.55 to 3.65 kg CO_2 eq, in line with LCA studies carried out in Greece (Anestis et al., 2020), Italy (Bava et al., 2017; Pirlo et al., 2016), and Spain (González- García et al., 2015). Bava et al. (2017) assessed the environmental impact of 6 intensive pig farms located in Northern Italy, producing heavy pigs. They reported an average CC of 4.25 kg CO_2 eq kg^{-1} LW. Pirlo et al. (2016) obtained similar results (3.3 kg CO_2 eq) using an economic allocation. They considered both the breeding and growing-fattening phase of heavy pig production and showed that 70% of the environmental impact could be attributable to the growing-fattening phase. Monteiro et al. (2019) found different results for the environmental impact of 8 pig farms located in Italy, but this may have been affected by the use of a local pig breed, the lower number of fattening pigs considered, and the inclusion of grazing emission and soil carbon sequestration. Finally, Anestis et al. (2020) and González-García et al. (2015) assessed the impact related to the production of pigs. Although the LW of fattening pigs considered was lower (around 105 kg), the values in those studies are close to those reported here. The most influential subsystems in CC are GHG emissions and feed production, as also reported by Bava et al. (2017), Dourmad et al. (2014), González- García et al. (2015) and McAuliffe et al. (2016).

In this study, feed was identified as the most important contributor to the environmental impact of pig farming. This is in line with the results of many other studies (Bava et al., 2017; Pirlo et al., 2016; Reckmann et al., 2013). In particular, soybean meal and oil represent the main protein sources. The replacement of soybean sourced from South America (mainly Argentina and Brazil) with locally produced material could certainly affect the final impact, as the contribution from transportation distances (e.g. diesel) and relative emissions other than emissions related to land-use change drastically reduce (van Zanten et al., 2018). Alternatively, other protein sources could be introduced, such as peas, rapeseed meal, and sunflower, even if optimising nutrient-use efficiency is probably the most effective step (Eriksson et al., 2005; Monteiro et al., 2016). In this regard, Andretta et al. (2018) evaluate the potential environmental impact of Brazilian pig production using precision feeding systems during the growing-finishing phase instead of the conventional feeding system. They obtained lower environmental impact for the precision daily feeding by group and by individual programmes compared with the conventional feeding program (4% and 6% savings in potential climate change impact, respectively).

For the other impact categories, the results cannot be compared because of the different units of measurement related to the choice of different characterisation methods. In this study, results have been calculated according to LCIA methodology, whereas, among the works mentioned above, Bava et al. (2017), Monteiro et al. (2019), and Pirlo et al. (2016) performed their evaluations using CML Baseline method. Moreover, different methodological choices (e.g. functional unit selected, system boundaries, emissions inventory, allocation factor choice) significantly influence the impacts and a substantial difference in the environmental impacts occurs.

Regarding air scrubbers in piggeries, De Vries and Melse (2017) assessed the environmental impact of an acid scrubber, and two kinds of biotrickling filter (nitrification only, and with nitrification and denitrification). For the acid scrubber, with a 90% NH_3 removal efficiency, they found that CC was 5.31 kg CO_2 eq, for biotrickling filter with nitrification only and 70% of NH_3 removal efficiency it was 6.73 kg CO_2 eq, and for biotrickling filter with nitrification and denitrification and 70% NH_3 removal efficiency it accounted for 121 kg CO_2 eq. Unfortunately, these results are not comparable since they use as FU 1 kg [NH_3-N] entering the scrubber. However, they similarly observed that the greatest NH_3-abatement effects can be observed on TA, PM, and ME, confirming that acid scrubbers are an effective tool to reduce NH_3-related impacts. Also, the sensitivity analysis highlighted the effect of changing the abatement efficiency on NH_3-related impacts; the higher is the removal efficiency, the lower are PM, TA, TE, and ME impacts. It can be concluded that implementing wet acid scrubbers can effectively mitigate NH_3-related impacts from pig housing. However, considering that a wide range of mitigation techniques is available for reducing NH_3 (Finzi et al., 2019; Philippe et al., 2011) and GHG emissions (Marszałek et al., 2018) in pig production, there is a need for future studies that, by combining different mitigation strategies, identify the best farm design in order to reduce the releases both inside pig houses and outside (e.g., during manure storage and spreading).

Moreover, the improvement of air quality and environmental conditions inside piggeries will improve the health of workers and animals living in the barns (Cao et al., 2021; Costantini et al., 2020), leading to a reduction of the insurgence of respiratory dis- eases and to a better evaluation at slaughterhouse for what concern lungs score. Although farmers are not open to innovation, data proving a beneficial effect on animal performance and welfare (i.e. higher feed conversation rate; reduced respiratory problems) will help to persuade them to test air treatment solutions.

7.5 Conclusions

The present study reports preliminary results for the environmental impact of a farm producing heavy pigs where a wet acid scrubber for air treatment has been installed to reduce NH_3 emissions in pig barns naturally ventilated. Though feed is the main factor responsible for the environmental load, the use of a wet acid air scrubber leads to an impact reduction for all the impact categories influenced by NH_3 (TA, TE, PM, and ME). Though emission from pig barns only represents part of the NH_3 emission during pig rearing, the application of the wet acid scrubber is an effective strategy to reduce the environmental impact of heavy pigs for NH_3-related impact categories. However, at the same time, it worsens other impact categories, such as CC, OD, POF, toxicity-related impact categories, and MFRD. To reduce the environmental load for these latter impact categories, the adoption of mitigation strategies at the feed level is fundamental and more promising. To improve the environmental performance of scrubber, efforts could be made to reduce water and citric acid consumption by increasing their recirculation. At the same time, a further small impact reduction could arise by realising the fertiliser value of ammonium citrate salt (formed by the reaction between NH_3 and citric acid). The outcomes of this study can be upscaled to other European countries were pig rearing takes place mainly in naturally ventilated facilities.

Future research activities should focus on the development of a microclimatic tool able to continuously monitor the air quality inside barns, to allow automatic management of the activation of the abatement system so that NH_3 levels fall within established thresholds, thus reducing NH_3 emissions and minimising energy consumption associated with its operation. This new technology, when completely automated, would help farmers to monitor pollutants and to control the environmental impact without unnecessary operation. Besides this the economic impact of wet acid scrubber on the economic performances of the process should be evaluated considering the increase of the production cost and, on the other side, the willingness to pay of consumer for pig meat produced in pig barns with improved air quality.

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8. Improving the Sustainability of Dairy Slurry by A Commercial Additive Treatment

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Abstract

Ammonia (NH_3), methane (CH_4), nitrous oxide (N_2O), and carbon dioxide (CO_2) emissions from livestock farms contribute to negative environmental impacts such as acidification and climate change. A significant part of these emissions is produced from the decomposition of slurry in livestock facilities, during storage and treatment phases. This research aimed at evaluating the effectiveness of the additive "SOP LAGOON" (made of agricultural gypsum processed with proprietary technology) on (i) NH_3 and Greenhouse Gas (GHG) emissions, (ii) slurry properties and N loss. Moreover, the Life Cycle Assessment (LCA) method was applied to assess the potential environmental impact associated with stored slurry treated with the additive. Six barrels were filled with 65 L of cattle slurry, of which three were used as a control while the additive was used in the other three. The results indicated that the use of the additive led to a reduction of total nitrogen, nitrates, and GHG emissions. LCA confirmed the higher environmental sustainability of the scenario with the additive for some environmental impact categories among which climate change. In conclusion, the additive has beneficial effects on both emissions and the environment, and the nitrogen present in the treated slurry could partially displace a mineral fertilizer, which can be considered an environmental credit.

Keywords: additive; slurry; storage; ammonia; greenhouse gas; environmental impact; LCA

8.1 Introduction

In recent decades, farming systems have evolved considerably. The intensification of livestock activity has significantly increased production levels. Moreover, intensification of livestock production (as the increased use of external inputs and services to increase the output quantity and/or value per unit input) has led to a considerable increase in the volumes of manure. To manage such an amount of manure, treatments, such as solid-liquid separation, anaerobic digestion, etc., is necessary. However, manure treatments could have a negative impact on the environment including ecosystem acidification, mainly due to ammonia (NH_3) volatilization, and climate change, as a result of the contribution of greenhouse gas (GHG) emissions, the most important of which are nitrous oxide (N_2O), carbon dioxide (CO_2), and methane (CH_4) [1,2]. In addition, the management of large volumes of excreta produced from livestock farms can cause malodorous emissions.

According to IPCC [3], agriculture globally contributes 10% to 12% of anthropogenic CO_2, 40% of CH_4, and 60% of N_2O emissions. Over a 100 year time period, CH_4 and N_2O have a Global Warming Potential of 28 and 265 CO_2-equivalent, respectively, and N_2O is also implicated in the reduction of stratospheric ozone [4,5].

GHG emissions are biogenic and affected by manure characteristics; therefore, emissions can be regulated by manure management, treatment and storage conditions. CH_4 and CO_2 are produced by the degradation of organic matter in manure, while the emission of N_2O is influenced by the nitrogen (N) and carbon content of manure, duration of storage and type of treatment. N_2O is principally produced by the nitrification-denitrification process subsequent to manure application [4,6].

Although NH_3 is not a GHG, it is considered a secondary source of N_2O due to its re-deposition on the land [3], and is mainly released from agriculture (94% of the total European NH_3 emissions [7]). Ammonia can also cause eutrophication, acidification, and disturbance of natural ecosystems after its deposition and is also a precursor of fine particulate matter ($PM_{2.5}$) formation. A recent study [8] highlighted that increased emissions of ammonia lead to an increased health risk with a nonlinear ratio (e.g., 20% agricultural output increase results in 24% increase in excess mortality in European Union).

Both the European Union (EU) and the United Nations (UN) have introduced policies, over the years, in order to improve the use of nutrients present in the manure and to decrease N emissions into the environment [9]. Among these policies, the EU National Emission Ceiling Directive [10] and the Gothenburg protocol of the UNECE Convention on Long-range Transboundary Air Pollution [11] were introduced specifically to decrease the emissions of NH_3 and NOx; the UN-FCCC Kyoto protocol has been adopted worldwide to decrease N_2O emission [12], while the EU Nitrates Directive is aimed at decreasing N leaching into ground and surface waters [13]. Countries that are party to the Kyoto Protocol are moving towards reducing global warming potential by reducing GHG concentrations in the atmosphere to "a level that would prevent dangerous anthropogenic interference with the climate system" [12]. Therefore, livestock farms and industries are developing mitigation strategies to reduce these emissions. In the development of appropriate mitigation technologies, it is fundamental to consider several aspects of manure management, such as manure excretion, storage, treatments and land application [14], as well as evaluating GHG emissions on the basis of a whole system approach, since the effects of mitigation methods used at one stage may affect emissions in downstream phases [15].

Various strategies for manure management practices and types of treatment can be adopted in order to exploit the nutrients from livestock manure whilst, at the same time, minimizing the environmental impact of NH_3 and GHG emissions [16,17]. In Europe, these strategies are listed in the horizontal BAT (Best Available Techniques) Reference Document (BREF), entitled "Emissions from Storage". In this section, BAT aim at reducing NH_3 emissions into the air and nitrate leakage into the water from manure storage and spreading techniques [18]. Among the most common treatments are solid-liquid separation, anaerobic digestion, manure storage covers and slurry acidification.

Solid-liquid separation reduces NH_3 volatilization into the atmosphere thanks to the reduced solid content of the manure that facilitates slurry infiltration, consequently reducing its exposure time on the soil surface [17]. This technique is useful for various applications, such as animal bedding, compost and commercial fertilizer.

The anaerobic digestion of manure converts the organic N (undigested feed protein) in the manure into ammonium (NH_4^+), which can be more rapidly utilized by the crops [19]. The manure effluent produced, ensures enough soil fertility, if applied in an adequate amount, and the availability of the nitrogen necessary for crop and forage production. However, the increased ratio of ammoniacal N to total N after anaerobic digestion may cause an increase in NH_3 loss after land application.

Covering stored manure provides a physical barrier to help reduce NH_3, particulate matter (PM) formation and odorous emissions [9]. The main solutions adopted involve reducing the free surface of the slurry by either constructing rooftops or covering the surface with different materials [20].

Slurry acidification enhances the fertilizing value of slurry and is used in some countries (e.g., Denmark) to reduce NH_3 emissions. The efficiency of acidification is dependent on the additive used (chemical formula, dosage, and application method), the target pH, the slurry type, and the position along the slurry management chain in which the additive is used. Usually sulfuric acid (H_2SO_4) is added to the slurry to lower the pH, consequently reducing NH_3 emissions, while it has no effect on GHG emissions [16,21,22].

Some of these manure management practices require a large amount of capital and high maintenance costs and necessitate specific knowledge for correct operation. Therefore, an alternative approach is the use of additives during slurry storage since they are able to affect the slurry properties by inhibiting or stimulating a particular microbiological process [14]. There are different types of additives that work on several processes simultaneously, some of which modify both the chemical composition and the biological process of the slurry, by acting on the N content. The potential of these additives (biochar, chemical additives, and gypsum) to mitigate NH_3 and GHG emissions from stored slurry are documented in recent literature. In particular the effectiveness of chemical additives (such as More Than Manure and Pro-Act Biotech) and biochar on manure NH_3 and GHG emissions were studied, showing no significant effects on NH_3 and GHG fluxes [17,23]. Instead, regarding gypsum, which is a sulfate mineral composed of calcium sulfate dihydrate ($CaSO_4*2H_2O$), diverse and inconsistent results have been reported in literature about its effects [24–26]. However, the addition of gypsum has a great capacity to prevent N loss from manure and reduce odor since it reduces NH_3 volatilization during the composting process [27]. NH_3 volatilization reduction has also been confirmed by other authors such as Febrisiantosa et al., Li et al., and Yang et al. [24,25,28]. Yang, et al. [25] found that phosphogypsum reduced CH_4 and NH_3 emissions from poultry manure composting whilst, on the other hand,

tended to increase emissions of N_2O. The weak aspect of these studies' findings, however, is that large amounts of material are required to achieve the desired results (approximately 10% in weight).
The aim of this study was to test the ability of a commercial additive made of 100% calcium sulfate dihydrate (agricultural gypsum based on calcium sulfate dihydrate processed with proprietary technology; SOP LAGOON www.sopfarm.com) processed with a proprietary technology to reduce NH_3 and GHG (CO_2, CH_4, and N_2O) emissions from the slurry.

8.2 Materials and Methods

To evaluate the impact of the additive on gas emissions from slurry storage, a pilot study was conducted. The study investigated both gas emissions (NH_3 and GHG) and the slurry chemical composition.
Furthermore, the Life Cycle Assessment (LCA) approach was used to evaluate the potential environmental impact and benefits associated to the adoption of this additive in slurry storage systems compared to traditional techniques.

8.2.1 Experimental Setup

Fresh slurry was collected from a dairy farm situated in Northern Italy, housing approximately 100 milking cows (Az. Menozzi, Landriano, PV; 45°18'41" N and 9°15'46" E). Six 220 L barrels were filled with 65 L of fresh slurry each. Three of these barrels (1, 2, and 3) were left as a control, the remaining three (4, 5, and 6) were destined to receive the treatment.
The slurry treatment was carried out by adding the additive at a dosage of 4 g/m^3, according to the manufacturer's specifications provided on the Technical Data Sheet of the product, i.e., 0.260 g/barrel in barrels 4, 5, and 6, and then mixed. The product was added once a week at day 0 (T0), 7 (T7), and 14 (T14) and then stirred to ensure the dispersion of the additive in the barrel as recommended by the manufacturer.

8.2.2 Emission Measurements

The measurements of the emissions (NH_3, N_2O, CH_4, and CO_2) were carried out on day 0, day 4, day 7, and day 26 (T0, T4, T7 and T26 respectively). Prior to measurement, the slurry was mixed in order to break up any crusts that could affect the emission stream, while the additive's application on day 0 and 7 was done after the measurements.
The measurement protocol was designed to investigate the short-term performance of the additive (T4 and T7) and whether potential washout effects occurred after two weeks when no further additive was applied (T26). The time interval for this study is in line with other works on emissions from manure [25,28].
The emissive fluxes from the barrels were measured by means of a static or closed chamber according to the "non-steady-state chamber-method". This method is widely reported in literature and used to measure NH_3 and GHG emissions from different emitting surfaces [6,20,26,29,30].

The closed chamber method is based on the determination of the increasing rate of gas concentration within the internal volume of the closed chamber, positioned on the emitting surface, avoiding any air replacement. The gas concentration detected typically shows a linear increasing trend followed by a saturation phase as represented in Figure 1. The slope of the regression line, calculated within the linear part of the saturation curve, multiplied by the chamber volume to area ratio, represents the emission potential of the surface, expressed in mg/m^2*h.

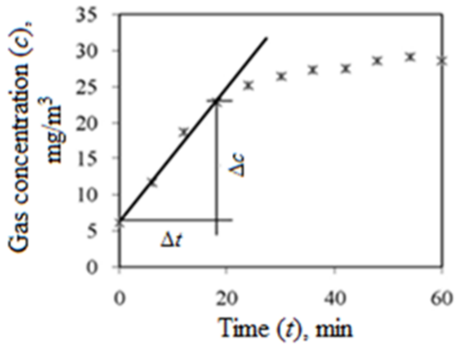

Figure 1. Gas concentration in the measuring chamber over time.

The emission factors (mg/m^2*h) were calculated according to Equation (1):

$$\text{Emission factor}_{\text{gas}}\left[\frac{mg}{m^2 \cdot h}\right] = \frac{\delta C\left[\frac{mg}{m^3}\right]}{\delta t[h]} \cdot \frac{V_{ch}[m^3]}{A_{ch}[m^2]}$$

where $\delta C/\delta t$ is the angular coefficient of the regression line of the linear branch of the gas saturation function, while V_{ch} and A_{ch} are the volumes and the area of the chamber.

The concentration of NH_3, N_2O, CH_4 and CO_2 (mg/m^3) were measured by means of an Infrared Photoacoustic Detector (Bruel&Kjaer, Nærum, Denmark, multi-gas monitor type 1302). The instrument automatically collects and analyzes air samples at regular time intervals (every 2 min until saturation values), and then the air analyzed is redirected into the barrel. During measurement, the barrels were sealed with a suitable lid, equipped with two holes, one for the inlet and one for the outlet of the gases, and a small fan, to allow the uniform distribution of the air within the top space of the sealed chamber (Figure 2) [31,32].

8.2.3 Chemical Analyses

The composition of the slurry was studied to evaluate the effects of the additive on the slurry's characteristics. Slurry was collected from each barrel and samples were analyzed for total solids (TS), volatile solids (VS), total nitrogen (TN), ammonium nitrogen (NH_4-N), nitrites (NO_2-N), and nitrates (NO_3-N), in order to verify that a reduction in emissions would not result in an increased concentration of noxious nitrogen compounds (i.e., nitrates).

Immediately after measurement of the emissions, each barrel was mixed again, in order to make it as homogeneous as possible, and 1 L of the material was sampled for lab analyses. The samples were stored at 4°C until the day of analyses, maximum two days after sampling.

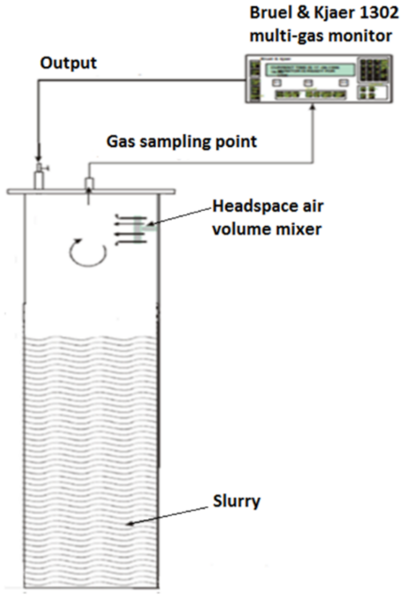

Figure 2. Schematic view of the sampling process.

The analytical methods used were:

- Total solids (TS): drying in the stove to 105°C for 48 h (APHA, 2012)
- Volatile solids (VS): incineration in muffle furnace 550°C for 2 h (APHA, 2012)
- Total nitrogen (TN): Koroleff digestion (peroxydisulfate) and photometric detection with 2,6-dimethylphenol (EN ISO 11905-1)
- Ammonium nitrogen (NH_4-N): indophenol blue (ISO 7150-1)
- Nitrites (NO_2-N): diazotization (EN ISO 26777)
- Nitrates (NO_3-N): 2,6-dimethylphenol (ISO 7890-1-2-1986).

The pH was determined using a pH meter at the beginning (T0) and at the end of the trial (T26).

8.2.4 Statistical Analyses

Statistical analysis was carried out using SAS software (SAS version 9.3; SAS Institute, Cary, NC, USA, 2012). The emission factors of each gas were calculated, in each barrel, with linear regression (PROC REG), using, as input data, the concentrations measured in the chamber at regular time intervals (every 2 min). The calculated emission factors were then used to evaluate if there was a difference among emission factors originating from treated and control samples.

The mean and standard error of the three replicates were reported in Tables.

8.2.5 Life Cycle Assessment

The goal of this LCA study is to evaluate the environmental performance of the additive during slurry storage. To this purpose, LCA was applied to the slurry storage with (Alternative Scenario, AS) and without (Baseline Scenario, BS) the additive and, finally, the environmental impact of these two solutions was compared in order to identify the most sustainable system for managing slurry from an environmental point of view.

The functional unit (FU) is the unit to which all inventory data refers, in accordance with the description of the function of the product to be analyzed. In this study, the selected FU is 1 ton of stored dairy cattle slurry. The system boundary is from cradle to gate and includes all the processes from the extraction of the raw materials included in the study to the end of the slurry storage as shown in Figure 3.

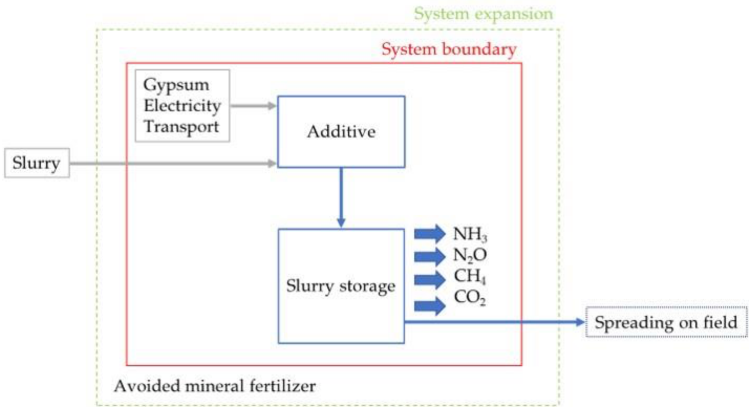

Figure 3. System boundaries.

Capital goods (e.g., slurry storage facility) were excluded from the system due to their long life span [33] and because their use is the same in the two scenarios (BS and AS). A "zero burden" approach was considered for the slurry; as in previous LCA studies [34–36], the slurry was considered a waste product deriving from the livestock production process. The distribution of the stored dairy cattle slurry was excluded from the system boundary; however, considering that by the end of storage the two slurries have different nitrogen contents, a system expansion approach was also applied, and the substitution of the mineral fertilizer was considered. More specifically, this means that the avoided use of a mineral fertilizer was quantified for highlighting the possible additional beneficial environmental effect of the additive. The amount of avoided fertilizer produced was calculated depending on the mineral fertilizer equivalence (MFE: is a measure of the fertilizer's ability to supply nitrogen to crops compared with a mineral fertilizer). MFE equal to 100% was considered for the N-NH_3 in the slurry [37] and the substitution of nitrogen by ammonium nitrate was taken into account.

Inventory data regarding the emissions of NH_3, N_2O, and CH_4 for BS were taken from Baldini et al. [38], while for AS they were collected during the experimental measurements previously described. The recommended storage period varies in different geographical areas, ranging from a few days in some areas of the United States to several months in other countries. For example, in Italy, the recommended storage period, before land application, is 120–150 days in zones vulnerable to nitrate pollution and 90 days in non-vulnerable zones. A period of 180 days was taken into account for the modeling of emissions in the LCA study to be more comprehensive of the different storage recommendations.

Emissions of NH_3, N_2O and CH_4 from slurry storage were estimated after 180 days from the relative reduction achieved by the addition of the additive. Once quantified, these values were also attributed to the amount of slurry produced per cattle per year and referred to the FU.

Inventory data regarding the additive were collected directly from the producer and the amounts adopted per FU are reported in Table 1.

Table 1. Main inventory data for additive manufacturing

Production factors	Unit	Quantity
Electricity	kWh/kg	0.899
Gypsum	kg	1
Transport distance	kg/km	45

Secondary data regarding the production of electricity, diesel fuel, and gypsum were obtained from Ecoinvent databases v 3.5 [39].

For the Life Cycle Impact Assessment (LCIA) step, in which the inventory data are transformed into indicators of its environmental impact, the ILCD 2011 Midpoint characterization method was selected. This method was widely adopted in recent LCA studies as it was recommended and endorsed by the European Commission, Joint Research Centre in 2012 [40]. In particular, it is characterized by the currently most reliable characterization factors and, being widely adopted, allows improvement in the standardization of the method and the comparability of results with other studies.

Twelve impact categories (namely environmental effects) were considered: climate change (CC), ozone depletion (OD), particulate matter formation (PM), human toxicity-no cancer effect (HTnoc), human toxicity with-cancer effect (HTc), photochemical ozone formation (POF), terrestrial acidification (TA), eutrophication of terrestrial ecosystems (TE), freshwater eutrophication (FE), marine water eutrophication (ME), freshwater ecotoxicity (FEx), and mineral and fossil resource depletion (MFRD).

8.3 Results

Table 2 summarizes the coefficients determination (R^2), that is a goodness-of-fit measure for linear regression models, and *p*-values for each emission factor that was calculated with the regression analysis. For all the samplings conducted, R^2 values amounted within a range of 67% e 99% and *p*-values showed high levels of significance. The high R^2 values are characteristic of a model that explains all the variation in the response variable around its mean, and *p*-values below the level of significance highlight how changes in the independent variable are associated with changes in the response at the population level.

Table 2. R-squared value for each measure and its *p*-value.

		T0		T4		T7		T26	
		Control	**Treated**	**Control**	**Treated**	**Control**	**Treated**	**Control**	**Treated**
NH_3	R^2	0.91	0.93	0.67	0.72	0.8	0.79	0.92	0.96
	p-values	<0.0001	<0.0001	0.013	0.011	0.003	0.006	<0.0001	<0.0001
N_2O	R^2	0.86	0.75	0.88	0.93	0.79	0.69	0.95	0.9
	p-values	0.05	0.05	<0.0001	<0.0001	0.05	0.05	0.0004	0.0007
CO_2	R^2	0.99	0.99	0.89	0.93	0.99	0.99	0.97	0.99
	p-values	<0.0001	<0.0001	0.008	0.0005	<0.0001	<0.0001	<0.0001	<0.0001
CH_4	R^2	0.99	0.99	0.86	0.89	0.97	0.99	0.96	0.98
	p-values	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001

8.3.1 The Effect on NH_3 and GHG Emissions

The emission factors per each gas (NH_3, and GHG) are shown in Table 3.

Table 3. Greenhouse Gas (GHG) emission factors (g/m^2*h).

	T0		T4		T7		T26	
	Control	**Treated**	**Control**	**Treated**	**Control**	**Treated**	**Control**	**Treated**
NH_3	1.109 (±0.047)	1.109 (±0.118)	0.091 (±0.065)	0.000 (±0.001)	0.408 (±0.032)	0.136 (±0.026)	1.654 (±0.034)	0.646 (±0.049)
N_2O	0.003 (±0.002)	0.003 (±0.002)	0.018 (±0.010)	0.000 (±0.001)	0.000 (±0.001)	0.001 (±0.001)	0.015 (±0.004)	0.018 (±0.002)
CO_2	28.078 (±0.999)	28.078 (±0.947)	603.73 (±136.191)	465.615 (±51.591)	74.723 (±2.485)	69.526 (±1.552)	92.968 (±6.629)	104.323 (±3.053)
CH_4	0.632 (±0.027)	0.632 (±0.031)	109.827 (±39.258)	86.184 (±12.185)	8.943 (±0.537)	9.318 (±0.212)	10.389 (±0.871)	12.699 (±0.693)

Data represents mean ± Standard Error (n = 24).

Regarding NH_3 emissions, the additive had a strong effect in the first week (T4 and T7). The measurements carried out at T4 showed an efficacy of 100%. A mitigation effect was also visible at T26, approximately two weeks after the last application of the additive (Table 3).

Also, as regards the GHG emissions, from the PROC REG, the treatment with the additive showed a strong reduction of emissions on day 4, where the control samples had the highest peaks of emissions for all gases. N_2O, CO_2, and CH_4 emissions, from the treated slurry, were respectively 100%, 22.9%, and 21.5% lower than the control at T4 when the emission peaks were recorded.

On day 7, the emission factors were much lower with respect to T4. At T7 the slurry treated with the additive had slightly lower emissions of CO_2 than the control (Table 3).

8.3.2 The Effect on Slurry Chemical Characteristics

Table 4 represents the slurry's chemical properties. The TS increased until T7 and then decreased at the end of the experiment (T26) for both the control and the treatment.

Table 4. Properties of slurry without additive (control) and with additive (treated) at T0, T4, T7, and T26.

	T0		T4		T7		T26	
	Control	**Treated**	**Control**	**Treated**	**Control**	**Treated**	**Control**	**Treated**
TS	78.41	78.41	97.48	99.91	99.36	103.88	91.37	93.21
(g kg^{-1} DM)	(±1.22)	(±1.37)	(± 1.63)	(±2.11)	(±1.62)	(±1.11)	(±3.28)	(±2.81)
VS	83.86	83.86	84.45	83.96	83.85	83.83	81.76	81.90
($\%_{TS}$ DM)	(±0.76)	(±0.41)	(± 0.42)	(±0.93)	(±0.47)	(±0.59)	(±0.35)	(±0.86)
TN	1.86	1.86	1.88	2.02	2.75	2.61	2.30	2.14
(g kg^{-1} WW)	(±0.10)	(±0.11)	(± 0.03)	(±0.04)	(±0.21)	(±0.24)	(±0.36)	(±0.02)
NH_4^+-N	0.88	0.88	0.99	1.00	1.00	0.99	1.03	1.00
(g kg^{-1} WW)	(±0.06)	(±0.03)	(± 0.03)	(±0.03)	(±0.01)	(±0.01)	(±0.04)	(±0.09)
NO_2-N	12.80	12.80	14.29	14.20	16.77	17.20	17.07	17.00
(mg kg^{-1} WW)	(±0.1)	(±0.2)	(± 0.3)	(±0.8)	(±0.4)	(±0.6)	(±0.8)	(±0.3)
NO_3-N	89.60	89.60	158.40	154.0	152.88	163.60	736.12	727.50
(mg kg^{-1} WW)	(±4.5)	(±2.8)	(± 18.0)	(±3.5)	(±5.3)	(±4.6)	(±54.1)	(±38.4)

TS: Total Solids; VS: Volatile solids; TN: Total Nitrogen; NH_4^+-N: Ammonium-nitrogen; NO_2-N: nitrites; NO_3-N: nitrates. DM: dry matter; WW: wet weight. Data represents mean ± Standard Error.

A VS reduction occurred throughout the duration of the study with more evident VS losses at the end of the experiment, both for the treated slurry and the control.
Subtle fluctuations were observed for NH_4^+-N and NO_2-N.
Instead, regarding TN and NO_3-N contents, the treatment's effect on the slurry was evident, in fact, the slurry with the additive showed lower values of these elements at the end of the experiment. In particular, the average TN content of the trial period was 2.20 and 2.16 g/kg for the control and treated, respectively.
The most interesting aspect regards the lower NO_3-N content observed in the treated samples.

8.3.3 Environmental Impact

Table 5 reports the comparison between the environmental impacts of the two scenarios while the relative comparison is reported in Figure 4.

Table 5. Potential environmental impact for Baseline Scenario and Alternative Scenario.

Impact Category	**Acronym**	**Baseline Scenario**	**Alternative Scenario**
Climate change	CC	132.6 kg CO_2 eq	110.1 kg CO_2 eq
Ozone depletion	OD	$-5\cdot10^{-10}$ kg CFC-11 eq	$-7.3\cdot10^{-12}$ kg CFC-11 eq
Human toxicity, non-cancer effects	HTnoc	$-1.6\cdot10^{-9}$ CTUh	$-8.3\cdot10^{-10}$ CTUh
Human toxicity, cancer effects	HT-c	$-1.8\cdot10^{-10}$ CTUh	$-1.7\cdot10^{-11}$ CTUh
Particulate matter	PM	0.062 kg $PM_{2.5}$ eq	0.039 kg $PM_{2.5}$ eq
Photochemical ozone formation	POF	0.052 kg NMVOC eq	0.043 kg NMVOC eq
Terrestrial acidification	TA	2.785 molc H^+ eq	1.769 molc H^+ eq
Terrestrial eutrophication	TE	12.452 molc N eq	7.911 molc N eq
Freshwater eutrophication	FE	$-1\cdot10^{-6}$ kg P eq	$4.95\cdot10^{-8}$ kg P eq
Marine eutrophication	ME	0.085 kg N eq	0.054 kg N eq
Freshwater ecotoxicity	FEx	−0.039 CTUe	−0.014 CTUe
Mineral, fossil & ren resource depletion	MFRD	$-3.3\cdot10^{-7}$ kg Sb eq	$-1.7\cdot10^{-7}$ kg Sb eq

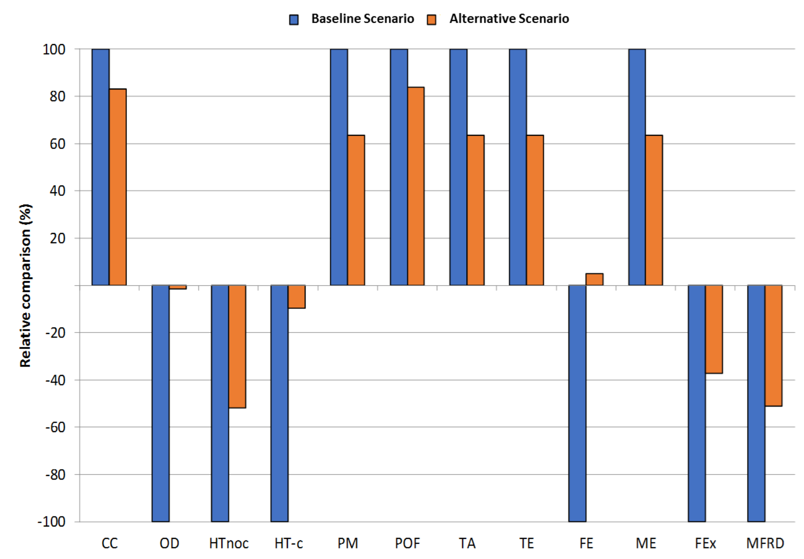

Figure 4. Relative comparison between Baseline Scenario and Alternative Scenario.

The results are reported as a percentage with the worst-performing scenario set at 100%.

The emissions of gases during storage are the main contributors to CC, PM, POF, TA, TE, and ME; regarding these impact categories, the contribution resulting from the production of the additive is negligible (<1% of the total impact). CH_4 and N_2O deeply affect CC, while NH_3 is the main gas responsible for PM, TA, TE and ME. The AS is the best for the CC impact category, since the use of the additive results in a reduction of CH_4 and N_2O emissions. Since NH_3 emissions are the main contributor to TA, TE, ME, and PM categories, the solution with the use of the additive can be considered the most appropriate. The impact of the production of the 12 g of additive required for the FU considered (gypsum extraction, processing, and transport, as well as energy consumption) is enough to affect the other six categories, whilst the BS scenario is not influenced by external factors.

Figure 5 reports the relative contribution of the additive, the emissions and the avoided production of mineral fertilizer for AS (the scenario in which the additive is used).

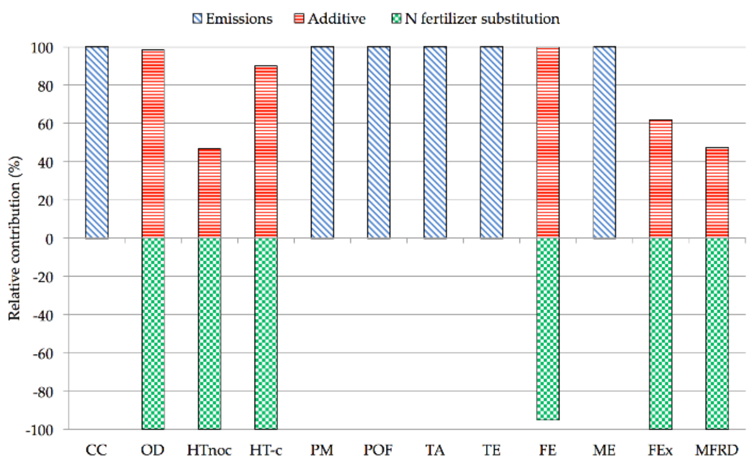

Figure 5. Relative environmental contribution for Alternative Scenario.

The avoided production of mineral fertilizer, considered as a credit in both scenarios, has considerable environmental benefits. For OD, toxicity-related impact categories and MFRD, this credit is higher than the impact and, consequently, the whole process has environmental benefits. For the other six evaluated impact categories, the avoided production of ammonium nitrate is not a key driver of the environmental results.
Considering the N-NH_3 content of the treated slurry a valid substitute for the nitrogen coming from mineral fertilizers, the avoided production of synthetic fertilizer results in considerable mitigation effects in terms of OD, HTnoc, HT-c, FEx, and MFRD, which globally offsets the impact that the production of the additive had on the mentioned categories.

8.4 Discussion

Sources of emissions of NH_3 and GHG in dairy farms are represented by manure on the barn floor, manure treatment, and storage. This study focused on the possible contribution of a commercial additive to reduce the emissions of NH_3 and GHG from slurry, checking also the slurry properties with particular attention to microbial N transformations.

8.4.1 Effects on NH_3 and GHG Emissions

Since slurry storage is a source of NH_3 and GHG emissions, it is important to take into account their global warming potential. Our data showed that the gypsum additive applied to the stored slurry decreased emissions of NH_3 and GHG and reduced the pH value of the slurry.
The reduction in NH_3 emissions may be attributed to the formation of ammonium sulfate and calcium nitrate. Indeed, in an acidic environment, NH_4^+ and NO_3^- ions, normally present in the slurry, react with SO_4^{2-} and Ca^{2+} deriving from gypsum.
Concerning GHG emissions, the additive has good reduction effects on CH_4, which is the predominant greenhouse gas emitted from slurry storage, and N_2O that play a role when surfaces become encrusted. This reduction could be attributed to several factors such as the increase in the SO_4^{2-} content of the slurry that has a toxic effect on methanogens, thus inhibiting CH_4 production. During the trial, there was a fluctuation of the emission factors, probably due to the increase of microbial activity in the slurry; nevertheless, the addition of the product led to a reduction of GHG emissions. The ability to reduce GHG emissions after 4 days and the rise of fluxes at the end of the trial period seem to indicate that more than three applications of the additive may be required to achieve maximum results.
With regard to the reduction of NH_3 emissions, the results were in line with Lim, et al. [41] that found that the addition of phosphogypsum led to an NH_3 volatilization reduction of 56–69% after 25 days. Febrisiantosa et al. [28] found that the supplementation of flue gas desulphurization (FGD) gypsum resulted in a reduction of NH_3 volatilization (26-59%). These reductions could be attributed to a lower pH and to NH_3 absorption

affected by the addition of these types of gypsum. In fact, the NH_3 volatilization is favored by an alkaline pH and a high concentration of NH_4^+ in the slurry. However, on the contrary to that found in this work, Febrisiantosa's study showed also an increase in the NO_3-N accumulation in the manure by 6.7–7.9 fold, compared to the initial value using FGD gypsum.

The acidification of the slurry obtained and the inhibition of the denitrification process, indicated by the lower accumulation of NO_3-N, seem to have favored the reduction of CH_4 and N_2O emissions in our study. Similar results were found by Berg, et al. [42], with the addition of lactic acid and Luo et al. [43], Hao et al. [44] and Yang et al. [25] with phosphogypsum. Regarding the reduction of N_2O emissions, some authors have found opposite results to ours, in fact they found a slight increment of the release of that gas after treatment [24,25,44].

8.4.2 Effects on Nitrogen Management

It is well known that nitrogen is essential for the development of field crops, but excessive nitrates deriving from manure and slurry represent a serious threat to the environment. Hence, best practices for slurry and manure management are widely recommended and known [20,45]. In general, slurry acidification, obtained by the use of gypsum, modifies the slurry characteristics and its subsequent application, as well as increases the fertilization value of slurry, without negative impacts on other gaseous emissions. The fertilization value may increase due to the lowered ammonia emission, and the increased inorganic dissolution [46,47]. The results of this study showed that the use of the tested additive could actually modify slurry characteristics, increasing the TS and reducing NO_3-N. These properties can improve the amendment features of the slurry, avoiding excessive leaching of the nitrates into groundwater and providing the right quantity of nutrients to the crops, safeguarding the environment.

8.4.3 Environmental Benefits

LCA shows that the environmental benefits achieved by reducing the use and production of synthetic fertilizers, together with the benefits related to the reduction of air emissions, make this treatment an interesting option for reducing the consumption of fossil resources for the production of synthetic fertilizers and to mitigate NH_3 emission into the air and the related Particulate Matter formation, Acidification and Eutrophication.

The results of this LCA study showed that, by making a projection of the values at 180 days, the treated sample would give off 1.6 times lower emissions than the raw slurry, as confirmed by the reduction of the CC impact when the AS is considered. The AS resulted as having higher environmental sustainability than the BS (unprocessed slurry) on most impact categories.

5. Conclusions

Compared to other additives studied, such as lactic acid or phosphogypsum, the additive studied here proved to be able to simultaneously mitigate NH_3, N_2O, CH_4, and CO_2 emissions from slurry storage.

It is also important to notice that these results were obtained with a much lower amount of product than that proposed in other studies for the other amendments: the additive under examination here was applied 3 times at 4 g/m^3 while other materials were used at a minimum of 3-10% of manure wet weight, proving, thus, to be a scalable solution.

The additive was able to control NH_3 and N_2O emissions, without accumulating nitrates. The use of slurry treated with this additive as organic fertilizer could be beneficial for the environment since its NO_3-N content appears to be lower. In addition, the use of treated slurry avoids the use and the purchase of mineral fertilizers, thus resulting in an economic advantage for the farmer.

The LCA study also confirms the environmental sustainability of this solution with the additive with respect to the environmental impact categories of Climate Change, Particulate Matter formation, Photochemical Oxidant Formation, Acidification and Terrestrial and Marine Eutrophication.

This study shows the alignment of this treatment with some of the UN Sustainable Development Goals (SDG), among which SDG number 13-Climate Action and 6-CleanWater and Sanitation.

For all these results, the application of the studied slurry additive can be considered an effective method to improve the sustainability of dairy slurry management.

Since the results of this study are promising, pilot and large-scale tests will be necessary to investigate the effectiveness and economic feasibility of this method also in other types of slurry management systems.

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CHAPTER 6

Adding insects to our menus could help overcome the world's food supply problems. Moreover, rearing, and consequently eating, terrestrial invertebrates could help reduce the contribution of livestock to climate change, limit biodiversity loss and pollution. Chapter 6 consists of two peer reviewed articles (*Paper 7 and 8*) that evaluate the environmental impact of the bioconversion of Fruit and Vegetable Waste (FVW) into earthworm meal to be used as new food/feed source.

9. Bioconversion of fruit and vegetable waste into earthworms as a new protein source: The environmental impact of earthworm meal production

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Abstract

Food waste is recognized as a global issue affecting the sustainability of the food supply chain. The unnecessary exploitation of natural resources (land, water and fossil energy) and production of greenhouse gas emissions (GHG) make the reduction of food waste a key point. In this context, the use of fruit and vegetable waste (FVW) as growth substrate for fresh earthworms to produce dried meal for feed and food purpose can be recognized as a viable solution.

Therefore, the aim of this study is to evaluate the environmental impact of the bioconversion of FVW into earthworm meal to be used as new food/feed source. This is carried out by adopting the Life Cycle Assessment (LCA) method with an attributional approach and solving the multifunctionality of the system with an economic allocation between earthworms and vermicompost.

The results show that themain process hotspots are the emissions of methane, dinitrogen monoxide and ammonia taking place during vermicomposting, as well as FVW transport and electricity consumed during fresh earthworm processing. Respect to the one used as feed, the dried meal with food purpose shows a higher impact due to the higher economic value and to the higher electricity consumed during freeze drying compared to the ovendrying process for feed meal production. Enhancing productivity and reducing energy consumption are necessary to improve the sustainability of earthworm meal as food/feed source.

Keywords: Life Cycle Assessment; Circular economy; Fruit vegetable waste; Earthworm; Novel food/feed protein; Sustainability

9.1 Introduction

Foodwaste is already recognized as an important global issue affecting the sustainability of the food supply chain (Tonini et al., 2018). The losses that occur during the whole lifecycle in terms of food scraps and wasted food in both the agricultural/industrial and domestic phases, can account for up to 60% of the initial weight of the food products (Notarnicola et al., 2017). According to the Food and Agriculture Organization of the

United Nations (FAO) about 1.3 billion tons of food produced worldwide every year are wasted along the supply chain (FAO, 2011).

The problem of food waste involves significant environmental, economic and social impacts. Food waste leads to an unnecessary exploitation of natural resources (land, water and fossil energy) and to notable greenhouse gas (GHG) emissions (Pham et al., 2015). Food produced but lost along the chain and wasted accounts for around one quarter of total freshwater resources and of total global cropland area used and lost, and has embedded production-phase emissions that should be counted (Kummu et al., 2012; Porter et al., 2016). The median GWP (Global Warming Potential) value for different food categories were estimated to account 0.37 kg CO_2-eq/kg for field-grown vegetables and 0.42 kg CO_2-eq/kg for field-grown fruit with meat from ruminants having the highest impact 26.61 kg CO_2-eq/kg bone free meat (BFM) (Clune et al., 2017).

Fruit and vegetable waste (FVW) is one of the major categories in the food waste generated, especially in industrialized regions (FAO, 2011; Porter et al., 2016). In EU contribute to almost 50% of the food waste generated by households (De Laurentiis et al., 2018). Moreover, FVW management poses disposal and environmental problems, due to its high biodegradability but also a great potential for reuse, recycling and energy recovery (Plazzotta et al., 2017). On the other hand, the patterns of growth in demand for animal-source foods (Alexandratos and Bruinsma, 2012; United Nations, 2017), poses terrestrial invertebrates as a suitable candidate to supplement other animal-based food proteins (FAO, 2013). A possible strategy is the utilization of FVW as feeding substrate for the rearing of terrestrial invertebrates to be used as potential protein source for feed and/or food supply chains. Terrestrial invertebrates represent a potential valuable solution to two problems: 1) the increasing amount of foodwaste, which can cause environmental pollution if not properly managed, and 2) the rising global demand for food and feed, necessary to supply human and animal nutrition (Salomone et al., 2017).

Up to now, the attention on alternative protein sources has regarded mainly terrestrial invertebrates from insects both as human food (Oonincx and De Boer, 2012; Halloran et al., 2016) and as animal feed (Smetana et al., 2016; Salomone et al., 2017; Tallentire et al., 2018; Thévenot et al., 2018). Among others terrestrial invertebrates, earthworms could be an interesting solution to be evaluated. In nature, earthworms grow on a wide variety of organic materials (Edwards, 1988) and are even called "ecosystem engineers" (Jones et al., 1994), as they are the main drivers of the decomposition of organic waste in soil ecosystems (Lim et al., 2016).

Earthworms provide an excellent ecosystem service modifying the physico-chemical properties of soil (Singh et al., 2016) maintaining aerobic conditions (Nigussie et al., 2016). They are voracious eaters and biodegraders of waste (Sims and Gerard, 1985; Sinha et al., 2008). Therefore, they can be fed on different types of waste, such as FVW (Chatterjee et al., 2014; Huang et al., 2016; Huang and Xia, 2018). Furthermore, vermicomposting is more effective in FVW disposal techniques to reduce GHG emissions and nitrogen losses than traditional composting (Yang et al., 2017; Swati and Hait, 2018; Colón et al., 2012). From an environmental point of view, vermicomposting process emissions of NH_3, CH_4 and N_2O are lower by three orders of magnitude than those coming from composting process (Lleó et al., 2013). Moreover, from the vermicomposting process, the derivative vermicompost is an excellent high quality bioactive amendment to improve soil fertility. It is pathogen-free thanks to earthworm gut transit mechanism which classifies vermicomposting as a promising sanitation technique in comparison to composting processes (Soobhany et al., 2017; Yang et al., 2017). Vermicomposting represents an option of FVW valorization, because it is a low-cost biotechnology that turns waste into a high-quality residue, namely vermicompost, through the joint action of earthworms and microorganisms (Dominguez, 2004; Yang et al., 2017). Currently, earthworms are employed to deal with food waste management in a bioconversion process to mitigate the food waste problem as a sustainable, cost-effective and ecological approach (Fernández-Gómez et al., 2010; Singh et al., 2011; Yadav and Garg, 2011; Huang et al., 2016; Lim et al., 2016). Nonetheless, earthworms grown on FVW can contribute to the waste disposal efficiency biotransforming FVW into two valuable products: (i) vermicompost, that can be sold as organic fertilizer, and (ii) earthworms themselves that can be a new food/feed source, thanks to their high protein content.

Like insects, earthworms are rich in proteins, particularly in essential amino acids (Cayot et al., 2009; Zhenjun and Jiang, 2017) and they can contribute to human and animal nutrition (Ncobela and Chimonyo, 2015; Bahadori et al., 2017; Zhenjun and Jiang, 2017). By the way, the production of edible terrestrial invertebrates as food or feed, have to be safe and wholesome. To ensure a high level of protection of human and animal health, terrestrial invertebrates and therefore earthworms need to be considered as “farmed animals” when they represent a food source and fed only with safe feed used as growth substrate. EU framework established restrictions or prohibitions on the feed for farm animals reared for producing food or feed (e.g. prohibited feeding: catering waste or household waste Regulation (EU) No 1069/2009 Regarding animal by-products and Regulation (EC) No 767/2009 on the placing on the market and use of feed, Annex III). Besides, safety rules for food or feed purpose were defined in the Hygiene Package (e.g. Regulation No 852/2004 and Regulation No 853/2004 on the hygiene of foodstuffs; Regulation No 183/2005 laying down requirements for feed hygiene) and on the levels of contaminants (Directive 2002/32/EC on undesirable substances in animal feed and Regulation (EC) No 1881/2006, setting maximum levels for certain contaminants in foodstuffs), according to which earthworm rearing and market as food or feed must respect the legislation in force for farmed animals and for the derivative products.

The aim is to evaluate the environmental impact of the earthworms' meal production for feed and food purpose obtained from earthworms reared on FVW discarded directly from juice and ready-to-eat processing industries. The propensity and willingness towards earthworms as a future food source has already been investigated in a previous study (Conti et al., 2018). In this study the environmental performances of earthworm meal production adopting Life Cycle Assessment (LCA) was analysed, in order to:

- evaluate the real effectiveness and sustainability to produce earthworms as food/feed protein source,
- test the environmental impact of the production system.

Two different scenarios (FEED and FOOD) were evaluated and a sensitivity analysis concerning key parameters, assumptions and methodological choices was performed.

9.2 Materials and methods

To evaluate the environmental impact of the earthworms' production system, the Life Cycle Assessment (LCA) method was used. LCA is a holistic approach, structured and recognized worldwide that consists of a systematic set of procedures to convert inputs and outputs of the studied system into its related environmental impact. To perform LCA, the ISO standard 14040/44 methodology (ISO, 2006) must be adopted.

In this study, the methodological framework of the attributional approach is used. It permits to model the production process of earthworms (*Eisenia fetida*) without considering potential effects on the market due to the use of vegetable waste and to the introduction of protein from earthworm's origin.

According to ISO 14040/44 (ISO, 2006), LCA involves four distinct and interdependent phases, all of which are discussed in detail in the next sections. These phases consist of defining and analysing:

i) goal and scope, which include the selection of a functional unit and definition of system boundary;
ii) life cycle inventory, which involves the definition of energy and material flows between the system and the environment and through the different subsystems and operations in the evaluated system;
iii) impact assessment, during which the inventory data are converted in environmental indicators (i.e. environmental impact categories); and
iv) discussion and interpretation of the results, where the results from the inventory analysis and impact assessment are summarized, sensitivity and uncertainty analysis are carried out and recommendations are drawn.

9.2.1 Goal and scope definition

Eisenia fetida is a widespread epigenic species of earthworms (Bouché, 1977; Sims and Gerard, 1985) that it is characterized by a high tolerance to a wide range of environmental factors, higher rates of consumption, digestion and assimilation of organic substances and high reproductive rate (Bhat et al., 2018; Domínguez and Edwards, 2010a). Adult earthworms of *Eisenia fetida* weigh up to 0.55 g (Domínguez and Edwards, 2010a) and reach up to 60–120 mm in length and 3–6mmin diameter (Sims and Gerard, 1985). They are hermaphrodites (Edwards and Bohlen, 1996), but usually reproduction occurs through copulation and crossfertilization, after which each of the mated individuals can produce cocoons (Dominguez and Edwards, 2010a). Approximately, the time from newly-laid cocoon through clitellate adult earthworm ranges from 45 to 51 days (Dominguez and Edwards, 2010a). Optimum growth conditions include a range of temperature between 25 and 30°C, moisture 75–90% (Edwards, 1988) and pH >5 and <9, optimum centered around 7.0 (Kaplan et al., 1980). Given the optimum conditions of temperature and moisture, about 5 kg of worms can vermiprocess 1 ton of waste into vermicompost in just 30 days (Sinha et al., 2010).

The scope is to investigate the environmental profile of the bioconversion process of FVW into earthworm dried meal as a novel food and feed protein source. The environmental impact analysis was carried out studying a small-scale production plant of earthworms housed in Northern Italy with LCA approach.

Currently, very few plants are present in Italy, and all of them are small-scaled or lab-scaled. Moreover, also the production processes are quite standardized.

The research questions for this study are as follows:

- What is the magnitude of the environmental impact of the production of earthworms' meal?
- What are the environmental hotspots for the evaluated process?

The outcomes of this study will be useful specially to decision makers, being the first results of a full environmental impact assessment about earthworms' protein production using FVW feed.

9.2.2 Functional unit

The selection of the functional unit (FU) is crucial to allow fair comparison with other studies and adequate assessments. According to ISO 14040 (ISO 14044, 2006), the FU is defined as the quantified performance of a product system and is used as a reference unit in an LCA.

In the studied context, very few studies were found in literature. Given the function of the earthworm process for novel food/feed protein production, the selected FU in this study was 1 kg of dried meal of earthworm.

9.2.3 Description of the production process of earthworms meal

The environmental impact analysis was carried out studying a small scale production system of earthworms located in the province of Lecco (North Italy) (45°55’23” N and 9°19’34” E). The production process of earthworm meal was divided in two subsystems (SS):

- SS1, characterized by the production of fresh earthworms and vermicompost,
- SS2, in which fresh earthworms are used for the meal production.

9.2.3.1 Subsystem 1: fresh earthworms and vermicompost production

The present study was approved by the Animal Ethics Committee of Milan University of Study (30.01.17; ethical code number 02/17).

Fresh earthworms and vermicompost were produced on a rearing area of 34 m^2 made up of a non-woven textile sheet used to avoid water stagnation and earthworms' escape. The area was covered with a net to avoid predators' damage. A mix of young-non-clitellum and adult-clitellate earthworms was provided by the earthworms' producer, added at an initial density of 1 kg/m^2 and reared on a feeding substrate consisting of FVW.

FVW was provided by a fruit and vegetable producer of ready-to-eat products and had a variable composition dependent on seasonality in vegetable growth and work process. The waste consisted mainly of tropical fruits, such as pineapple, papaya, mango, kiwi as well as of melon, tomatoes and grapes. The fiber components of FVW (e.g., pineapple tufts) were grinded with a gardening shredder to make them biodegradable by earthworms' activity. To reach a C:N ratio optimal for earthworm growth the FVW was mixed with straw (10:1). Fruit and vegetable wastes were left to rot for a few days before being fed to earthworms, which allowed having a narrow range of favorable chemical and environmental conditions more favorable for microbial activity and further decomposition of the growth substrate (Dominguez and Edwards, 2010b). FVW were added to feed earthworms three times a month, by introducing them on the top of the production area. In order to guarantee optimum growth conditions, moisture, temperature and pH of the growth substrate were monitored and kept under control and supplied with water if needed (moisture 84–88%; temperature 20–25°C; pH 6.07–8.02; C/N 25.34). These values are in the range of the recommended values of process factors for vermicomposting (Dominguez and Edwards, 2010b).

Earthworms were reared for three months in the most favorable environmental conditions in order to obtain the best conversion efficiency. After 3 months, they were separated mechanically from the vermicompost with the use of a trommel.

Besides earthworms, during the decomposition of FVW also an odor free and humus-like substance is produced (Suthar, 2009). Vermicomposting is the stabilization of organic material through the joint action of earthworms and microorganisms (Dominguez, 2004) and its final product is the vermicompost. In our study vermicompost represents the co-product of the production system, with possible beneficiations as a valuable replacer of conventional soil fertilizers. Specifically, it is a residue produced by earthworms, characterized by low C/N ratio, high porosity, water-holding capacity and available nutrients (Lim et al., 2015).

9.2.3.2 Subsystem 2 — from fresh earthworms to food/feed meal

Once collected, earthworms were repeatedly washed with running tap water to clean the body surface and kept in water until their digestive system were clean. Finally, washing water was removed and earthworms were packaged in plastic bags and stored at −28°C to let them enter quiescence and kill them.

For the production of dry meal, two technological transformation processes were considered depending on the final destination of the meal: food scenario and feed scenario.

For food scenario the vacuum freeze drying technology method was chosen for producing high quality dehydrated earthworm meal because this is a typical technology for food purposes in order to avoid affecting the nutritional characteristics. Earthworms were freeze-dried at a pilot scale level. Finally, freeze-dried earthworms were ground to obtain the meal.

For feed scenario, the dry meal was produced in laboratory by drying earthworms in an oven at 65°C to a constant weight and grinding.

9.2.4 System boundary definition

A "from cradle to gate" system boundary was considered. More in details, the life cycle of each sub process for both subsystems (SS1 and SS2) was considered. Consequently, the following activities were included: raw materials extraction (e.g., fossil fuels, metals and minerals), inputs manufacture (e.g., diesel fuel, electricity, tap water and trucks for FVW transport), inputs use (diesel fuel emissions), maintenance and final disposal of capital goods (e.g., the trucks used for the FVW transport).

The emissions into atmosphere (e.g., dinitrogen monoxides, methane, etc.) related to vermicomposting of FVW were also included.

Packaging, distribution, use and end-of-life of the produced meal were excluded from the system boundary. Fig. 1 summarizes the system boundary considered.

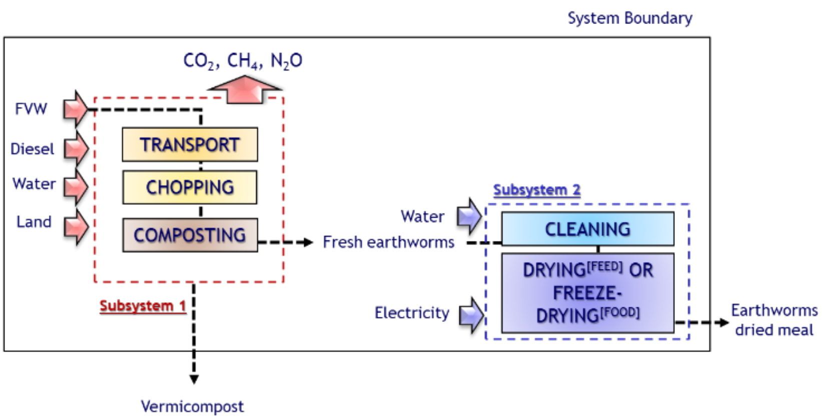

Fig. 1. System boundary (for FEED and FOOD scenario).

9.2.5 Life cycle inventory (LCI)

Inventory data relevant to the production of earthworms' biomass were collected over a three-month experimental test performed in year 2017.

Primary data were collected with questionnaires during interviews with the farmer. These regarded mainly the amount of FVW used as feed, transport, diesel, fossil energy for preparing the feed substrate, water volumes and land occupation for earthworms breeding and water for washing earthworms. Secondary data about electricity for processing earthworms into dried meal and fossil fuel for transport activities were obtained from Ecoinvent database (Weidema et al., 2013).

The main inventory data collected during the experimental trials are reported in Table 1 for SS1 and in Table 2 and Table 3 for the two scenarios in SS2.

Table 1. Main inventory data for SS1.

		Unit	Amount
Input	Fruit and vegetable waste	kg	45.73
	Transport of FVW	km	25
	Diesel	kg	0.196
	Water	m^3	3.66
	Land	m^2	0.41
Output	Vermicompost	kg	12.8
	Fresh earthworms	kg	1

Table 2. Main inventory data for SS2 for the FEED scenario.

		Unit	Amount
Input	Fresh earthworms	Kg	6.25
	Water	m^3	22.41
	Electricity	kWh	2.0
Output	Meal for feed	Kg	1

Table 3. Main inventory data for SS2 for the FOOD scenario.

		Unit	Amount
Input	Fresh earthworms	kg	6.25
	Water	m^3	22.41
	Electricity	kWh	3.67
Output	Meal for food	kg	1

Vermicomposting is a process that inevitably involves emissions of greenhouse gases (Lleó et al., 2013; Wang et al., 2014; Nigussie et al., 2016; Swati and Hait, 2018), although vermicomposting process emissions are clearly lower than those coming from composting (Colón et al., 2012; Lleó et al., 2013; Nigussie et al., 2016; Yang et al., 2017). Various factors such initial waste characteristics, process parameters like aeration, moisture content, temperature regime, contribute to fully understand the influence of process parameters in gas emissions during vermicomposting (Swati and Hait, 2018). However, the effects of earthworms on gas emissions are complicated and no consensus has been reached yet (Wang et al., 2014). Notwithstanding all this, an estimation of the gaseous emissions of CH_4, N_2O and NH_3 during SS1 was based on the relationship of growth substrate quality parameters with the gaseous emission as reported by Yang et al. (2017): consequently, the gaseous emissions per kg of fresh earthworm, were assumed equal to 5056 g CH_4, 1.53 g N_2O and 15.84 g NH_3.

9.2.6 Allocation

Considering that the production system entails the production of different products, allocation should be dealt with. In some cases, among which this study, the choice of the allocation procedure may be difficult and questionable. However, the present rearing system produces as main product an earthworm biomass growing on a mix of FVW, which is further processed into earthworm meal that leaves the holding.

Vermicompost is produced alongside as co-product. Since these two outputs are produced in very different amounts, the sharing of the environmental impact was performed with an economic allocation in order to avoid attributing an unbalanced impact, although commonly the mass allocation is suggested. Thus, economic allocation was selected and based on the estimation of earthworm meal and vermicompost prices as reported in Table 4. In detail, at the time being, no reference prices for food and feed do exist. Therefore, the earthworm meal price for both food and feed was estimated considering the economic sustainability to be achieved by this new food/feed sources in comparison with other animal protein sources currently used. For the feed dried meal, it was estimated 1.1 €/kg dry matter. As reference, the prices of fishmeal (1.46 €/kg of dry matter) (Milan Grain Association, 2018) and of insect dried meal (1.09 €/kg of dry matter) (as proposed by Salomone et al., 2017) were considered. Concerning earthworm food meal, to enable comparison with other animal food products, meat prices were recalculated to dry matter content. The estimated 15 €/kg of dry matter (22.4 €/kg protein) for food earthworm meal is related to the comparison with animal food products prices such as pork (14–16 €/kg of dry matter, 19–19.5 €/kg protein), poultry (17–18.3 €/kg of dry matter, 20.4–22.0 €/kg protein) and beef (26.6–28.6 €/kg of dry matter, 33.6–37.6 €/kg protein) (Borsa Merci Modena, 2018; ISMEA, 2018). The economic value for vermicompost was considered 0.30 €/kg (CONITALO, 2018).

Because allocation is a key methodological choice and is here based on the estimate of prices subject to variability, sensitivity analysis was performed on this issue (± 30%).

Table 4. Prices of product and co-product used in the economic allocation.

Scenario	Product	Price €/kg
FEED	Vermicompost	0.3
	Meal for feed	1.1
FOOD	Vermicompost	0.3
	Meal for food	15.0

9.2.7 Life Cycle Impact Assessment (LCIA)

The LCIA consists in transforming inventory data into environmental indicators. This step is achieved by using defined characterization factors that are gathered from characterization methods. Among the available ones, ILCD (International Reference Life Cycle Data System) midpoint characterization method (ILCD, 2012) is endorsed by the European Commission and adopted in this study. According to ILCD, the following impact categories were evaluated: Climate change (CC) midpoint (IPPC, 2007), Ozone depletion (OD) midpoint (WMO, 1999), Human toxicity midpoint, cancer effects (HT-c) and non-cancer effects (HT-noc) USEtox (Rosenbaum et al., 2008), Ecotoxicity freshwater (FEx), midpoint USEtox (Rosenbaum et al., 2008), Particulate matters (PM), midpoint RiskPoll model (Rabl and Spadaro, 2004 and Greco et al., 2007), Photochemical ozone formation (POF), midpoint (Van Zelmet al., 2008 as applied in ReCiPe2008), Acidification (TA), midpoint (Seppälä et al., 2006; Posch et al., 2008), Eutrophication terrestrial (TE), midpoint (Seppälä et al., 2006; Posch et al., 2008), Eutrophication aquatic freshwater/marine (FE and ME), midpoint (ReCiPe2008, EUTREND model — Struijs et al., 2009), Resource depletion — mineral and fossil fuels (MFRD), midpoint (CML 2002; Guinée et al., 2002).

The choice of considering these indicatorswas related to the need of providing a comprehensive evaluation of the environmental impact of *Eisenia fetida* meal production as food and feed supplements.

9.3 Results

9.3.1 Fresh earthworm production

Fig. 2 shows the environmental hotspots for SS1 (earthworm production).

Except for CC, PM, TA and TE, for the other 7 evaluated impact categories, transport of fruit and vegetable waste (FVW) from the food industry to the vermicomposting plant represents the largest contributor to the environmental impact. More in details, the contribution of transport ranges from 1.9% in TA and TE to 95% in MFRD and it is larger than 75% for 6 of the 12 evaluated environmental effects. The emissions during the FVW vermicomposting are the main contributors for CC (78%) due to the emissions of dinitrogen monoxide and methane, PM (94%), TA (97%), TE (98%) and ME (78%) due to the emissions of ammonia.

Methane emissions are responsible also of about 4% of POF. Diesel consumption during the partial chopping of FVW is responsible for a share of the environmental impact lower than 5% for all the assessed impact categories, except for POF (17%, mainly due to refinery activities). The impact related to water consumption is little (<10%) except for FE (37%, due to the emission of phosphate in water) and FEx (12%).

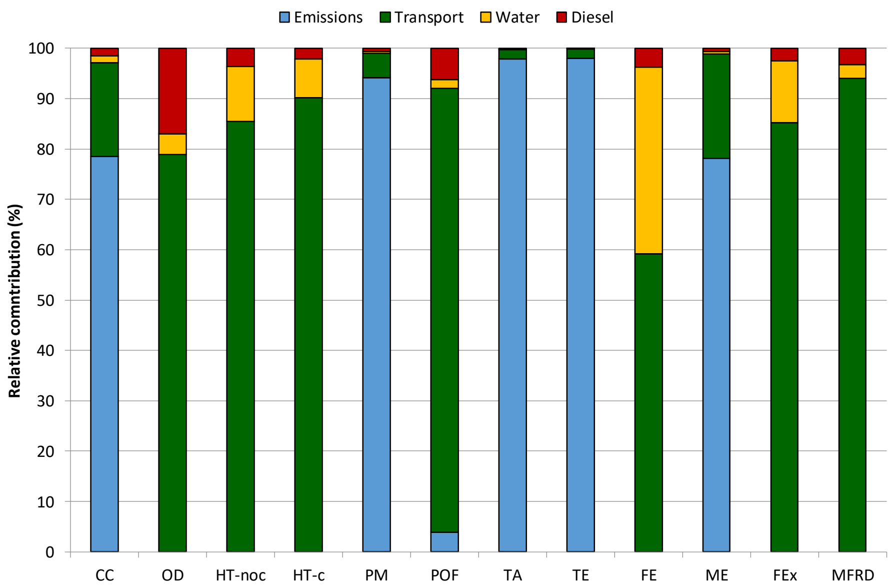

Fig. 2. Environmental hotspots for SS1.

Table 5 reports the absolute impact for producing 1 kg of fresh earthworms in the two scenarios (i.e. FEED and FOOD). The differences between the two scenarios are related to the different allocation factors (see Table 4).

Table 5. Absolute environmental impact for 1 kg of fresh earthworms in the two scenarios

Impact Category	Unit	FEED	FOOD
CC	kg CO_2 eq	0.162	0.593
OD	mg CFC-11 eq	0.0063	0.023
HT-noc	CTUh	6.07×10^{-9}	2.28×10^{-8}
HT-c	CTUh	2.03×10^{-9}	7.39×10^{-9}
PM	mg PM2.5 eq	246.8	897.38
POF	g NMVOC eq	0.293	1.064
TA	molc H+ eq	0.0107	0.0391
TE	molc N eq	0.048	0.175
FE	mg P eq	4.655	16.97
ME	g N eq	0.410	1.492
FEx	CTUe	0.140	0.498
MFRD	mg Sb eq	1.112	4.044

9.3.2 Earthworms' meal production

Fig. 3 shows the comparison between the earthworm meal produced in the two scenarios (FEED and FOOD) as well as the related environmental hotspots.

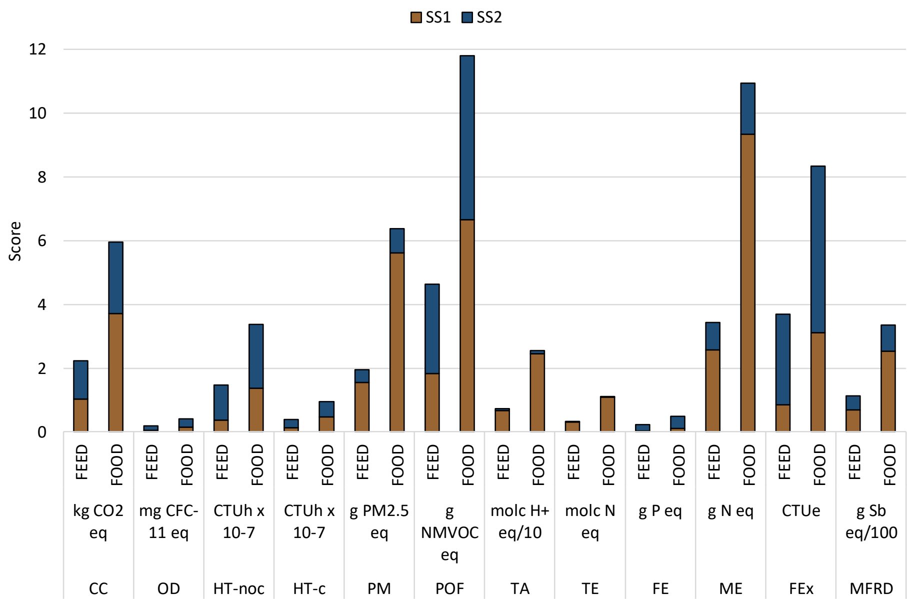

Fig. 3. Comparison between the dried meal produced in the two scenarios (FEED in red and FOOD in blue). Layout with horizontal lines: fresh earthworms, oblique lines: electricity. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

The absolute impact for producing 1 kg of earthworm meal in the two scenarios is reported in Table 6. The earthworm meal produced in the FOOD scenario shows an impact higher than the one produced in the FEED scenario. This difference ranges from 2.06 times more for FE to 3.58 times more for TE and is related to the different allocation between meal and vermicompost and, secondarily, to the higher electricity consumption in SS2 of FOOD scenario (where the fresh earthworms are freeze-dried instead of being only dried as in FEED scenario).

Table 6. Absolute impact for 1 kg of earthworm dried meal produced in the two scenarios.

Impact category	Unit	FEED	FOOD
CC	kg CO_2 eq	2.238	5.944
OD	mg CFC-11 eq	0.187	0.414
HT-noc	CTUh	1.46×10^{-7}	3.37×10^{-7}
HT-c	CTUh	3.96×10^{-8}	9.55×10^{-8}
PM	g PM2.5 eq	1.960	6.374
POF	g NMVOC eq	4.635	11.801
TA	molc H+ eq	0.073	0.255
TE	molc N eq	0.309	1.108
FE	g P eq	0.234	0.482
ME	g N eq	3.441	10.936
FEx	CTUe	3.698	8.328
MFRD	mg Sb eq	11.397	33.436

The production of fresh earthworms (SS1) is responsible of >75% of the environmental impact for the following impact categories PM, TA, TE and ME in both the scenarios and in MFRD (in FOOD scenario) and of 46% and 62% of CC in FEED and FOOD scenario, respectively. For CC, the impact related to the fresh earthworm production is mainly due to the emissions of CO_2, CH_4 and N_2O that occur during earthworms' rearing. For all the other environmental impact categories (OD, HT-noc, HTc, FE, FEx, and, even if only for the FEED scenario, also in CC and POF), the SS2 is the main responsible of the environmental impact of the dried meal, with a share of the impact ranging from 54% (CC) to 88% (FE).

9.3.3 Sensitivity analysis

A sensitivity analysis was carried out to investigate the effect of key parameters, assumptions and methodological choices of the study as well as to test the robustness of the achieved environmental results. Thus, the following aspects were considered:

- the electricity consumption during SS2. A variation ±25% was taken into account for the electricity consumed during drying in the FEED scenario and during freeze-drying in the FOOD scenario.
- The substitution of the Italian electric mix with to renewable electricity produced from a photovoltaic plant.

- the price of earthworm meal. The price of vermicompost kept constant, a variation ±30% of the price of earthworm meal was considered for both scenarios. Consequently, the allocation factors for vermicompost and earthworm meal in FEED scenario became equal to 83% and 17% and to 73% and 27% at the decrease and increase in price, respectively; in the FOOD scenario, instead, allocation factors were set to 27% and 72% and to 16% and 84% at the decrease and increase in price, respectively;
- the procedure for solving the multifunctionality issue. Considering that the ISO standards suggest to avoid the allocation, instead of the economic allocation, the multifunctionality was solved taking into account a mixed functional unit composed by earthworm meal and vermicompost. More in detail, considering that 6.25 kg of fresh earthworm are needed to produce 1 kg of earthworm meal and 12.8 kg of vermicompost are produced with 1 kg of fresh earthworm the mixed FU is composed by 1 kg of earthworm meal and 80 kg of vermicompost.

The results of the sensitivity analysis are reported in Table 7.

Table 7. Results of the sensitivity analysis: Impact variation respect to the values reported in Table 6 considering different electricity consumption (EE) in SS2, electricity produced from a photovoltaic plant (PV) and a mixed FU to avoid allocation.

Impact category	FEED						FOOD					
	EE - 25%	EE + 25%	Low price	High price	PV EE	Mixed FU	EE - 25%	EE + 25%	Low price	High price	PV EE	Mixed FU
CC	-10.9%	13.6%	-10.4%	10.4%	-33.6%	161.5%	-7.5%	9.4%	-5.5%	5.5%	-49%	15.6%
OD	-15.8%	19.8%	-4.7%	4.7%	-55.4%	74.0%	-13.1%	16.4%	-3.0%	3.0%	-67%	8.6%
HT-noc	-14.8%	18.5%	-5.9%	5.9%	36.5%	91.8%	-11.8%	14.8%	-3.6%	3.6%	46%	10.2%
HT-c	-13.6%	17.0%	-7.3%	7.3%	-15.1%	113.7%	-10.3%	12.9%	-4.2%	4.2%	-20%	12.1%
PM	-4.3%	5.3%	-17.9%	17.9%	-8.5%	279.1%	-2.4%	3.0%	-7.7%	7.7%	-15%	22.0%
POF	-12.1%	15.1%	-9.0%	9.0%	-35.4%	140.0%	-8.7%	10.9%	-4.9%	4.9%	-49%	14.1%
TA	-1.6%	2.0%	-20.9%	20.9%	-3.4%	326.0%	-0.8%	1.1%	-8.4%	8.4%	-6%	23.9%
TE	-0.6%	0.7%	-22.1%	22.1%	-1.2%	344.2%	-0.3%	0.4%	-8.6%	8.6%	-2%	24.6%
FE	-17.5%	21.9%	-2.8%	2.8%	-32.8%	44.1%	-15.6%	19.5%	-1.9%	1.9%	-37%	5.5%
ME	-5.1%	6.4%	-17.0%	16.9%	-11.9%	264.3%	-2.9%	3.7%	-7.5%	7.5%	-21%	21.3%
FEx	-15.4%	19.2%	-5.3%	5.3%	298.9%	82.1%	-12.5%	15.7%	-3.3%	3.3%	367%	9.3%
MFRD	-7.8%	9.8%	-13.9%	13.9%	786.1%	216.2%	-4.9%	6.1%	-6.6%	6.6%	1257%	18.9%

The change of electricity consumption in SS2 involves an impact variation ranging from −17.5% to 21.9% in the FEED scenario and from −15.6% to 19.5% in the FOOD scenario. FE is the impact category most affected by electricity consumption, therefore it shows the highest impact variation in both scenarios. On the opposite, PM, TA, TE and ME mainly affected by emissions occurring during fresh earthworm production, are the impact categories less affected by variations in electricity consumption during drying/freeze-drying.

When the electricity is produced from a photovoltaic plant, 9 of the 12 evaluated impacts are reduced (from 1.2 to 67%) but HT-noc, FEx and MFRD increase. These two last impact categories show a remarkable impact increase mainly due to the manufacturing and disposal of the photovoltaic plant. FEx is 3.67 and 2.98 times higher for FEED and FOOD scenario, respectively while MFRD is 12.57 and 7.86 times higher for FEED and FOOD scenario, respectively.

The variation in price of the earthworm meal (±30%) and the consequent variation in the allocation factors involves an impact variation that ranges from ±22.1% for TE to ±2.8% for FE. More in details, when the price varies, the impact grows more for the FEED scenario respect to the FOOD one.

The choice of a mixed FU to avoid allocation, as expected, deeply affect the environmental results. Without allocation, the environmental impact is no more divided between the two products (vermicompost and dried meal) but it is fully attributed to the mixed FU. Respect to the impact of 1 kg of dried meal assessed considering economic allocation, with the mixed FU, the higher impact increase occurs for the FEED scenario where the earthworm price is lower and, consequently, also the allocation factor assessed using the earthworm meal price is low. In the FOOD scenario, where already with economic allocation, the higher impact is attributed to earthworm meal, the use of a mixed FU is less impacting on the environmental indicators.

9.4 Discussion

Turning waste into a resource is part of 'closing the loop' in circular economy systems (EU COM, 2015). To this scope, earthworms play an interesting role as they are considered effective in organic waste transformation (Edwards, 1988). The earthworms process of FVW results in two excellent products: the vermicompost, a high-quality bioactive soil amendment, and the earthworms that are grown on FVW that and can be a new food/feed source, thanks to their high protein content. The ecological impacts of this process were estimated with the LCA method.

The environmental impact of the production of 1 kg of fresh earthworm (SS1) from FVW substrate and the subsequent dried process (SS2) to produce 1 kg of earthworm meal for feed purpose shows interesting outcomes; on CC the impact is 0.162 and 2.238 kg CO_2eq, for 1 kg of fresh earthworm and 1 kg of dried meal, respectively. This is mostly related to the emissions but also to the FVW transport to the plant and to the electricity use for the drying process in SS2. To reduce the role of transport activities on the sustainability assessment, vermicomposting process could optimally take place at the FVW production site. Regarding the reduction of the energy use, a photovoltaic system or other renewable energy sources could be introduced for reducing the impact of oven-dry meal production. In any case, more research on earthworms' meal production for feed purpose using FVW could permit achieving interesting further improvements for alternative protein sources in view of reducing the environmental impact for their production and the food waste (Conti et al., 2018).

Making even more sustainable the earthworms' meal and adopting it in livestock rations would also bring to a second subsequent beneficial effect. In fact, Europe's reliance on imported protein to feed livestock, especially soybean, is inconsistent with sustainability goals. Soybean production is associated, among others, with deforestation, use of pesticides and long-distance transportation (Tallentire et al., 2018; Thévenot et al., 2018) which bring to an environmental burden for 1 kg of soybean meal equal to 3.05 kg CO_2eq (Tallentire et al., 2017). Another issue to take into account is the total content of protein (or essential amino acids e.g. lysine) in the comparative emissions evaluation for future feed application of novel ingredients. The protein of earthworm meal ranged from 63.8 to 72.4% of the dry matter (Tava et al., 2018), higher than the protein of soybean meal, 43–44% of the dry matter (FEEDIPEDIA, 2019).

Considering the earthworm meal produced for food purpose, the environmental impact for 1 kg of fresh earthworm production (SS1) and the subsequent freeze-drying process (SS2) to produce 1 kg of earthworm meal still had interesting results, 0.593 and 5.944 kg CO_2eq for fresh earthworm and dried meal, respectively. Comparing earthworm meal for food purpose with other different food categories is only partially possible because of the different functional units adopted. Commonly, LCA studies on animal food products such as pork, chicken and beef adopted as functional units the kg edible meat from carcass or kg bone free meat (BFM). Non-ruminant livestock had average GWP values equal to 3.49 kg CO_2eq/kg BFM for fish, 3.65 kg CO_2eq/kg BFM for chicken and 5.77 kg CO_2eq/kg BFM for pork. Ruminant livestock show the highest average GWP values in lamb (25.58 kg CO_2eq/kg BFM) and in beef (26.61 kg CO_2eq/kg BFM) had (Clune et al., 2017). Referring these data to 1 kg of meat protein production, the highest impact is reported with beef (75–170 kg CO_2eq), followed by pork protein (21–53 kg CO_2eq), and finally by chicken protein (18–36 kg CO_2eq) (de Vries and de Boer, 2010).

Earthworm meal has a high protein content equal to 60–65% in the range of 54.6% to 71.0% dry matter (Zhenjun et al., 1997). Moreover, it is a low-demanding source of land and water use and it can be interesting in contrasting biodiversity loss. Thus, bioconverting FVW to produce earthworm meal has the potential to be considered an environmentally sustainable strategy.

9.5 Conclusions

Starting from a fruit and vegetable waste material, the goal was to outline innovative and sustainable models to design and develop more efficient regeneration and re-use systems. In this study, this is achieved by valorising organic waste and transforming it into high value-added products. The turning of FVW into earthworm meal could contribute to make more sustainable the protein production and to meet the future global protein demands.

By means of LCA method, the environmental impact of the production of earthworm meal was quantified with an attributional approach. Both 1 kg of fresh earthworm and 1 kg of dried meal were evaluated.

The feed substrate for earthworms is made of FVW so being highly valorized respect to wasting. Given the increasing importance worldwide of issues related to food waste, the transformation into feed and/or food meal is very promising.
Similarly, to other protein sources, earthworm meal currently has high environmental impacts mostly due to emissions during vermicomposting, transport of FVW for fresh earthworm production and energy use during processing. Freeze-drying instead of only oven-drying determines a higher environmental impact for the FOOD scenario respect to the FEED one. To make earthworm meal sustainable and competitive on the market, enhancing earthworm productivity and reducing energy consumption during processing by shifting towards renewable energy sources is essential. From a methodological point of view, the analysis highlighted that the choice made to solve the multifunctionality issue deeply affects the environmental results. In this regard, a comparison with future studies should be drawn using the same criteria for allocation or, even better, avoiding allocation by using a mixed functional unit.
Earthworms as feed protein ingredient can help to replace at least partially the use of soybean and fishmeal in animal nutrition or be used as feed additive, whereas the earthworm grown on a safe feeding substrate, can be an optimal food source or dietary supplement.
Additional research and integration with innovations among different sectors are the key drivers for the near future. However, the outcomes of this study can be useful for the development of a subsidy framework supporting the earthworm dried meal production chain thanks to the identification of the hotspot stages and their possible mitigations.

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10. Earthworms for feed production from vegetable waste: environmental impact assessment

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Abstract

In the European Union, 88 million tons of food is wasted annually, 30% of which comes from the production and processing sectors. Among the different food waste, vegetable ones represent a remarkable share and their management is complicated by the usually high-water content and the difficult storage. In this context, the earthworms are an interesting solution because transform vegetable waste into valuable products: the vermicompost, that can be sold as organic fertilizer, and the earthworms that, thanks to their high protein content can be used for feed and food production. This study aims to evaluate the environmental impact related to the production of vermicompost and dry earthworm meal. LCA approach was applied, 1 kg of dry meal for feed production was selected as functional unit. Inventory data were collected during experimental tests carried out in 2017 in a composting plant located in Northern Italy where earthworms were fed with vegetable waste. Secondary data were used about emissions during earthworms rearing. A quantity of 1 kg of fresh earthworms (16% of dry matter with 67% of protein content) and 13 kg of vermicompost were produced from 45 kg of vegetable wastes. Between earthworm rearing and processing, the first one is the main responsible for the environmental impact for all the evaluated impact categories except for freshwater eutrophication and ecotoxicity. GHG emissions during composting are the main hotspots for Climate Change.

Keywords: feed, protein source, vermicompost, waste valorization

10.1 Introduction

Food loss and waste have a negative environmental impact due to the natural resources used for food production as well as for their management and disposal. In the European Union, 88 million tons of food is wasted annually, 30% of which comes from the production and processing sectors. In particular, the fruit and vegetable retail sector generates large amounts of waste. In industrialized countries, fruit and vegetable waste (FVW) are mainly generated before reaching consumers, during all phases of the supply and handling chain, such as market oversupply or nonfulfillment of aesthetic and quality standards (Plazzotta et al., 2017). Even without official quality standards, food retailers generally do not offer food with abnormal appearance, based on the assumption that consumers do not purchase or consume foods that deviate from regular products, which can mean yielding lower profits (Loebnitz et al., 2015). For this reason, related to not reflecting aesthetic standards (shape, color or size), many products are discarded, even if they were produced for human consumption, they are still healthy, safe, and edible and could still reach the consumers (Stuart, 2009). FVW poses environmental problems due to the squandering of environmental, human and economic resources used to produce it and represents also a loss of valuable biomass (Plazzotta et al., 2017).

In order to reduce the impacts associated with food waste and to avoid the squandering of valuable resources, the search for sustainable solutions to the valorization of food waste is highly necessary and encouraged. A possible strategy is the utilization of FVW as feeding substrate for the rearing of terrestrial invertebrates to be used as potential protein source for feed and/or food supply chains. Among terrestrial invertebrates, earthworms could be an interesting solution. Earthworms grown on FVW can contribute to the waste disposal efficiency and bio-transform FVW into valuable products: the vermicompost, which can be sold as organic fertilizer, and the earthworms themselves that, thanks to their high protein content, can be a new food/feed source. Earthworms are rich in proteins, particularly in essential amino acids and they can contribute to human and animal nutrition (Yadav and Garg, 2011; Zhejun and Jiang, 2017). Currently, earthworms are just employed to convert food waste (FW) in a bioconversion process to mitigate the FW problem as a sustainable, cost-effective and ecological approach in dealing with FW management (Huang et al., 2016). Up to now, the attention on alternative protein sources has regarded mainly the insects both as human food (Halloran et al., 2016; Oonincx and De Boer, 2012; San Martin et al., 2016) and as animal feed (Smetana et al., 2016; Salomone et al., 2017). No studies addressed the environmental performances of dried meal production from earthworms.

In this context, this study aims to evaluate the environmental impact of the earthworms' dried meal production for feed purposes using fruit and vegetable waste (FVW) as feedstock. Primary data collected during field trials were combined with secondary data coming from literature; the environmental impact was quantified, and the environmental hotspots identified

10.2 Methodology

Life Cycle Assessment (LCA) is a holistic approach, structured and recognized worldwide that consists of a systematic set of procedures to convert inputs and outputs of the studied system into its related environmental impact (ISO 14040, 2006; ISO 14044, 2006).

In details, there are 4 steps in LCA:

(i) goal of the study definition that foresees the selection of the functional unit, the definition of the system boundary and the solving of multifunctionality;

(ii) Life Cycle Inventory (LCI) data collection, in which the flow of materials and energy from the studied systems and the environment are identified and quantified;

(iii) Life Cycle Impact Assessment; during which, thanks to specific characterization factors, the inventory data are converted in few numeric indicators of environmental impact;

(iv) interpretation of the results and identification of the process hotspots.

Over the last years, although originally developed for industrial processes, LCA has been more and more applied also to agricultural systems (Moudry et al., 2018; Schmidt Rivera et al., 2017) and waste to energy processes (Bacenetti and Fiala, 2015; Lijó et al., 2015; Vida and Tedesco, 2015) and waste treatment solutions (Bacenetti et al., 2016; Bjelic et al., 2017; Lijó et al 2017; Salomone et al., 2017; Smetana et al., 2016).

10.2.1 Goal and scope definition

The goal of the present study is to evaluate the environmental impacts of the earthworms' (*Eisenia foetida*) production system reared on a low-quality substrate made of fruit and vegetable waste (FVW).

Concerning the functional unit, in this study, to avoid allocation between vermicompost and earthworms dried meal, a mixed functional unit was selected. According to ISO standards for LCA (ISO, 2006), the functional unit is defined as the quantified performance of a product system, and is used as a reference unit in an LCA. In this study, the FU is the production of 1 kg of dried earthworm meal and 80 kg of vermicompost.

Concerning the system boundary, a "from cradle to gate" approach was applied. Fig. 1 reports the system boundary for the evaluated process; two different subsystems were identified:

- Subsystem 1 (SS1), a mix of young-non-clitellum and adult-clitellate earthworms was reared on a feeding substrate consisting of FVW and straw. FVW, constituted mainly by tropical fruits, was ground and then used as feed for earthworms three times a month. Besides earthworms, during the decomposition of FVW also an odour-free and hummus-like substance is produced: the vermicompost. Vermicompost is the co-product of the production system, it can be used as organic fertilizers. After 3 months of earthworms rearing, the vermicompost and the earthworms were collected through mechanical separation;
- Subsystem 2 (SS2), the collected earthworms were processed to produce meal. First, they were repeatedly washed, then kill by cooling and, finally, dried. During the experimental trials, the dry meal was produced in a laboratory by drying earthworms in an oven at 50°C and then proceeding with grinding.

The following activities were included: raw materials extraction (e.g., fossil fuels, metals and minerals), manufacture of the different inputs (e.g., diesel, electricity, water and trucks for FVW transport), use of the inputs (diesel fuel emissions), maintenance and final disposal of capital goods (e.g., the trucks used for the FVW transport). The emissions of methane, dinitrogen monoxide and ammonia related to the vermicomposting of FVW were included too. The packaging, the distribution as well as use and end-of-life of the produced meal were excluded from the system boundary.

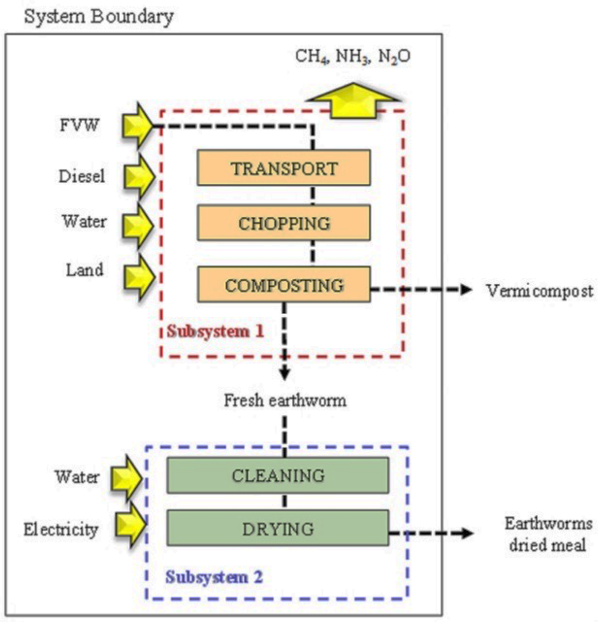

Fig. 1. System boundary

10.2.2 Description of the process

Earthworms were provided by a small-scale production system, located in the province of Lecco (North Italy). Earthworms were reared on an area of about 30 m^2 made up of FVW (growth substrate), placed above a non-woven textile sheet and covered with a net. During the rearing phase, moisture, temperature, and pH of the growth substrate were kept under control in order to guarantee optimal living conditions.

After 3 months, samples of *Eisenia foetida* at the adult stage of development were collected. The first cleaning procedure consisted of a mechanical separation from the growth substrate with the use of a trommel.

As the material rolls, anything smaller than the holes in the screen falls through, and the rest continues until it comes out the output end. Subsequently, they were washed with running tap water and soaked for some hours, to remove the residual particles of waste and to clear their gut. Finally, to produce the meal, after being frozen at -28°C, they were dried at 65°C and ground.

10.2.3 Life Cycle Inventory

Inventory data concerning inputs and outputs relevant to the production of earthworms' biomass were collected over a three-month experimental test performed in year 2017.

Primary data were collected with questionnaires during interviews with the farmer and during surveys to the experimental site. More in detail the following data were directly collected: amount of FVW used as feed, fossil energy for preparing the feed substrate, water volumes and land occupation for earthworms breeding. The main secondary data refers to the emissions during vermicomposting. These emissions were retrieved from literature (Yang et al., 2017). Table 1 reports the main inventory data for the analyzed production process.

Background data was retrieved from the Ecoinvent Database v.3.5 (Moreno Ruiz et al., 2018; Weidema et al., 2013).

Table 1. Inventory data

Subsystem	*Inputs/Outputs*	*Amount*
1	Fruit and vegetable waste	285.8 kg
1	Transport of FVW	25 km
1	Diesel	1.2 kg
1	Water	22.9 kg
1	Land	2.6 m^2
1	Ammonia	99.03 g
1	Dinitrogen oxide	9.56 g
1	Methane	31.60 g
2	Electricity	2.0 kWh
2	Water	22.4 kg
2	Vermicompost	80.0 kg
2	Dried meal	1.00 kg

10.2.4 Life Cycle Impact Assessment

The systems considered here have been modeled using SimaPro LCA software 8.05 and the impacts estimated according to the ReCiPe method (Goedkoop et al., 2009). The following 10 impacts are considered: Climate change (CC), Ozone depletion (OD), Terrestrial acidification (TA), Freshwater eutrophication (FE), Marine eutrophication (ME), Human toxicity (HT), Photochemical oxidant formation (POF), Particulate matter formation (PM), Metal depletion (MD) and Fossil depletion (FD).

10.3 Results and discussions

Table 2 reports the environmental results for the different evaluated impact categories while Fig. 2 shows the environmental hotspots (i.e. the inputs or emissions mainly responsible for the total impact).

The main environmental hotspots are:

- Diesel production: the consumption of diesel fuel for grinding a share of the FVW (e.g., pineapple leaves) is the main contributor for OD (72%) and FD (69%) while for the other evaluated impact categories it is responsible of a share of the total impact ranging from 10% in CC to 31% in POF;
- Transport of the FVW to the composting plant is the main responsible for HT and POF (51% and 52%, respectively, mainly due to the emissions of pollutants related to the diesel combustion) and MD (54%, mainly due to the manufacturing of the truck). Similarly, to the diesel production, the transport plays a non-negligible role for all the other evaluated impact categories (from 2% for TA to 21% in TE);

- Electricity consumption during earthworm processing in SS2 is the main contributor of TE (57%) and it is responsible for about one-third of HT. For the other evaluate impact categories the role of electricity ranges from 1.8% in ME and 18% in MD. For CC, the consumption of electricity; is responsible for 13% of the total impact;
- Ammonia emission during vermicomposting is the main contributor to TA (94%), ME (94%) and PM (85%, due to the formation of secondary particulate);
- Dinitrogen oxide emission deeply affects CC with 45% of the total impact.

With regard to the other inputs or emissions:

- The consumption of water in SS1 (for rearing humidity maintenance) as well as during SS2 (for cleaning) is responsible for a small impact (<2% for all the evaluated impact categories);
- The emissions of methane only slightly contribute to CC (about 12%) and POF (2.6%, due to the emission of CH_4).

Table 2. Environmental impact for the selected FU

Impact category	*Acronym*	*Unit*	*Score*
Climate change	CC	kg CO_2 eq	6.327
Ozone depletion	OD	mg CFC-11 eq	1.142
Terrestrial acidification	TA	kg SO_2 eq	0.257
Freshwater eutrophication	TE	g P eq	0.429
Marine eutrophication	ME	g N eq	9.637
Human toxicity	HT	kg 1,4-DB eq	0.618
Photochemical oxidant formation	POF	g NMVOC	12.332
Particulate matter formation	PM	g PM10 eq	36.997
Metal depletion	MD	g Fe eq	72.318
Fossil depletion	FD	kg oil eq	2.212

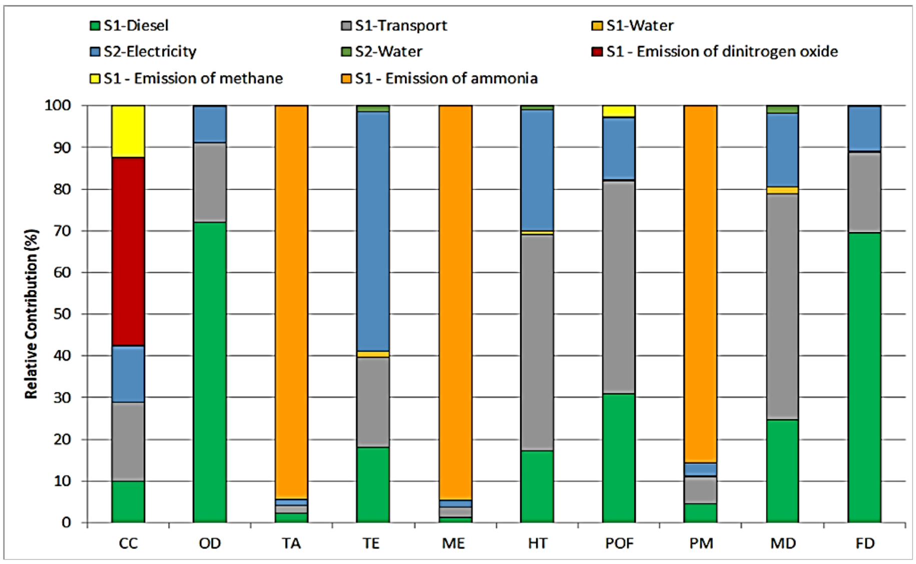

Fig. 2. Identification of the environmental hotspots (S1 = subsystem 1, S2 = subsystem 2)

Between the two subsystems, SS1, with a share of the total impact ranging from 70% in HT to 98% in TA and ME, is the main contributor for all the evaluated impact categories except than for TE. For this last impact category, 59% of the impact is related to SS2.

The environmental impact of the earthworms’ dried meal production for feed purposes using fruit and vegetable waste (FVW) as growth substrate showed a higher CC value associated with its production; this was caused by the considerable energy input for FVW transport and drying process. This could be reduced if the vermicomposting process takes place at the FVW production site. Moreover, Europe's reliance on imported protein, particularly soybeans, to feed livestock is inconsistent with sustainability objectives because soybean is associated with deforestation and impacts from pesticide use and transportation (Tallentire et al., 2018). The environmental burden for soybean meal is 3.05 kg CO_2 eq kg^{-1} (Tallentire et al., 2017). This means that improvements of the earthworms’ dried meal production for feed purpose using FVW could be a promising research field how even the necessity of alternative protein sources in terms of minor warming potential and reduction of food waste (Conti et al., 2018).

10.4 Conclusions

By means of the Life Cycle Assessment (LCA) method, the environmental impact of the production of earthworm-dried meal was quantified. The feed substrate for earthworms is made of fruit and vegetable waste (FVW) that, therefore, is highly valorised respect to wasting. Given the increasing importance worldwide of issues related to food waste, the transformation into feed and/or food meal is very promising. Besides the not negligible environmental impact, this production system brought benefits such as the recovery of FVW as feeding substrate, the earthworm production as a food/feed source with high nutritional profile, and the availability of vermicompost as an organic fertilizer that allows reducing the use of mineral fertilizers in other production systems.

Similarly, to other protein sources, earthworm dried meal currently has high environmental impacts mostly due to the transport of FVW for fresh earthworm production and energy use during processing. To make earthworm meal sustainable and competitive on the market, enhancing earthworm productivity and reducing energy costs of the processing stages by shifting towards renewable energy sources is essential.

Additional research and integration with innovations among different sectors are the key drivers for the near future. However, the outcomes of this study can be useful for the development of a subsidy framework supporting the earthworm dried meal production chain thanks to the identification of the hotspot stages and their possible mitigations.

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CHAPTER 7

11.General discussion and conclusions

This thesis deals with the air quality issue and the environmental impact of intensive livestock systems in the Po Valley area. Being the emissions from this activity one of the main responsible for the environmental impact of agriculture, particular attention is dedicated to the evaluation of different abatement systems to reduce emissions of NH_3 and VOCs from intensive livestock farming in the Po Valley area.

The investigation was conducted at several levels, firstly considering the contribution of livestock activities to air pollution (*Paper 1, 2 and 3*), and secondly taking into account two different aspects related to the applied mitigation systems: i) their odor, NH_3 and GHG removal efficiency (*Paper 4, 5 and 6*), ii) the quantification of their environmental performance through the Life Cycle approach (*Paper 5 and 6*). Finally, alternative proteins (i.e., earthworms) were considered to investigate their sustainability (*Paper 7 and 8*).

Besides being the major causes of NH_3 emissions, as confirmed by emissions and concentration levels also during the Covid-19 pandemic (*Paper 1 and 2*), agriculture and livestock farming represent an important source of odorant compounds. In Chapter 3, some interesting aspects emerged. NH_3 emissions and concentration in the Po Valley area were unaffected during lockdown periods imposed by Covid-19 spread. Regardless of the methodologies used (ground-based or satellite) no reduction effects of atmospheric NH_3 as a consequence of Covid-19 were observed, confirming the relevant role of livestock activities for this airborne pollutant. This situation leaves agriculture with extensive room for improvement, which could be partially achieved thanks to the application and adoption of mitigation strategies, applicable at all levels of agriculture and livestock management.

Although agriculture is recognized as the main responsible for NH_3 emissions, it largely contributes also to odor emissions, causing odor annoyance to nearby residential areas. In Chapter 4 (*Paper 3*), the literature reviewed highlighted that odor needs to be considered an airborne pollutant, such as GHG and NH_3, due to its negative effect on the environment and human health. To quantify odor annoyance, an integrated approach combining sensorial or analytical techniques with air dispersion models could provide complete information on "odor issue" (quantitative, qualitative, and assessment of odor impact). Indeed, each of them presents different advantages. Approaches based on chemical measurements of tracer compounds in the environment entail the advantage of being easier and more reliable than the measurement of low odour concentration values by dynamic olfactometry. Conversely, sensorial techniques entail the double advantage of allowing the direct determination of ambient air concentration close to the odour detection threshold and of presenting higher sensitivity. However, the evaluation of odor concentration alone is not sufficient for the simulation of dispersion of any pollutant. Odor dispersion models take into account also the air flow associated with the monitored odor source, thus allowing to simulate how odour disperses into the atmosphere, and, finally, calculate ground odour concentration values in the simulation space-time domain. These represent a particularly useful tool to investigate the efficiency of mitigation strategies.

Although odors occupy a leading position among air quality issues, at the moment, the quantification of the odor impact from an environmental point of view is still missing. Some authors tried to include an Odor Impact Potential in Life Cycle Assessment, but the methodology is still critical. A specific procedure to calculate odor impact and a dedicated indicator does not exist, besides it is difficult to quantify its contribution to the other impact categories (Cadena et al., 2018; Peters et al., 2014). It should be useful providing a standardize definition of odor impact. Also other airborne pollutants, such as NH_3, have a characteristic odor but their environmental impact is already quantified by different impact categories (e.g. eutrophication, acidification, and particular matter formation). Environmental impacts occur on “objects” or “areas” we wish to protect: human health, ecosystem functions and natural resources. To protect people from odor annoyance a wide range of mitigation techniques is available but further research is needed to explore efficient and cost-effective management systems other than to ensure effective implementation. Paper 4 was dedicated to the quantification of the mitigation effect of two air treatment systems installed in a pig farm. Specifically, a dry filter and a wet scrubber prototype were tested. The odor removal efficiency was quantified with dynamic olfactometry. The adopted abatement technologies affect odor concentration, even if some trade-off between the systems emerged. Wet acid scrubber constitutes a good choice to control odor, especially if installed in mechanically ventilated buildings as underlined by (Melse and Ogink, 2005; Van der Heyden et al. 2015). Instead, the dry filter had limited to no effect. This preliminary result could be promising considering that the two air treatment technologies were tested in a naturally ventilated building, a typical situation of most Italian pig farms, and also considering that the wet acid scrubber used a citric acid solution instead of a sulphuric one, traditionally used. In Paper 5 a full LCA study was conducted to evaluate, for the same intensive pig farm, the potential reduction in environmental impact linked to the installation of a wet acid scrubber prototype that used acid citric solution. Scrubbers usage in pig farms located in North Europe countries, such as Belgium and Netherlands, is well consolidated (Zhuang et al., 2019), but, based on authors knowledge, they have never been tested in an Italian context with naturally ventilated buildings. Assuming a 60% NH_3 abatement efficiency, findings outlined that the use of the wet acid scrubber leads to an impact reduction for all the impact categories influenced by NH_3 (acidification, eutrophication, and PM formation), confirming that it represents an effective strategy to reduce the environmental impact of heavy pigs for NH_3-related impact categories. Indeed, the other impact categories, such as climate change, ozone layer depletion, photochemical oxidant formation, toxicity-related impact categories, and mineral, fossil and renewable resource depletion, were worsened by the consumable materials for scrubber operation (energy, citric acid, and water) and construction. A further small impact reduction could arise by realising the fertiliser value of ammonium citrate salt (formed by the reaction between NH_3 and citric acid) as nitrogen fertilizer could further reduce the environmental impacts due to the replacement of mineral fertilizer. In addition, the field application of the discharge water is another valuable strategy to reduce the use of mineral fertilizer, as demonstrated by de Vries and Melse (2017). Finally, the development of artificial intelligence able to manage the activation of the abatement system based on NH_3 levels recorded inside the farms would help farmers to monitor pollutants and to control the environmental impact without unnecessary operation. This last aspect will be evaluated during the third year of the APPROAch project from which data were collected for the LCA study.

Emissions could be mitigated with different technologies, air treatment systems could affect air quality both outside and inside livestock buildings, but considering that manure management is a considerable source of GHG and NH_3 emissions, various practical options could be implemented, such as usage of additives or of covering systems during slurry storage, slurry treatments (e.g., anaerobic digestion and acidification) and others (Finzi et al., 2019). In the third article included in Chapter 5 (*Paper 6*), the LCA was used to evaluate the potential environmental impact and benefits associated with the adoption of a commercial additive during slurry storage. With this strategy NH_3 and GHG emitted during slurry storage could be decreased. Although the use of additives is still controversial as some provide a short-term efficacy and/or require frequent reapplication, with high costs (McCrory and Hobbs 2001; Wheeler et al. 2011), different authors reported beneficial effects of gypsum (Febrisiantosa et al., 2018; Lim et al., 2017). It has a great capacity to prevent N loss from manure, reduce odor and especially NH_3 volatilization (Febrisiantosa et al., 2018; Li et al., 2018; Tubail et al., 2008). Moreover, the acidification of the slurry obtained by the addition of gypsum seems to favor the reduction of CH_4 and N_2O emissions (Berg, et al. 2006; Hao et al. 2005; Luo et al. 2013). The above-mentioned mitigation effects were also confirmed by the LCA study. Being the additive capable of partly reducing GHG and NH_3 emissions, also GHG and NH_3-related impact categories were beneficially affected.

The outcomes of these studies could be useful for farmers and their associations to understand the actual environmental impacts of livestock production and the consequences and benefits arising from the reduction of airborne pollutants emissions. Moreover, they can be useful for companies and technicians working with farmers to offer innovative solutions.

Mitigation strategies are not always sufficient to improve livestock environmental sustainability. Some emissions, such as CO_2 direct emissions from the respiratory process or CH_4 emissions from enteric fermentation could not be cut-off. At the same time, agriculture and livestock activities are asked to improve their sustainability even if they should produce more to satisfy the increasing global food demand. Indeed, the world population is rapidly growing and current food production systems may not sustain both the projected world population and the projected consumption patterns of meat products, in particular in the context of climate change. Although entomophagy is slowly gaining mainstream visibility, and terrestrial invertebrates are already consumed in many countries, this practice remains marginal in Western countries. Other having a high protein content, they could represent an advantage compared to traditional livestock farming from an environmental point of view (van Huis and Oonincx, 2017). They require less space, energy and water and produce fewer emissions. Consumers acceptability can be facilitated in different ways: i) propose insects processed or as ingredients inside familiar food items (Caparros Megido et al., 2016; Gmuer et al., 2016; Tan et al., 2015); ii) improve information and awareness of future global food demand; and iii) enhance nutritional characteristics of insects (Tan et al., 2015). In Chapter 6 the environmental sustainability of an alternative protein source (i.e., earthworms) was assessed. Earthworms could grow on fruit and vegetable waste (FVW), avoiding the squandering of valuable resources, contributing to the waste disposal efficiency and bio-transforming FVW into a valuable product: the vermicompost. Moreover, according to the processing method (drying or freeze-drying) earthworms could be used as a feed or food protein source. At the moment, the main limit to their "environmental sustainability" is linked to three processes: i) the consumption of diesel fuel for grinding a share of the FVW, ii) the transport of FVW, and iii) the electricity consumption during earthworm processing into meal. Regarding the first aspect, various substrates could be studied, thus avoiding the grinding process. The impact related to the transport could be reduced if the vermicomposting process takes place at the FVW production site. Finally, to make earthworm meal sustainable and competitive on the market, enhancing earthworms' productivity and reducing energy costs of the processing stages by shifting towards renewable energy sources is essential. For the moment, based on Paper 7 and Paper 8 results, earthworms are more sustainable if used as a feed protein source. This limits the problem of consumers' acceptability.

In conclusion:

- Livestock farming represents an important source of airborne pollutants emissions, that could be reduced thank to the application of abatement measures. Mitigation strategies are available for all steps in the sequence of animal production and manure management, i.e. feeding, housing, manure storage, and spreading. For every strategy, it is necessary to consider that the desired mitigation level, as well as the applicability, are strictly dependent on animal species, housing conditions, and manure management, besides being influenced by external factors, such as weather. Also, it must be accepted that only a partial reduction of pollutants emissions can be achieved. For example, a strategy effective in abating NH_3 emissions may worsen GHG and vice-versa. So, a combination of solutions is often needed to control the whole-chain GHG, NH_3 and odor emissions.
- Environmental benefits are achievable by implementing mitigation options. Each strategy can affect different impact categories; wet acid scrubber, for example, have proved to be effective on NH_3-related impact categories, but unfavorable on the others. Finally, LCA could help in identifying the processes that contribute most to the environmental impact of each specific livestock farming system. This result stresses once again that the modulation of environmental impact of livestock farming can succeed only through the adoption of a proper combination of measures.
- With the booms of the world's population and the increasing intensification of animal husbandry, the demand for addressing airborne pollutants emission in livestock farms will increase. Thus, new technologies are required to increase resource efficiencies at the global level, improving at the same

time environmental sustainability. From a policy perspective, a mix of regulation, incentives and education are likely to be necessary to support the implementation of interventions. In this context, authorities should urgently assume their responsibilities by orienting and supporting the appropriate and sustainable foodstuff productions and consumptions. Sustainable food policies should consider, in a coherent manner, both agriculture and the health sector, with the aim to ultimately benefit agriculture, human health and the environment. Earthworms and insects could be included among "livestock" sustainable solutions.

11.1 Limitations and opportunities for further studies

For each paper, the limitations and opportunities for improvement can be resumed as follow:

- In Paper 1 and 2, NH_3 emissions and concentrations were compared in the period (i.e., from year 2013–2019) before the lockdown determined by the emergency of Covid-19 and the pandemic period (i.e., year 2020, with the strict lockdown lasting from March–June 2020), adopting two measurement systems, ground-based units and satellites. The two techniques showed similar patterns, such as the higher yearly average concentrations in country zones. However, some limitations emerged, such as some differences for individual months and in the relative contribution of the different zones to total NH_3 concentrations. Moreover, for the entire Lombardy region (surface area about 23,000 km^2), the availability of NH_3 measured data analyzed in the two Papers was limited to 6 and 10 ground-based control units, respectively. In addition, for the same control units, data of different pollutants (e.g., NH_3, NOx, and PM), were not always available, making comparisons more difficult. As concerns satellite images, to avoid data with high uncertainty, it was not possible to create a grid finer than 0.5 × 0.5°. Also, the comparison between data collected with ground-based units or satellites was complicated basically for the different units of measure and assessment method but even for the different spatial resolution. Given the limitations of both measurement approaches, among which the temporal and spatial resolution, improvements should be introduced both in the number and density of ground stations, especially in areas where agriculture is widespread, and in the availability of satellite data with a higher spatial resolution, which might also make validation efforts easier in comparison with ground observations. Finally, to confirm that agriculture is the main responsible for NH_3 emissions, other areas characterized by a large concentration of intensive livestock farms and agricultural activities could be included in future studies.
- In Paper 3, a literature review of odor measurements and major sources of complaints was carried out to point out the state of the art of this concern with a focus on odors from waste management and livestock farming. For quantitative and qualitative characterization of odors, both sensorial and analytical techniques can be used, in alternative or combination. The choice of the appropriate

technique should be made considering the individual situation, the objective of the study, and the advantages and disadvantages of each one. Regardless of the measurement technique adopted, the quality of the results obtained is heavily dependent on appropriate sampling, which is one of the main issues relating to odor characterization and measurement. If the goal of the study is to determine the odor plume extent, the evaluation of odor concentration alone is not sufficient, but air dispersion models should be applied. To be performed these models require topographic/orographic, meteorological and emission data, which goodness affects the quality of the results. Based on the model applied (i.e., Gaussian, Lagrangian or Eulerian) an underestimation or an overestimation can be obtained, which is also affected by the quality of the input data. Concerning the livestock sector, specific odor dispersion models should be developed to include all the specific features of livestock odor dispersion (e.g., short distance of transportation, multiple sources, animal mass, and number). As the extent of odor emissions depends on different factors, odor emission reduction can be obtained thanks to the application of one or more mitigation strategies. Some solutions have been longer investigated and are well known (e.g., scrubber, additives, manure covers, etc.), while for others (e.g., activated carbon adsorption and activated sludge diffusion) further research is needed to explore efficient and cost-effective management systems other than to ensure effective implementation. Since odors have such a big impact on the surrounding environment, it would be useful to quantify the nuisance also from an environmental point of view. Thus, it is reasonable to integrate odors as an indicator to be used in an LCA framework, and consequently, to develop a specific impact category. In fact, up to now, although the different compounds responsible for the odor, such as NH_3 and VOCs, affect some impact categories (e.g., eutrophication, acidification, and particular matter formation) an impact category specifically referred to the odor is missing. Future studies should focus on this aspect trying to propose an approach that combining LCA methodology and exiting sensorial/analytical techniques could "environmentally" quantify the "odor impact".

- In Paper 4, the odor abatement efficiency using two different abatement technologies was evaluated with dynamic olfactometry methodology. The first limit of the study was linked to the two air treatment systems tested. Indeed, the dry filter was a technology already used in the industrial context to mainly abate dust particles and not odors, even if it is known that some odorous compounds are transported by dust, whereas the scrubber was a prototype with an acid citric solution. Scrubbers with sulphuric acid are generally applied, especially in Dutch countries where sulphuric acid is the only acid admitted by national regulations. Moreover, the trial was conducted in a naturally ventilated pig facility, although scrubbers are usually applied in forced ventilation buildings to reach higher removal efficiencies. This condition was tested as fattening cycles in naturally ventilated buildings represent a traditional Italian farming system. Despite these limits, the two air treatment systems were used inside the barns thus allowing to improve indoor air quality and not only to abate outside emissions. Concerning the methodology used, despite dynamic olfactometry is regulated by the European Standard EN 13725:2004, which requires the panel to be tested with *n*-butanol as a standard reference odorant, this gas does not reflect the characteristics and intensities of odors associated with pig farming. Consequently, knowing the major odorous compounds associated with swine facilities, using for example gas chromatography coupled to mass spectrometry (GC-MS), and training panelists to recognize "pig odor" could improve the accuracy of olfactometric sessions. Moreover, GC-MS could help understand how the abatement technologies affect individual odorous compounds, such as organic or inorganic molecules. To improve the accuracy of these preliminary results, collecting odor samples also in other seasons, with different weather conditions and pig weights (fattening phase starts at 60 kg live wight and ends at 160 kg), could provide a more complete range of achievable abatement efficiencies.

- The same wet scrubber prototype studied in Paper 4, was considered also in Paper 5 to evaluate the environmental performance of a typical Italian pig rearing system where this air treatment technology was applied with the aim of reducing NH_3 emissions from livestock housing and the related environmental impacts. As in the previous paper, a prototype was tested. Since experimental results on the actual effectiveness of NH_3 removal were still not available, a 70% reduction of NH_3 emissions was considered as in some Western-European countries this is the required minimum removal efficiency in pig houses. The inventory data related to scrubber energy, water, and acid consumption as well as raw materials and energy needed for the construction of the machinery were taken from the literature, but the use of primary data is to be preferred as reflects the effective situation and achievable environmental benefits. Although promising preliminary results were achieved for NH_3-related impact categories, there is still a big margin of improvement to reduce NH_3 emissions and enhance farm sustainability. Future studies could take into consideration the adoption of different mitigation strategies, as well as improvements in this considered. As an example, for the wet acid scrubber, water and acid citric solution recirculation could represent a margin of improvement also for non-NH_3-related impact categories. Moreover, in this preliminary study, the wet acid scrubber was considered to be working 24/7, but in the next steps of the APPROAch project, its operation time will be regulated by an air quality monitoring tool that will turn on the scrubber only when NH_3 indoor recommended threshold limits will be exceeded, thus reducing energy consumption.
- Regarding Paper 6, a first limitation of the study was the short time testing period of the additive, instead of considering a greater period ranging from 90 to 150 days as recommended for Italian non-vulnerable and vulnerable zones, respectively. Although the preliminary results are promising, a longer evaluation could have also provided a better understanding of slurry properties and on

emission reduction. For this reason, in the LCA study 180 days were taken into account for the modeling of emissions to be more comprehensive of the different storage recommendations. Secondly, it is necessary to consider that the results achieved were obtained with a much lower amount of product than that proposed in other studies for other amendments, so also considering different dosage and application periods, could have affected the final abatement efficiency. Finally, pilot and large-scale tests could be interesting to investigate the effectiveness and economic feasibility of this method also in other types of slurry management systems.

- Finally, regarding the last two Papers (*Paper 7 and 8*), to the authors' knowledge, they represent the first LCA studies of earthworms rearing. Other having a good nutritional profile, earthworms can be fed on different types of waste and the produced vermicompost is an excellent high quality bioactive amendment to improve soil fertility. For the moment, the main limit to earthworm consumption is the consumers degree of distaste. To enhance consumers' willingness to taste foods containing terrestrial invertebrates, minced or powdered earthworms need to be incorporated into ready-to-eat preparations, as Western countries people are not ready to add earthworms/insects to their diets in "whole form." However, their processing worsens the environmental impact. Alternative processing methods could be considered to compare the final environmental results. Further improvement could be made by shifting towards renewable energy sources during the transformation phase.

In the future, a significant reduction in livestock emissions is expected as a consequence of the abatement measures that are being introduced in farms (e.g., closed tank storages, manure and slurry treatments, precision application of slurry on the field). Thanks to improvements introduced in the European Union, NH_3 was already reduced by 24% from 1990 to 2017. Therefore, supporting farmers in this direction is fundamental to increase the spread of such ameliorative techniques and solutions and it is also of great importance from an environmental point of view to avoid damages to ecosystems.

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12. List of publications

12.1 Publications in peer review journals

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