A versatile method for groundwater vulnerability projections in future scenarios

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**Abstract** 

Water scarcity and associated risks are serious societal problems. A major challenge for the future will

be to ensure the short-term and long-term provision of accessible and safe freshwater to meet the needs

of the rapidly growing human population and changes in land cover and land use, where conservation

and protection play a key role. Through a Bayesian spatial statistical method, a time-dependent

approach for groundwater vulnerability assessment is developed to account for both the recent status

of groundwater contamination and its evolution, as required by the European Union (Groundwater

Directive, 2006/118/EC). This approach combines natural and anthropogenic factors to identify areas

with a critical combination of high levels and increasing trends of nitrate concentrations, together with

a quantitative evaluation of how different future scenarios would impact the quality of groundwater

resources in a given area. In particular, the proposed approach can determine potential impacts on

groundwater resources if policies are maintained at the status quo or if new measures are implemented

for safeguarding groundwater quality, as natural factors are changing under climatic or anthropogenic

stresses.

Keywords: Groundwater vulnerability, Time dimension, Statistical method, Land use management

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### 1 Introduction

Water crises are ranked among the most important global risks likely to occur with severe social, economic and environmental impacts, mainly due to the increasing demand from population growth, intensive land management, and economic expansion (World Economic Forum, 2015).

One of the major future challenges will be to ensure the short- and long-term provision of accessible and safe freshwater to meet the needs of the fast growing human population and changes in land use, where conservation and protection have a key role (Kumar, 2015). Rapid land use change can modify the amount and distribution of point and non-point sources of contamination, while climate change may significantly impact water availability and quality (IPCC, 2012). In this context, groundwater represents the most strategically important water resource, which could be less impacted by both climate change and contamination compared to surface water. Nevertheless, groundwater is certainly vulnerable to contamination with increasing risks in changing environments (Rebelo et al., 2014). Thus, there is an urgent need to better quantify time-dependent processes that can affect groundwater quality (Stevenazzi et al., 2015) and to develop methods to evaluate the effects of future scenarios on groundwater vulnerability.

The objective of this study is to develop a time-dependent approach for groundwater vulnerability assessment to account for the recent status and the evolution of groundwater contamination, as required by the European Union (Groundwater Directive, 2006/118/EC). Such an approach also allows a quantitative evaluation of future potential scenarios and their impacts on groundwater quality. Specifically, this study focuses on the status of nitrate contamination in 2011 and its evolution in the decade 2000 to 2011, and assesses groundwater vulnerability due to natural and anthropogenic factors and their evolution in the decade of the 2000s. The future scenario will be referenced to year 2020, and consider changes in anthropogenic sources of nitrate and natural factors observed in the period 2011 to 2020.

The development of new techniques (i.e., statistical or processed-based methods) to estimate groundwater vulnerability introduced questions as to whether vulnerability or risk is assessed. In some cases, no distinction is made between specific vulnerability and risk assessment, with hazard types, distribution, loading, and transport all included at the risk-assessment stage (e.g., Focazio et al., 2002).

Without unambiguous and universally accepted definitions of vulnerability and risk in the assessment methods, this study follows the same terminology used in previous studies on groundwater vulnerability assessment using spatial statistical techniques (e.g., Arthur et al., 2007; Sorichetta et al., 2011). Hence, the term "groundwater vulnerability to nitrate contamination" will be used. In addition, the term "severity" is used in this paper to describe areas where the probability of groundwater quality deterioration occurring in the future is higher or lower compared to the previous conditions.

#### 2 Materials and methods

#### 2.1 Study area

The study area is in the Po Plain area of Lombardy Region (Fig. 1), covering a mainly agricultural area of 13,400 km<sup>2</sup>. Urban areas, covering about 22 % of the area, have consistently increased due to urban sprawl since the beginning of the 2000s, while the population did not follow the same growth (EEA, 2006).

This region is surrounded by important rivers that influence groundwater flow in the unconfined aquifer (Fig. 2a): the Po River to the south, Ticino and Sesia Rivers and another small section of the Po River to the west, and Mincio River to the east. It is also constrained by mountain chains forming the boundary of the plain: the Lombardy Prealps along the north and the Appennines along the southwest. This area has a complex hydrogeological setting consisting of multiple aquifers with various properties and interactions. Plio-Pleistocene sediments, the upper unit of which forms shallow unconfined aquifers, characterize the subsoil of the Lombardy plain. Sediments are mainly gravels and sands, although the presence of finer sediments increases from the north to the south where shallow aquifers are mainly constituted by fine sands and are partially confined. These aquifers have high transmissivity ranging from  $10^{-2}$  to  $10^{-4}$  m<sup>2</sup>/s, and medium-high hydraulic conductivity ranging from  $10^{-4}$  to  $10^{-6}$  m/s; while average thickness ranges from 40 to 80 m (Regione Lombardia and ENI, 2002; Fig. 2b).

The groundwater flow is generally oriented north–south toward the base level defined by the Po River, with a deviation to the east-south-east in the south-eastern area of the plain. The groundwater depth decreases from north to south, ranging from values higher than 70 m to less than 2 m. There are also some groundwater-fed streams, where the local groundwater depth reduces to zero. During the last 15

years, the groundwater level has remained steady in most of the study area with some sectors characterized by an increase in the groundwater level after 2010 (Fig. 2).

#### 2.2 Weight of Evidence technique

The Weight of Evidence (WofE) is a statistical based technique that has proven to be a useful and reliable method to assess groundwater vulnerability (Arthur et al., 2007; Masetti et al., 2007, 2008; Sorichetta et al., 2011, 2012; Uhan et al., 2011). WofE is a pixel-based modeling technique, which combines different spatial datasets in a GIS environment to analyze and describe their interactions and generate predictive patterns (Bonham-Carter, 1994; Raines et al., 2000). WofE can be defined as a data-driven Bayesian method in a log-linear form that uses known occurrences (training points) to generate predictive probability maps (response themes). This is done accounting for multiple weighted evidences (evidential themes representing explanatory variables) related to the spatial distribution of the occurrences in the study area (Raines, 1999).

In groundwater vulnerability assessments of a specific region, the training points are a selected subset of the monitoring well network in the area, the evidential themes are natural and anthropogenic factors influencing groundwater vulnerability, and the posterior probabilities represent the relative groundwater vulnerability of the area.

Please refer to Arthur et al. (2007), Uhan et al. (2011), Sorichetta et al. (2012), and Stevenazzi et al. (2015) for a detailed description of how WofE has been used for assessing groundwater vulnerability in different study areas. We note here several relevant concepts:

- The response theme is generated using the training points, and calibration and validation are performed using a combination of training and control points and the control points alone, respectively.
- The prior probability represents the probability that a pixel within the study area contains an occurrence without considering any evidential themes, and is given by the ratio between the number of pixels containing a training point and the total number of pixels of the area.

- Each evidential theme is generalized in statistically significant classes, by assuming a minimum confidence value of |1.282| corresponding approximately to a 90% level of significance, and a pattern distribution justifiable from a hydrogeological point of view.
- The posterior probability represents the relative probability that a pixel contains an occurrence based on the evidences provided by the evidential themes.

In this study, the WofE response themes are generated using the Spatial Data Modeler (Sawatzky et al., 2009) for ArcGIS 9.3 (ESRI, 2008).

### 2.3 Monitoring network and explanatory variables

Nitrate is one of the most common contaminants found in groundwater in the study area (Cinnirella et al., 2005), and its presence has been monitored via a network of about 500 wells since 2001. Data were collected every 6 months from 2001 to 2012 (Regional Environmental Agency – ARPA, unpublished data, 2013). From the network, only the wells monitoring the shallow aquifer and having a minimum of eight measurements are selected (Grath et al., 2001, Fig. 1c).

In the conceptual hydrogeological model, six explanatory variables are considered as factors influencing groundwater vulnerability to nitrate contamination in the study area. These explanatory variables, representing both natural and anthropogenic factors (Tab. 1) were derived from multiple sources of information. These variables were selected to capture the pathway and the main regional-scale processes that characterize the nitrate contamination pattern. This includes: potential release from the surface (sources) and eventual degradation through superficial soils (soil protective capacity), vertical spreading (hydraulic conductivity of the vadose zone) to the saturated zone (groundwater depth), and transport and dilution in the aquifer (groundwater velocity). The importance of each variable in influencing groundwater vulnerability can differ according to its local spatial relation with the other variables.

Table 1 – Explanatory variables used as evidential themes.

Explanatory variable	Туре	Range
QuikSCAT-DSM [2000, 2001,, 2009] (dB)	Continuous	-11.91 ÷ -2.53
Slope QuikSCAT-DSM [2000 ÷ 2009] (dB/year)	Continuous	-0.0699 ÷ 0.1268
Nitrogen fertilizer load [2002] (kg/ha/year)	Continuous	0 ÷ 767
Nitrogen fertilizer load [2010] (kg/ha/year)	Continuous	0 ÷ 664
Soil protective capacity	Categorical	Low, Moderate, High
Groundwater depth [2003] (m)	Continuous	0 ÷ 70
Groundwater depth [2014] (m)	Continuous	0 ÷ 75
Groundwater velocity (m/s)	Continuous	$4.7 \times 10^{-8} \div 7.3 \times 10^{-5}$
Hydraulic conductivity of the vadose zone (m/s)	Continuous	$4.1 \times 10^{-8} \div 4.0 \times 10^{-2}$

Natural factors characterizing geological and hydrogeological conditions in the study area, with the exception of groundwater depth, are considered to be time-independent in this study. The soil protective capacity map was prepared by the Agency of Services of Agriculture and Forest, at a 1:250,000 scale, and assigns soil to three protective capacity classes: high, moderate and low.

Groundwater depth was derived from the difference between the topographic level and groundwater piezometric levels from two regional surveys conducted in 2003 and 2014. In both surveys, groundwater depth decreases from north to south, ranging from values higher than 70 m to 0 m. Groundwater piezometric levels (Fig. 2a) from several wells in the network show that groundwater depth, albeit a seasonal variability, has not significantly changed over the Po Plain between 2000 and 2010. At the beginning of 2010, in some sectors of the study area, an increase of the piezometric levels occurred (Fig. 2c, e), while in other sectors the piezometric levels remain stable (Fig. 2d, f). Thus, the piezometric levels map from the 2003 survey is used for the analyses related to the period 2000 to 2010 while the map from the 2014 survey is used to evaluate the groundwater depth in the period 2011 to 2020 as described in § 4.2.

Groundwater velocity was estimated by interpolating hydraulic conductivity values obtained from pumping tests, executed in more than 1,200 wells, together with the local hydraulic gradient derived from the regional groundwater potentiometric surface (Fig. 2a). Hydraulic conductivity of the vadose zone was determined from the interpolation of hydraulic conductivities calculated via the equivalent

vertical permeability method (Anderson and Woessner, 1992), applied to more than 1,500 well stratigraphy records. Thickness of the vadose zone in each well was evaluated as the difference between the topographic surface and groundwater potentiometric surface, both expressed in m a.s.l.

Anthropogenic sources of nitrate contamination can be associated with both urban (leakages from the sewage system or septic tanks) and agricultural sources (fertilizers and manures). Urban areas derived from satellite radar remote sensing are used as a proxy to represent urban nitrate sources (Stevenazzi et al., 2015). Such a dataset, with a pixel posting of 1 km², was obtained by using the Dense Sampling Method (DSM, Nghiem et al., 2009) to process the data collected by the SeaWinds scatterometer aboard the QuikSCAT satellite (QuikSCAT) in the decade of the 2000s.

The Agency of Services of Agriculture and Forest, which regulates the amount of fertilizers and manures sold to farmers every year in each district of the region, provided the nitrogen loadings data for this study.

### 3 Spatio-temporal approach

## 3.1 General aspects

To obtain a model that can account for both the last status of nitrate contamination and its evolution, the following steps were implemented (Fig. 3):

- 1) A spatial model is developed to represent the status of nitrate contamination and nitrate sources at time  $t_1$ . It identifies areas where the combination of natural and anthropogenic factors results in nitrate concentrations higher than a specific value.
- 2) A temporal model is constructed to account for the evolution of both nitrate contamination and nitrate sources between  $t_0$  and  $t_1$ . It identifies areas where the combination of natural and anthropogenic factors results in an increase of nitrate concentration.
- 3) A spatio-temporal model is generated to capture both the status at  $t_1$  and the evolution between  $t_0$  and  $t_1$  of nitrate contamination and nitrate sources. It identifies areas where the combination of natural and anthropogenic factors causes the presence of nitrates and their increasing concentration trends in groundwater.

- 4) A predictive spatio-temporal model to project a potential status of nitrate contamination and nitrate sources at time  $t_2$  and their evolution between  $t_1$  and  $t_2$ . Various scenarios can be considered with: (a) a different evolution of nitrate contamination sources (e.g., different growth rates of urban areas), or (b) a variation of hydrogeological conditions (e.g., groundwater depth variations).
- 5) The spatio-temporal (3) and the predictive spatio-temporal (4) models are compared in order to determine the differences between the results at  $t_1$  and at  $t_2$ .
- 6) A severity model of groundwater contamination is developed by combining the comparison model (5) and the spatial model (1). It identifies severity areas where the combination of natural and anthropogenic factors involves both the presence of nitrates and their increasing contamination in groundwater at t<sub>2</sub>, considering the vulnerable areas at t<sub>1</sub>.

The spatial model and the temporal model have been already developed and presented, with various examples within the Po Plain area of the Lombardy Region (Masetti et al., 2007, 2008, 2009; Sorichetta et al., 2011, 2012, 2013; Stevenazzi et al., 2015). Note that only the results of the spatial model can be compared with those obtained via other methods for evaluating groundwater vulnerability, since none of them uses the time dimension in the analyses. In this regard, various authors (Worrall and Besien, 2005; Twarakavi and Kaluarachchi, 2006; Uhan et al., 2011) showed how the high flexibility of statistical methods allow more reliable "spatial" groundwater vulnerability assessments over large areas compared to the ones obtained with other methods.

## 3.2 Response variable

WofE requires a binary formulation of the response variable. Thus, it is necessary to establish a threshold value to distinguish between occurrences and non-occurrences (a training and a control set, respectively). The response variable is represented either by nitrate concentration in groundwater at time  $t_1$ , by the nitrate concentration trend from  $t_0$  to  $t_1$  or by their combination as described below. In the spatial model with respect to a specific time  $t_1$ , the value representing the inflection point of the cumulative probability plot of nitrate concentrations is selected as an appropriate threshold (Masetti et al., 2009). Thus, wells with a nitrate concentration higher than the threshold value represent the training

set, and they are selected to produce the post probability maps. In contrast, wells with a nitrate concentration lower than the threshold value form the control set.

The temporal model considers the evolution of nitrate concentrations. The change in nitrate concentration is quantified by the slope of the regression line from the interpolation of concentration data. Thus, following Stevenazzi et al. (2015), wells with a clear increase in concentration trends (positive slope values) represent the training set, and wells with a decreasing trend or a trend close to zero (non-positive slope values) represent the control set.

In the spatio-temporal model, the training set is identified to account for both the nitrate concentration measured in the wells and the nitrate concentration trends expressed as the slope of the regression line from the interpolation of concentration data. Each well in the network is classified based on its nitrate concentration at time  $t_1$  and its contamination trend from  $t_0$  to  $t_1$ . Eleven classes of nitrate concentration and five classes of contamination trend are defined for the analysis (please refer to the Supplementary Material for additional information regarding the classification systems). Then, to rank each well with respect to both its concentration and trend class, the following formula is used (Stillwell et al., 1981):

$$R = \frac{(n - r_j + 1)}{\sum_{j=1}^{n} (n - r_j + 1)}$$
 (1)

where n is the number of classes and  $r_j$  is the rank of class j. Once each well is ranked with respect to both its concentration and trend, the average rank (6<sup>th</sup> column, Tab. 2) for identifying the training set to be used in the analysis can be computed as a simple average of the concentration rank value (3<sup>rd</sup> column, Tab. 2) and the trend rank value (5<sup>th</sup> column, Tab. 2).

Table 2 – Classification of the wells in the monitoring network based on both their nitrate contamination trend from 2001 to 2011 and last measured nitrate concentration (in 2011, or the closest year to 2011).

Well ID	Concentration class	Concentration rank	Trend class	Trend rank	Average rank
001	6	0.0909	1	0.3333	0.2121
002	7	0.0758	5	0.0667	0.0712
003	11	0.0152	4	0.1333	0.0742
221	1	0.1667	2	0.2667	0.2167

The frequency histogram of the average rank shows a slight right-skewed (non-normal) distribution (Fig. 4), thus the median value is identified as an optimal measure of its central tendency and it is selected as the threshold (Masetti et al., 2009). With the median value of 0.1167 representing a "reference critical value" for groundwater quality, wells with an average rank higher than 0.1167 constitute the training set, and those below 0.1167 form the control set.

The same procedure, used for the spatio-temporal model, is used to identify the training set for the predictive spatio-temporal model, with the exception that the nitrate concentration at time  $t_2$  is calculated using a hypothetical trend between  $t_1$  and  $t_2$ . Then, in order to consistently compare the spatio-temporal and the predictive spatio-temporal models, the same threshold value is selected and the same number of training points is used. These choices imply that the prior probability of the two models is the same and so is the "reference critical value" of groundwater quality.

#### 3.3 Evidential themes

The six explanatory variables selected for the analyses (§ 2.3) need to be consistent with the bases used to develop the spatio-temporal and predictive spatio-temporal models.

Natural factors are stable and considered as time invariable. Anthropogenic factors, which represent urban and agricultural nitrate sources, are handled by applying a formula that considers both the status of the variable at  $t_1$  (or  $t_2$ ) and its evolution from  $t_0$  to  $t_1$  (or  $t_1$  to  $t_2$ ), as expressed below:

Nitrate source "SpatialTemporal" value = 
$$\frac{|Value\ at\ t_1|}{Mean\ of\ the\ value} + \frac{Variation\ from\ t_0\ to\ t_1}{Mean\ of\ the\ variation}$$
(2)

where the means are calculated over the entire study area and are used to normalize the two terms of the equations. The formula is applied separately to urban and agricultural evidential themes (QuikSCAT-DSM datasets and nitrogen fertilizer loadings, Tab. 1).

### 3.4 Post-probability maps and map of severity

Once all the models are evaluated through the WofE, the post-probability maps can be obtained. The spatial, temporal and spatio-temporal models are categorized in five classes, which represent five degrees of groundwater vulnerability increasing from 1 to 5 based on criteria used to identify vulnerability classes (Sorichetta et al., 2011) and on visual analytic techniques (Cowan, 2001).

The post-probability maps obtained from the spatio-temporal and the predictive spatio-temporal models are compared by calculating the difference in percentage of the post probabilities in each pixel, and the obtained map is categorized in five classes of percentage (columns, Tab. 3). The map shows areas where the post-probability increases or decreases between the conditions at time  $t_1$  and  $t_2$ .

Then, the comparison map is combined with the spatial model as shown in the matrix in Table 3. The five new classes represent five degrees of severity from 1 to 5, with the higher values representing more severe conditions. The purpose of the severity map is to identify critical areas at  $t_2$ , based on the vulnerable areas at  $t_1$ . For example, two areas with the same increment of post probability shown by the comparison map (e.g., higher than 50 %) have two different implications if the class according to the spatial map at  $t_1$  is the least vulnerable (1) or the most vulnerable (5). The former area is less severe ( $3^{rd}$  class) than the second one ( $5^{th}$  class).

Table 3 – Matrix of severity. Severity increases from 1 to 5.

		Difference of Post Probabilities (%)				
		<-25%	-25÷0%	0÷25%	25÷50%	>50%
		1	2	3	4	5
SS	1	1	1	1	2	3
.y cla	2	1	1	2	3	4
abilit	3	1	2	3	4	5
Vulnerability class	4	2	3	4	5	5
<b>→</b>	5	3	4	5	5	5

### 4 Demonstration of the method

## 4.1 Spatial, Temporal and Spatio-Temporal models

The spatio-temporal approach is illustrated for the Po Plain area in the Lombardy Region (§ 2.1). Spatial, temporal and spatio-temporal models refer to the status of nitrate contamination and nitrate sources in 2011 and their evolution from 2000 to 2011. Table 4 shows the combinations of the significant evidential themes used to generate the response themes. For each model, only the statistically or physically significant evidential themes are considered in the analyses.

Table 4 - Combination of evidential themes used to obtain response themes and model area-under-the curve (AUC) values. spc = soil protective capacity; dsm [2009] = land use from satellite data in 2009;  $dsm/slope [2000 \div 2009] = land$  use changes from satellite data between 2000 and 2009; gwd [2003] = groundwater depth measured in 2003; gwv = groundwater velocity; hcv = hydraulic conductivity of the vadose zone. \* Data from Stevenazzi et al., 2015.

Model	Combination of evidential themes	Success Rate: AUC value	Prediction Rate: AUC value
Spatial	spc, dsm [2009], gwd [2003], gwv, hcv	78.4%	43.2%
Temporal	dsm/slope [2000 ÷ 2009], gwd [2003], gwv, hcv	74.3%*	62.1%
Spatio-temporal	dsm [2009] + dsm/slope [2000 ÷ 2009], gwd [2003], gwv, hcv	75.5%	51.9%

Spatial, temporal and spatio-temporal response themes are shown in Figure 5. Each response theme is categorized so that each vulnerability class in the corresponding map contains approximately the same number of different posterior probability values, according to the geometric interval method (Sorichetta et al., 2011).

The general quality of each response theme (i.e., post probability map) can be evaluated by the success and predictive rate curves (SRC and PRC, Chung and Fabbri, 1999; Sorichetta et al., 2013), expressed as the area-under-the curve (AUC) values. AUC is a direct measure of the performance of the statistical approach, and is given by the AUC integral for cumulated vulnerable area/cumulated wells as a percentage, using the training set for the SRC and the control set for the PRC. The calculated AUC values are presented in Table 4 showing the quality of the different maps. The high AUC values, obtained for the spatio-temporal model, and their similarity with the values obtained for the spatial and temporal models affirm the reliability of this approach in groundwater vulnerability assessment.

Then, the reliability of each classified map is evaluated based on its overall performance in classifying the occurrences. The average of the response variable of all wells in each vulnerability class is evaluated, using either the nitrate concentration, the nitrate concentration trend or the average ranking for the spatial (e.g., Sorichetta et al., 2011), temporal (Stevenazzi et al., 2015) and spatio-temporal models, respectively. These analyses are carried out using data from all available monitoring wells.

The evaluation of the average ranking of all wells,  $R_{AVG}$ , is expressed as:

$$R_{\text{AVG}} = \frac{\sum_{i=1}^{T_{Wj}} R_{ij}}{T_{Wi}} \tag{3}$$

where  $R_{ij}$  is the average ranking (6<sup>th</sup> column, Tab. 2) of well i in the vulnerability class j, and  $T_{Wj}$  is the total number of wells in the same class j. The average should monotonically increase as the degree of vulnerability increases and the central vulnerability class should give a value close to the overall mean value. In the three histograms, each regression line shows the overall expected direct correlation between degrees of vulnerability and (a) average nitrate concentrations, (b) average trends, and (c) average ranks (Fig. 6). The spatio-temporal model shows a good performance, with a regression coefficient close to 1, and presents the closest value to the mean of the whole distribution in the central vulnerability class, with respect to the other two models.

The groundwater vulnerability maps obtained through the spatial and temporal models show different classes of vulnerability when the combination of natural and anthropogenic factors involves either the presence of nitrate contamination or increasing nitrate concentrations. Since the two maps have different meanings, they cannot be meaningfully compared. Instead, the groundwater vulnerability map obtained through the spatio-temporal model shows the areas vulnerable due to the presence of nitrate contamination and/or an increasing trend of nitrate concentrations. The distribution of the vulnerable areas reflects the distribution of those urban areas that are characterized by the presence of a notable and continuous urban pattern and/or show an increase in extension or modifications of their pattern (e.g., revitalization of industrial areas and transformation in residential or green areas).

#### 4.2 Predictive Spatio-Temporal model

The predictive spatio-temporal model considers a potential/hypothetical scenario of nitrate contamination of groundwater in 2020, and its variation during the period from 2011 to 2020, in the Po Plain area of the Lombardy Region (§ 2.1). Two scenarios are investigated: the first (Scenario 1) considers only a possible evolution of urban sources of nitrate contamination; the second (Scenario 2) considers also a possible evolution of a natural factor, which is taken as the groundwater depth.

In Scenario 1, the evolution of nitrate contamination and urban nitrate sources (i.e., urban areas) during the period of 2011 to 2020 is taken as equal to their evolution in the decade of 2000 to 2010. Natural factors are considered stable and remain unchanged in the two decades.

The response variable is obtained using the approach described in § 3.2. Wells are classified considering the trend in the period from 2011 to 2020 and the concentration value is calculated for 2020, which depends on the trend. In Scenario 1, the equal trends in 2011 to 2020 and in 2000 to 2011 means that the slope of the regression line obtained from the interpolation of concentration data from 2000 to 2011 is used as trend for 2011 to 2020 in order to calculate the nitrate concentration in 2020.

The evidential theme representing the urban nitrate sources (i.e., QuikSCAT-DSM) is obtained using the method described in § 3.3 and by applying the formula in Equation 2, as:

Urban nitrate source "SpatioTemporal" value

$$= \frac{|\text{Value QuikSCAT. DSM}_{2020}|}{\text{Mean of the value QuikSCAT. DSM}_{2020}}$$

$$+ \frac{\text{Slope QuikSCAT. DSM}_{2011 \div 2020}}{\text{Mean of the slope QuikSCAT. DSM}_{2011 \div 2020}}$$

$$(4)$$

where the value of QuikSCAT-DSM in 2020 is calculated using the slope of QuikSCAT-DSM for 2011 to 2020, which is equal to the slope obtained for 2000 to 2009 (Stevenazzi et al., 2015). Changes of nitrate concentrations in groundwater and rates of urbanization for the 2020-scenario are assumed to be consistent with the trend observed in the decade since 2000 (§ 4.1).

Scenario 2 is an implementation of Scenario 1 where the possible evolution of a natural factor is considered as an additional changing factor alongside the evolution of nitrate contamination and urban nitrate sources. The evolution of groundwater depth is evaluated starting with the piezometric levels measured in 2003 and 2014. Groundwater depth for the year 2020 is calculated with the formula below:

$$gwd_{2020} = gwd_{2003} + 1.5 \times (gwd_{2014} - gwd_{2003})$$
(5)

where gwd is groundwater depth. The formula assumes that groundwater depth varies according to a linear regression equation, which considers that the slope of the regression line between 2014 and 2020 is equal to half of the slope between 2003 and 2014, as to model realistic changes in groundwater depth.

While more complicated scenarios can be assumed, in this analysis it is assumed that the variation of nitrate contamination in groundwater is the same as in Scenario 1, although groundwater depth changes could cause different nitrate concentration trends with respect to the assumed variability.

The evidential themes used to generate the models are listed in Table 5. Soil protective capacity and nitrogen fertilizer load are not statistically significant, and they are not considered in the analysis. The reliability of both predictive models, measured via the AUC values, (Table 5), is similar to the reliability of the spatio-temporal model (Table 4).

Table 5 - Combination of evidential themes used to obtain the response themes of the predictive spatio-temporal models and AUC values: dsm [2020] = land use from satellite data in 2020;  $dsm/slope [2010 \div 2020] = land$  use changes from satellite data between 2010 and 2020; gwd [2003] or gwd [2020] = groundwater depth measured in 2003 or calculated for 2020; gwv = groundwater velocity; hcv = hydraulic conductivity of the vadose zone.

Predictive model	Combination of evidential themes	Success Rate: AUC value	Prediction Rate: AUC value
Scenario 1	dsm [2020] + dsm/slope [2010 ÷ 2020], gwd [2003], gwv, hcv	74.5%	53.1%
Scenario 2	dsm [2020] + dsm/slope [2010 ÷ 2020], gwd [2020], gwv, hcv	75.3%	53.1%

The groundwater vulnerability maps, in the form of post-probability maps, obtained through the predictive spatio-temporal model are calibrated and partially validated. The AUC values of the SRCs are high and are consistent with those obtained via the other models (spatial, temporal and spatio-temporal models). Alternatively, the AUC values of the PRCs are comparable with that obtained for the spatio-temporal model, confirming their capability to adequately describe groundwater vulnerability in the study area. As the predictive spatio-temporal models are developed based on a potential or hypothetical evolution of nitrate contamination and nitrate sources, the validation technique used in the other three cases is not applicable, as the future change is merely a projection of future groundwater contamination (§ 4.1).

Finally, as described in § 3.4, in order to recognize the severely contaminated areas in 2020, given the vulnerable areas in 2011, maps of severity are generated (Fig. 7).

Maps of severity show that groundwater contamination remains a major problem in the northern sector of the study region, with the prevalence of high classes of severity. The southern sector is generally less vulnerable, while medium-high levels of severity are present in some areas in the central and east-southern sectors. This highlights potential problems in groundwater quality if land-use policies are maintained as assumed in the two scenarios.

#### 5 Discussion and conclusion

The provision of safe groundwater is a crucial global challenge to sustain healthy living conditions in the ever-growing urban areas. Consequently, the associated increase of potential sources of contamination together with increasing urbanization causes a close interrelation between the presence of groundwater-contaminated areas, and the difficulties of using groundwater without considering investments needed to maintain the water quality.

Groundwater vulnerability must be evaluated considering the role of potential sources of contamination on groundwater quality. These data must be integrated with parameters related to climatic and hydrogeological factors in order to select a combination of factors most effective in characterizing groundwater quality.

The development of a dynamic and time-dependent groundwater vulnerability approach is a crucial tool to support sustainable land use practices that preserve groundwater quality within a regional hydrogeological context. By determining which natural and anthropogenic factors are the most responsible for groundwater deterioration, adequate environmental policies can be developed for the most effective implementation. The proposed approach of a spatio-temporal vulnerability model is demonstrated to be valid at identifying areas where natural and anthropogenic factors are responsible for combined effects of current nitrate concentration and increasing nitrate concentration trends. The approach utilizes the advantages that statistical methods have over other methods used for assessing groundwater vulnerability, and further expands their capability by introducing the time dimension. The new advances enable an evaluation of the efficiency of new land use and urban development decisions

on aquifer protection to achieve mid- to long-term safeguarding of groundwater quality (Lavoie et al., 2015). The proposed approach allows for determination of what can happen to groundwater resources if policies are maintained at the status quo or if new measures are introduced as natural factors change under climatic or anthropogenic stresses. In fact, the final calibrated spatio-temporal map can be projected into the future to predict potential effects within realistic scenarios that rely on observed land use and contamination trends.

The introduction of the time dimension makes this method the first that can include the recent history of contamination in groundwater vulnerability assessment to meet the requirements from the European Union (Groundwater Directive, 2006/118/EC) on both the contamination levels and their trends for groundwater protection and management. Moreover, the approach shows how time series of land use changes derived from satellite data can be efficiently integrated in groundwater vulnerability assessments. The methodology can be then used to investigate more realistic future scenarios, based on more complex models of the spatial evolution of both time-dependent natural and anthropogenic factors.

In conclusion, the inclusion of the time dimension in groundwater vulnerability studies can provide breakthrough advances in identifying and mapping vulnerable areas on a regional or sub-regional scale, and in assessing the efficacy of land-use planning and policies for groundwater protection:

- a) The proposed spatio-temporal groundwater vulnerability maps derived through the spatial statistical approach are a reliable tool to combine natural and anthropogenic factors in order to identify areas with a critical combination of increasing nitrate concentration trends and current nitrate concentration.
- b) The introduction of a map of severity offers a new perspective in the projection of how different future scenarios would impact the quality of groundwater resources in a given area, and to help develop pro-active, and thus effective, land use policies to preserve groundwater quality.
- c) Through this approach, decision makers can have a high-level guide on the compatibility of a given policy or activity in a given area to alleviate new groundwater diffuse contamination situations or avoid the deterioration of the existing ones.

d) Applicable to global satellite data together with ancillary ground data, the method can be used to obtain extensive results for an improved protection of groundwater quality, which has clear advantages for the environment and for groundwater dependent ecosystems and a benefit to society that transcends artificial political boundaries across the world.

### 6 Acknowledgments

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# 8 Figures

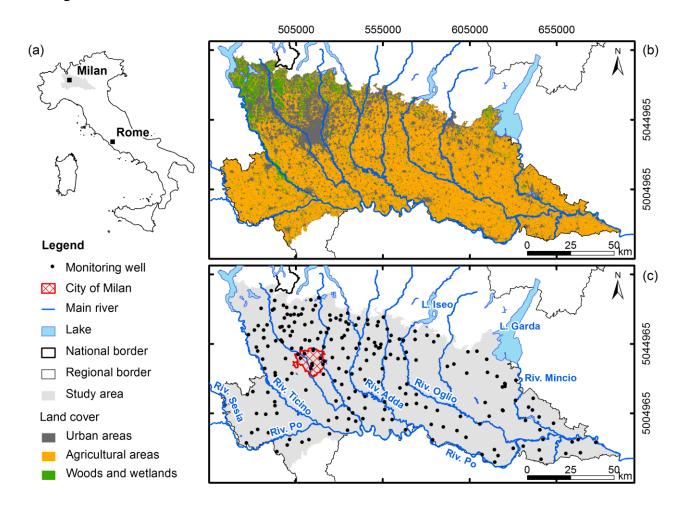


Fig. 1 – (a) Location of the study area in light grey. (b) Land cover in 2012. (c) Monitoring-well network.

Coordinates refer to WGS 1984 – UTM Zone 32 N projection.

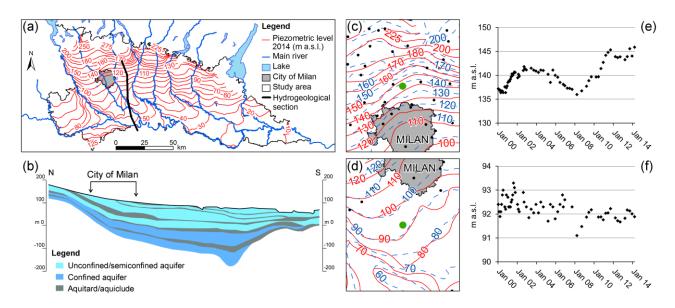


Fig. 2 – (a) Piezometric levels in 2014 (m a.s.l.) and location of the hydrogeological section; (b) N-S hydrogeological section, modified from Regione Lombardia and ENI (2002); (c, d) Piezometric levels (m a.s.l.) in 2003 (dashed blue lines) and in 2014 (solid red lines) and monitoring well network (black dots) in areas around the city of Milan; (e, f) Piezometric levels measured in two monitoring wells located in the northern (e) and in the southern (f) sector of the area, located as the green dots in maps (c) and (d).

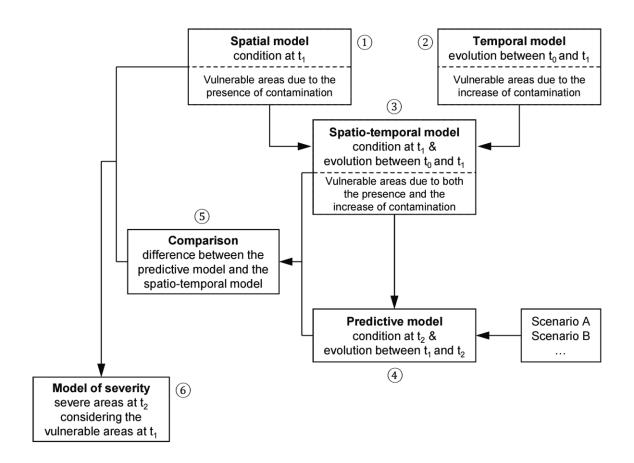
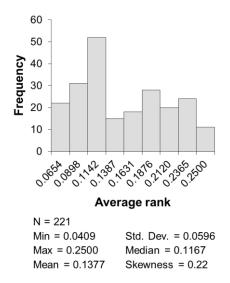


Fig. 3 – Study flow for groundwater vulnerability mapping.



*Fig. 4 – Frequency histogram of the average rank.* 

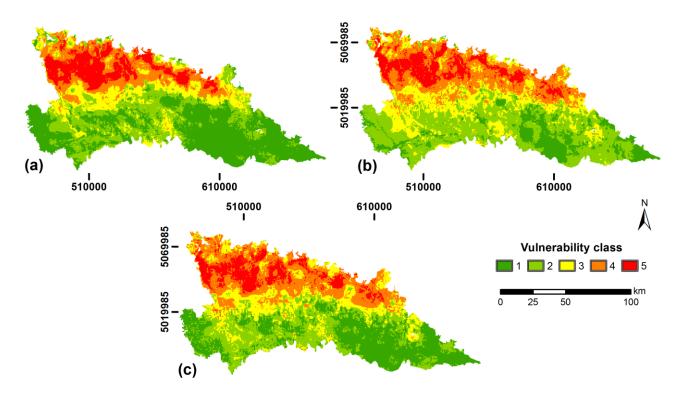


Fig. 5 – Vulnerability maps representing: (a) spatial model, (b) temporal model (from Stevenazzi et al. 2015), (c) spatio-temporal model. Coordinates refer to WGS 1984 – UTM Zone 32 N projection.

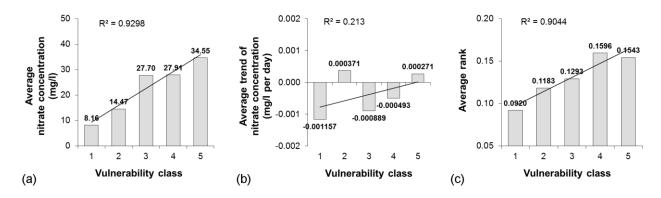


Fig. 6 – Histograms of (a) the average nitrate concentration, (b) the average nitrate concentration trend\*, and (c) the average ranking in each vulnerability class of the spatial, temporal and spatio-temporal model, respectively. The degree of vulnerability increases from 1 to 5. \*Data from Stevenazzi et al., 2015.

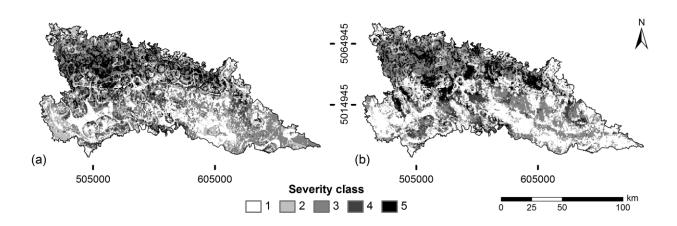


Fig. 7 – Maps of severity in 2020 according to Scenario 1 (a) and Scenario 2 (b). Coordinates refer to WGS 1984 – UTM Zone 32 N projection.

## **Supplementary Material**

Nitrate concentrations and nitrate concentration trends have been categorized in eleven and five classes according to the classification in Table S1 and S2, respectively. The high reliability of nitrate concentration measurements allows to distinguish a high number of classes, according to a 5 mg/L-stepping. Instead, nitrate concentration trends follow a wider discretization to recognize wells showing a clear increasing trend, wells showing a clear decreasing trend and wells characterized by an uncertainty in the slope coefficient value.

Table S1 - Classification of the wells based on the last measured nitrate concentration, in 2011, or the closest year to 2011 (Sorichetta, 2011).

Concentration class	Concentration in mg/L	
1	C > 50	
2	$45 < C \le 50$	
3	$40 < C \le 45$	
4	$35 < C \le 40$	
5	$30 < C \le 35$	
6	25 < C ≤ 30	
7	20 < C ≤ 25	
8	15 < C ≤ 20	
9	10 < C ≤ 15	
10	5 < C ≤ 10	
11	C ≤ 5	

*Table S2 - Classification of the wells based on the nitrate concentration trend of the period 2001–2011.* 

Trend class	Trend in mg/L per day
1	T > + 0.0015
2	$+0.0003 < T \le +0.0015$
3	$-0.0003 < T \le +0.0003$
4	$-0.0015 < T \le -0.0003$
5	$T \le -0.0015$

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