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2 **Groundwater vulnerability maps derived from a time-dependent method using satellite**  
3 **scatterometer data**

4 Stevenazzi, Stefania (contact)<sup>1</sup>; Masetti, Marco<sup>1</sup>; Nghiem, Son V.<sup>2</sup>; Sorichetta, Alessandro<sup>3</sup>

5 <sup>1</sup>: Dipartimento di Scienze della Terra “A. Desio”, Università degli Studi di Milano, Via Luigi  
6 Mangiagalli, 34, Milano 20133, Italy, stefania.stevenazzi@unimi.it, marco.masetti@unimi.it

7 <sup>2</sup>: Jet Propulsion Laboratory, California Institute of Technology, 4800 Oak Grove Drive, Pasadena, CA  
8 91109, USA, son.v.nghiem@jpl.nasa.gov

9 <sup>3</sup>: Geography and Environment, University of Southampton, Highfield Campus, Shackleton Building  
10 44, Southampton SO17 1BJ, UK, A.Sorichetta@soton.ac.uk

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14 **Abstract**

15 Introducing the time variable in groundwater vulnerability assessment is an innovative approach to study  
16 the evolution of contamination by non-point sources and to forecast future trends. This requires a  
17 determination of the relationship between temporal changes in groundwater contamination and in land  
18 use. Such effort will enable breakthrough advances in mapping hazardous areas, and in assessing the  
19 efficacy of land-use planning for groundwater protection. Through a Bayesian spatial statistical  
20 approach, time-dependent vulnerability maps are derived by using hydrogeological variables together  
21 with three different time-dependent datasets: population density, high-resolution urban survey, and  
22 satellite QuikSCAT (QSCAT) data processed with the innovative Dense Sampling Method (DSM). This  
23 approach is demonstrated extensively over the Po Plain in Lombardy region (northern Italy). Calibrated  
24 and validated maps show physically consistent relations between the hydrogeological variables and  
25 nitrate trends. The results indicate that changes of urban nitrate sources are strongly related to

26 groundwater deterioration. Among the different datasets, QSCAT-DSM is proven to be the most  
27 efficient dataset to represent urban nitrate sources of contamination, with major advantages: a worldwide  
28 coverage, a continuous decadal data collection, and an adequate resolution without spatial gaps. This  
29 study presents a successful approach that, for the first time, allows the inclusion of the time dimension  
30 in groundwater vulnerability assessment by using innovative satellite remote sensing data for  
31 quantitative statistical analyses of groundwater quality changes.

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## 34 **1. Introduction**

35 Groundwater is among the most important freshwater resources. In Western Europe, it contributes 60 %  
36 of the drinking water supply (EuroGeoSurveys 2014). Increasing numbers of contamination sources in  
37 developed and developing countries critically threaten groundwater resources. Reactive remediation  
38 measures can be excessively expensive when groundwater becomes contaminated beyond the required  
39 quality standards for safe consumption.

40 Groundwater vulnerability studies are crucial to understand the cause-effect relationship between  
41 groundwater quality and both natural and anthropogenic factors to develop effective groundwater  
42 protection plans. Mapping areas where groundwater is most vulnerable to contamination and identifying  
43 primary factors influencing the contamination level are imperative to manage and protect groundwater  
44 and thus human health.

45 As groundwater resources have become more vulnerable in recent years, it is necessary to urgently close  
46 the gap between the information required for land use planning to efficiently safeguard groundwater  
47 quality and techniques required to accurately assess groundwater vulnerability. In fact, the European  
48 Union (EU) Groundwater Directive (2006/118/EC) requires the identification of areas where  
49 groundwater suffers increasing trends in contaminant concentration, highlighting the need to carefully  
50 manage such areas even if the concentration is below the regulatory limit.

51 A current limitation in groundwater vulnerability studies is related to the lack of consideration of  
52 temporal trends (Stuart et al. 2007), and this emphasizes the need to consider the time dimension in

53 assessing groundwater vulnerability. Methods currently used to assess groundwater vulnerability at a  
54 regional scale (Focazio et al. 2002) can be subjective (i.e., knowledge-driven) or objective (i.e., data  
55 driven). Subjective methods include overlay and index methods (e.g., DRASTIC, Aller et al. 1987;  
56 GOD, Foster 1987; AVI, Van Stempvoort et al. 1993; and EPIK, Doerfliger and Zwahlen 1997) and  
57 their modifications (Sener and Davraz 2013). They are easy to implement and require a limited amount  
58 of data to derive a subjective categorization of groundwater vulnerability. On the other hand, objective  
59 methods are based on the use of statistical methods, ranging from descriptive statistics (e.g., Welch et  
60 al. 2000) to regression and conditional probability analyses (e.g., Eckardt and Stackelberg 1995;  
61 Tesoriero and Voss 1997; Nolan 2001; Alberti et al. 2001; Worrall and Besien 2005; Masetti et al. 2009),  
62 which allow an objective determination of relations between the predictor factors and the level of  
63 contamination in the study area. In this regard, only objective methods allow scientifically defensible  
64 end products (Focazio et al. 2002) and, most importantly, enable an explicit integration of the time  
65 dimension in the groundwater vulnerability assessment (Sorichetta 2011).

66 Objective methods, however, face a major challenge that requires an extensive dataset, including a series  
67 of contaminant concentration measurements and natural and anthropogenic variables, to be consistent  
68 both in space and in time (Brunner et al. 2007). Addressing such a challenge demands a determination  
69 of the relationship between temporal changes in groundwater contamination and in land use across a  
70 vast spatial extent encompassing natural environments, agricultural regions, and urban areas. This effort  
71 will enable breakthrough advances to improve the mapping of hazardous areas with different levels of  
72 vulnerability, and to assess the efficacy of land use planning toward groundwater protection.

73 In this context, this study focuses on advancing the use of statistical methods to assess groundwater  
74 vulnerability by explicitly introducing the time dimension in the analysis. The objectives are to address  
75 recent requirements from transnational policies and to close the critical information gap described  
76 earlier.

77 In view of current and projected acceleration in global urbanization, urban areas are widely considered  
78 as one of the most important non-point sources of contamination impacting groundwater quality (Kuroda  
79 and Fukushi 2008). The Environmental European Agency in 2006 reported that the expansion of urban  
80 areas in many eastern and western European countries has increased by over three times the growth of

81 population between 1986 and 2006 (EEA 2006). Urban sprawl is one of the most important types of  
82 land-use changes impacting the regional environment, the social structure, and the economy in Europe.  
83 Urban sprawl generally follows periods of rapid urbanization associated with population growth and  
84 with the excessive migration of people from rural to urban areas. The Po Plain in northern Italy is one  
85 of the most populated regions in Europe with a similar pattern: an initial phase of urban area expansion  
86 from the 1950s to the 1970s followed by an urban sprawl in the subsequent decades. This pattern  
87 qualifies the Po Plain as a representative “pilot area” to identify the interplay of urbanization and  
88 environmental, social and economic impacts after the rapid urban increase.

89 Nitrate is an abundant contaminant of groundwater. With a high mobility and multiple sources, nitrate  
90 is an effective indicator of groundwater contamination. A sufficient frequency for monitoring nitrate  
91 concentration in groundwater over the long term allows the use of nitrate in temporal analyses to  
92 determine the contamination trend.

93 Recent studies (Masetti et al. 2008, 2009; Sorichetta et al. 2011) have shown that, in some areas of the  
94 Po Plain, nitrate occurrence in groundwater is strongly related to urban sources (using population density  
95 as a proxy) more than to agricultural activities; however, the problem has never been analyzed in the  
96 time dimension. It is unclear whether a relationship exists between recent changes in groundwater nitrate  
97 contamination and in land use. To analyze how urban development could affect groundwater quality in  
98 the 2000s, recent trends in groundwater nitrate concentration need to be correlated with the evolution of  
99 potential urban nitrate sources across this region.

100 While satellite data have been typically used to qualitatively assess the availability of groundwater  
101 resources (Jha and Chowdary 2007; Tweed et al. 2007; Al Saud 2010; Jha et al. 2010; Jasmin and  
102 Mallikarjuna 2011; Frappart et al. 2011; Wang et al. 2014), only a limited number of studies used  
103 satellite data to quantitatively assess groundwater quality (Werz and Hötzl 2007). The use of an  
104 innovative dataset to delineate urban areas with satellite scatterometer data has been explored to identify  
105 zones where different rates of urban growth occurred across the entire study area, and in which an  
106 increase of potential urban sources may exist and consequently impact groundwater. Radar backscatter  
107 data acquired by the SeaWinds scatterometer aboard the QuikSCAT satellite together with the Dense  
108 Sampling Method (QSCAT-DSM; Nghiem et al. 2009) have been used to identify and map urban extent

109 and surface features at a posting scale of about 1 km<sup>2</sup>. QSCAT-DSM results are to be compared with  
110 those obtained from two different sources of urban information: 1) changes of population density and  
111 2) changes in land use derived from high-resolution aerial images acquired in different years.  
112 In addition, to address the time dimension in groundwater vulnerability assessment, this study can be  
113 considered the first to use remote sensing data to obtain a quantitative assessment of groundwater quality  
114 changes through time. Moreover, it also represents one of the first applications of QuikSCAT data to  
115 environmental and hydrogeological problems with an optimal spatial scale enabled by DSM.  
116 In this study, a “zone vulnerable to nitrate contamination” can be defined as an area where the  
117 combination of natural (e.g., groundwater depth and velocity) and anthropogenic factors (e.g., growth  
118 of urban areas) involves a deterioration trend of groundwater quality. In a static system, a “zone  
119 vulnerable to nitrate contamination” can be defined as an area where the combination of the same factors  
120 involves a given absolute level of contamination in the aquifer.

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## 126 **2. Study area**

127 The study area is located within the Po Plain area of Lombardy region, and covers an area of 13,400  
128 km<sup>2</sup>, where urban, industrial, livestock and agricultural activities are extensively and heterogeneously  
129 present.

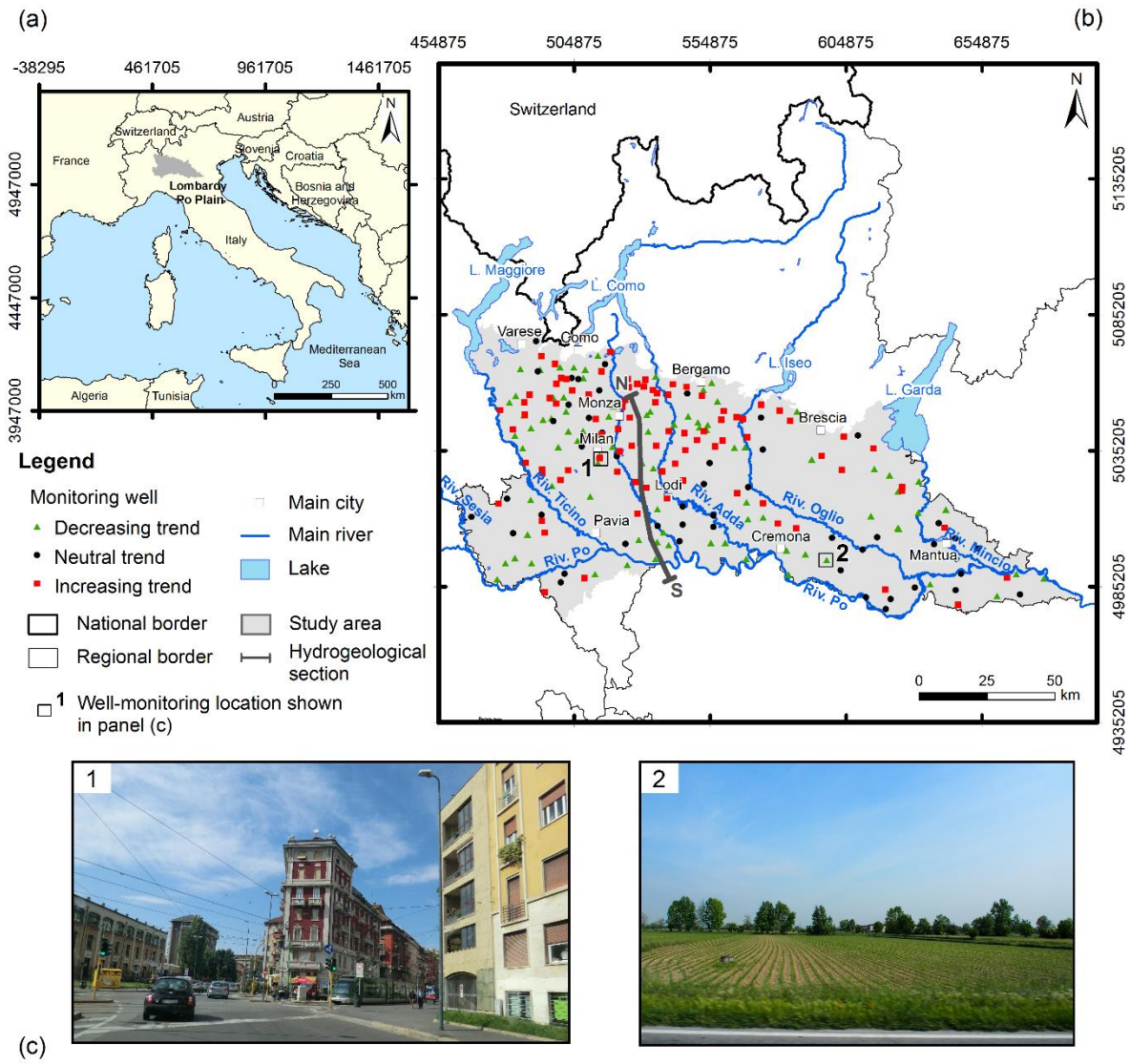
130 This region is surrounded by important rivers influencing groundwater flow in the unconfined aquifer:  
131 Po River along the south; Ticino, Sesia and Po rivers along the west; and Mincio River along the east  
132 (Fig. 1). It is also constrained by mountain chains forming the boundary of the plain: Lombardy Prealps  
133 along the north and Appennines along the southwest.

134 This area has a complex hydrogeological setting consisting of multiple aquifers with various properties  
135 and interactions. The Lombardy plain subsoil is characterized by Plio-Pleistocene sediments whose  
136 upper unit forms the shallow unconfined aquifers (Fig. 2). Sediments are mainly gravels and sands

137 although the presence of finer sediments increases from the north to the south where shallow aquifers  
138 are mainly constituted by fine sands and are partially confined. These aquifers have high transmissivity,  
139 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,  
140 while its thickness ranges from 40 to 80 m (Regione Lombardia and ENI 2001).

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

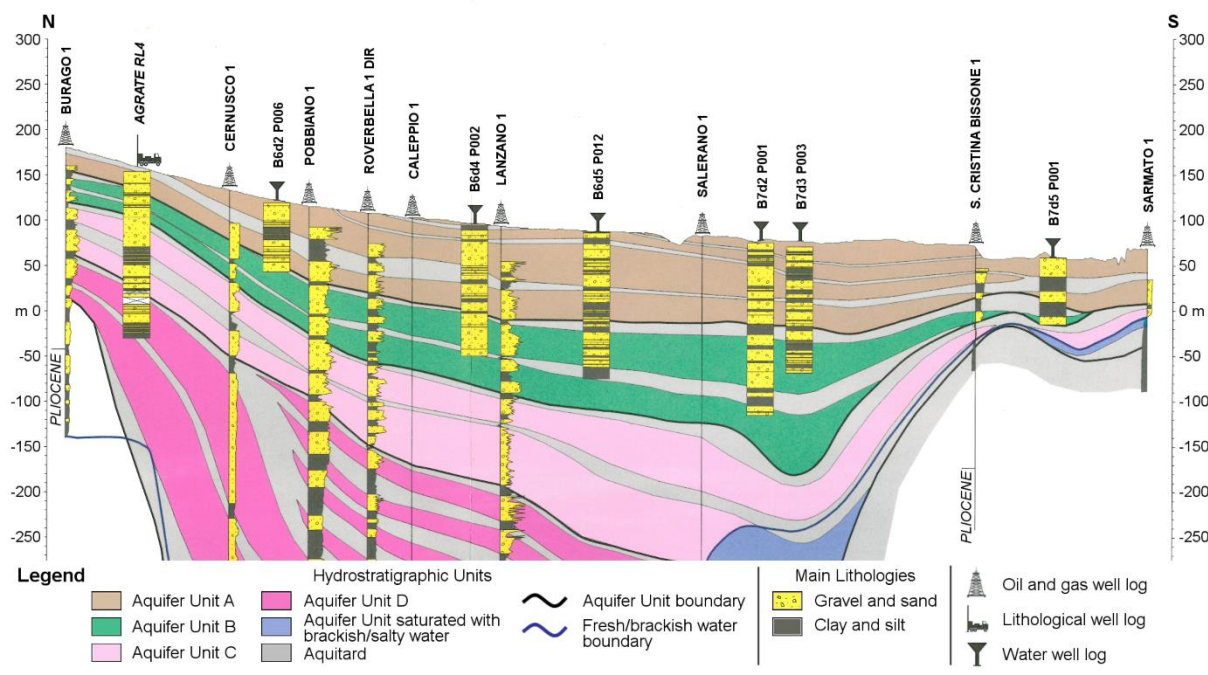
145 Nitrate ( $\text{NO}_3^-$ ) is the most common non-point-source contaminant found in groundwater in the Po Plain.  
146 Nitrate concentrations have been monitored by a network of about 500 wells covering the entire area  
147 with a nearly uniform spatial distribution, where data have been collected every six months from 2001  
148 to 2011 (Regional Environmental Agency – ARPA, unpublished data, 2012). From the network, only  
149 the 221 wells monitoring the shallow aquifer and having a minimum of eight measurements were  
150 selected for being used in the analysis. As an example (Fig. 1), two photographs taken during the PO  
151 PLain EXperiment (POPLEX) in May 2014 (Masetti et al. 2014; Nghiem et al. 2014a) show rural and  
152 urban areas, as two major contrastive types of land use where the monitoring wells are located.



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155 This study focuses on the evolution of nitrate concentration in groundwater related to changes of urban  
 156 areas. The change in nitrate concentration is quantified by the slope of the regression line from an  
 157 interpolation of concentration data. The slope defines the rate of nitrate concentration change in mg/L  
 158 per day. Positive slope values show increasing concentration trends representing water quality  
 159 deterioration, while non-positive slope values indicate steady or decreasing concentration trend  
 160 characterizing unaffected or improved groundwater quality.



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Until the end of the considered monitoring period in 2011, about 28 % of wells show increasing concentration trends and concentrations exceeding the guideline value of 25 mg/L defined by the EU standard (91/676/EEC), while 35 % of wells show decreasing concentration trends and concentrations lower than the same guideline value. Only 3 % of wells show increasing concentration trends and concentrations exceeding the established threshold of 50 mg/L (91/676/EEC; 2006/118/EC) (Table 1). Wells with concentrations higher than the guideline value of 25 mg/L are located mainly in the northern sector, while those with concentrations lower than the guideline value are mostly located in the southern sector.

172 **Table 1** Nitrate concentration trends related to the last measured concentration (percentage of wells)

	Increasing concentration trend	Decreasing concentration trend
Concentration $\geq$ 25 mg/L in 2011	28 %	18 %
Concentration < 25 mg/L in 2011	19 %	35 %
Concentration $\geq$ 50 mg/L in 2011	3 %	6 %
Concentration < 50 mg/L in 2011	44 %	47 %



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### 175 **3. Method and Materials**

#### 176 **3.1. Method**

177 The Weights of Evidence (WofE) modeling technique combines different spatial datasets in a  
178 Geographical Information System (GIS) environment to analyze and describe their interactions and  
179 generate predictive patterns (Bonham-Carter 1994; Raines et al. 2000). WofE can be defined as a data-  
180 driven Bayesian method in a log-linear form that uses known occurrences representing the response  
181 variable as training sites (training points). These data are used to obtain predictive probability maps  
182 (response themes; i.e., groundwater vulnerability maps) from multiple weighted evidences (i.e.,  
183 evidential themes representing explanatory variables or factors that influence groundwater  
184 vulnerability), which determine the spatial distribution of the occurrences in the study area (Raines  
185 1999).

186 Training points (TPs) are used in WofE to calculate the prior probability, the weights for each class  
187 representing a different range of values of each generalized evidential theme, and the posterior  
188 probability values in the response theme.

189 Prior probability is based on prior knowledge of the TPs' locations in the study area. Prior probability  
190 is simply defined by the ratio between the area containing occurrences (i.e., the number of pixels  
191 containing a training point  $D$ ) and the total area (i.e., the total number of pixels). Thus, the prior  
192 probability represents the probability that a pixel within the study area contains an occurrence without  
193 considering any evidential themes, and it can be expressed as (Bonham-Carter 1994):

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$$P\{D\} = \frac{N_D}{N_T} \quad (1)$$

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196 where  $N_D$  and  $N_T$  are respectively the number of pixels containing a training point and the total number  
197 of pixels in the study area.

198 For each class of each evidential theme, a positive and a negative weight are computed based on the  
 199 location of the TPs with respect to the study area. For a given class  $B$ , the positive weight  $W^+$  and the  
 200 negative weight  $W^-$  are, respectively, higher and lower than zero or lower and higher than zero. The  
 201 resulting combination depends on whether  $B$  has more or fewer TPs than expected by chance.

202 The weights can be expressed as (Bonham-Carter 1994):

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$$W^+ = \log_e \frac{P\{B|D\}}{P\{B|\bar{D}\}} \quad (2)$$

$$W^- = \log_e \frac{P\{\bar{B}|D\}}{P\{\bar{B}|\bar{D}\}} \quad (3)$$

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205 where  $P\{B|D\}$  and  $P\{B|\bar{D}\}$  are respectively the probability of a pixel of being in the class  $B$  when the  
 206 same pixel contains or does not contain a training point, and  $P\{\bar{B}|D\}$  and  $P\{\bar{B}|\bar{D}\}$  are respectively the  
 207 probability of a pixel of not being in the class  $B$  when it contains or does not contain a training point.

208 The contrast (positive weight minus negative weight) represents the overall degree of spatial association  
 209 between each class of a given evidential theme and TPs. Thus, it is a measure of the usefulness of the  
 210 considered class in predicting the location of TPs (Raines 1999).

211 A confidence value for the ratio between the contrast and its standard deviation must be selected to  
 212 provide a useful measure of the significance of the contrast (Raines 1999). For this study, a confidence  
 213 value of 1.282, corresponding approximately to a 90 % level of significance, was chosen as the minimum  
 214 acceptable value to consider an evidential theme class as statistically significant.

215 The posterior probability represents the relative probability that a pixel contains an occurrence based on  
 216 the evidences provided by the evidential themes (i.e., based on the calculated weights). The posterior  
 217 probability can be expressed as (Bonham-Carter 1994):

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$$\log_e O\{D|B_1^k \cap B_2^k \cap B_3^k \dots \cap B_n^k\} = \sum_{j=1}^n W_j^k + \log_e O\{D\} \quad (4)$$

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220 where  $n$  identifies each single class used to categorize each evidential theme,  $k$  is either + or - depending  
221 on whether the prediction spatial class,  $B_n$ , is either present or absent, and  $O\{D\}$  is the odd form of the  
222 probability that a pixel within the study area contains an occurrence..

223 The relative probability means that a pixel having a higher posterior probability is more likely to contain  
224 an occurrence than a pixel having a lower probability, and it represents a measure of the relative  
225 likelihood of occurrence of an event (Raines 1999).

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

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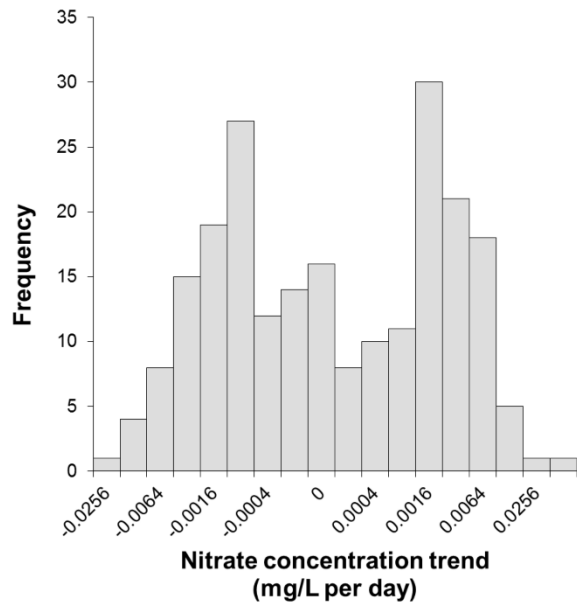
### 229 **3.2. Response variable**

230 For the purpose of this study, the response variable is represented by nitrate concentration trend in  
231 groundwater. The WofE modelling technique requires a binary formulation of the response variable.

232 A frequency histogram of nitrate concentration trend shows a nearly bimodal distribution with two main  
233 relative peaks at about -0.0008 and +0.00016 mg/L per day (Fig. 3). Another minor peak can also be  
234 identified at value 0. The intermediate values of -0.0004 and +0.0004 mg/L per day, which separate  
235 three populations, were considered to be appropriate values to be used as thresholds.

236 Wells showing concentration trends higher than +0.0004 mg/L per day are considered as “increasing”  
237 wells (87), and those below -0.0004 mg/L per day as “decreasing” wells (86). Wells showing  
238 concentration trends included in the range -0.0004 and +0.0004 mg/L per day are considered as “neutral”  
239 wells (48). In these wells, the uncertainty in the slope coefficient value, which is close to zero, does not  
240 allow one to precisely categorize them as “increasing” or “decreasing” wells.

241 The “increasing” wells, showing a clear increase in concentration trends, represent the training set, and  
242 they have been selected to be used in the analysis. While “decreasing” and “neutral” wells are grouped  
243 in a unique set, representing the control set.



N = 221  
 Min = -0.03095                      Std. Dev. = 0.004754  
 Max = 0.03040                      Median = -0.000035  
 Mean = -0.0002175                  Skewness = -0.60

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247 **3.3 Evidential themes**

248 Both natural and anthropogenic factors have been used as evidential themes in the analysis. Natural  
 249 factors include geological and hydrogeological conditions of the study area and are considered static for  
 250 the purpose of the study. Three different factors have been selected to represent changes of urban nitrate  
 251 sources through time, i.e. land use change derived from 1) satellite data, 2) aerial photographs, and 3)  
 252 data on population density changes. Details on each of the factors and on how they are used in the study  
 253 are presented below.

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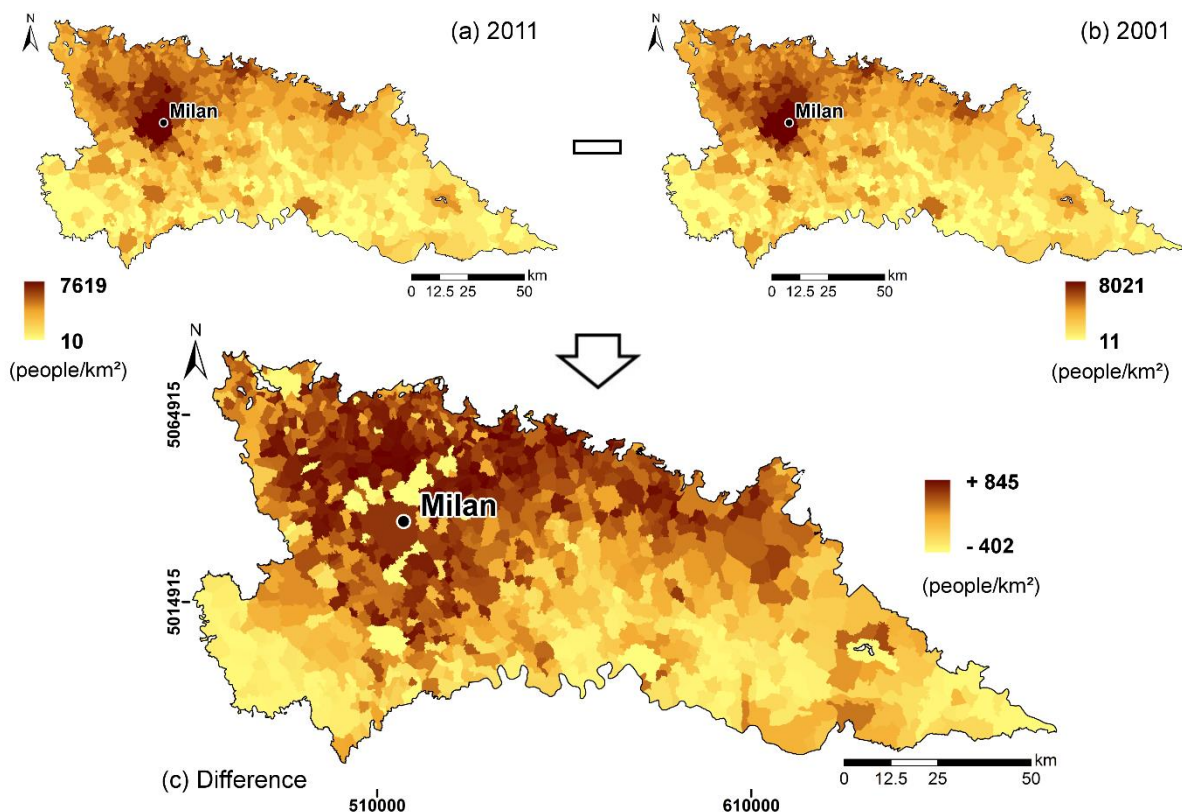
255 **3.3.1 Urban nitrate sources: anthropogenic factors**

256 Nitrogen loading derived from urban areas (presence of sewer leakage or septic tanks) cannot be easily  
 257 or directly estimated quantitatively. For this reason, it is necessary to explore other variables that can be  
 258 used as a proxy.

259 For this purpose, it is crucial to have a temporally and spatially consistent dataset delineating the urban  
 260 area extent through time in order to investigate the potential relationships between its variation and the  
 261 evolution of groundwater contamination. Even in the data-rich European and North American countries,  
 262 such information is not collected consistently or during consistent periods of time and is often spatially  
 263 and/or temporally limited.

264 Population density has been used often as a proxy for urban nitrate sources in groundwater vulnerability  
 265 assessments (Nolan 2001; Nolan et al. 2002; Masetti et al. 2009; Sorichetta et al. 2011). Population  
 266 density is generally referred to administrative units at the specific time of the demographic census or  
 267 survey. Official national censuses are usually done once every ten years. Consequently, analyses based  
 268 on population census cover a period of ten years, missing changes in shorter periods.

269 In this study, the population-density change is calculated as the difference between population densities  
 270 in each district referred to two successive national censuses, in 2001 and 2011 (ISTAT 2001, 2011).  
 271 Positive values indicate a growth of population, and negative values represent a reduction of population.  
 272 Between 2001 and 2011, population density changed in the range from -402 to +845 people/km<sup>2</sup> across  
 273 Lombardy (Fig. 4).

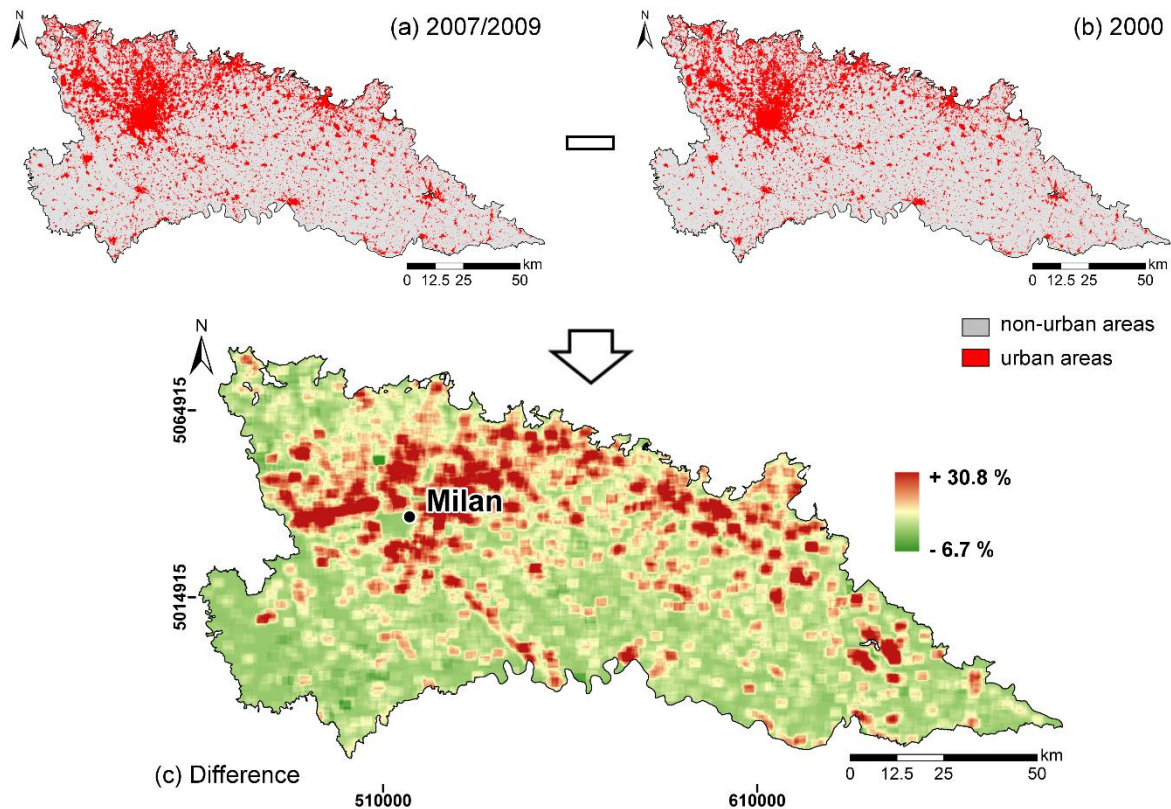


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High-resolution aerial images in Lombardy have been periodically acquired by the Agency of Services of Agriculture and Forest (ERSAF), creating a database called DUSAF (ERSAF 2014), to identify and categorize the land cover in five main land use classes: urban areas, agricultural areas, woods and semi-natural environments, wetlands and surface water areas. The technical maps are at a 1:10,000 scale. For the purpose of the study, vector maps have been transformed to raster format. DUSAF is updated at irregular intervals that can be different for different sectors of the region. This limitation does not allow the maps to represent the urban land use at the same time across the whole region.

To observe changes in urban extent, the two relevant groups are: urban areas, and non-urban areas consisting of the remaining four classes in DUSAF. Urban-extent changes are calculated as the percentage change of urban areas in each 1 km<sup>2</sup> pixel, between two successive compilations, in 2000 (DUSAF version 1.1) and in 2007/2009 (DUSAF versions 2.1 and 3.0), depending on the last available data in different sectors of the study area. Positive values indicate an expansion of urban areas, while negative values indicate a reduction of urban areas. According to DUSAF data, urban-area extent changed in the range from -6.7 % to +30.8 % (Fig. 5).



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Radar satellite remote sensing data can be used to identify and delineate urban areas. In fact, satellite radar backscatter is dependent on the number, density, size and material of buildings (e.g., higher backscatter for more buildings, for larger and taller buildings, and for stronger materials like steel rather than wood). Crucially, the satellite global coverage with regular data acquisitions in time spanning over a decadal period allows continuous monitoring of urban changes, and thus enables the trend analysis together with changes in nitrate sources, capturing more detailed variability in annual, interannual, and decadal time scales. Such a satellite dataset has been collected by the SeaWinds scatterometer aboard the QuikSCAT satellite (QSCAT) in the decade of the 2000's. QSCAT backscatter measurement is accurate to 0.2 dB ( $3\text{-}\sigma$ ) (Nghiem et al. 2004), which is equivalent to approximately 1.57% in root-mean-square error, enabling QSCAT to detect not only large and rapid changes as well as small and slow variations. Applied on the original QSCAT backscatter data, the Dense Sampling Method (DSM), based on a newly invented mathematical transform called Rosette Transform (Nghiem et al. 2009), is a breakthrough enabling quantitative measurements of urban parameters (i.e., location, shape, extent, and

307 typology) to map land cover features at a posting pixel scale of 1 km<sup>2</sup>, and to calculate the rate of urban  
308 change in the decadal period of 2000–2009 in every pixel across the world.

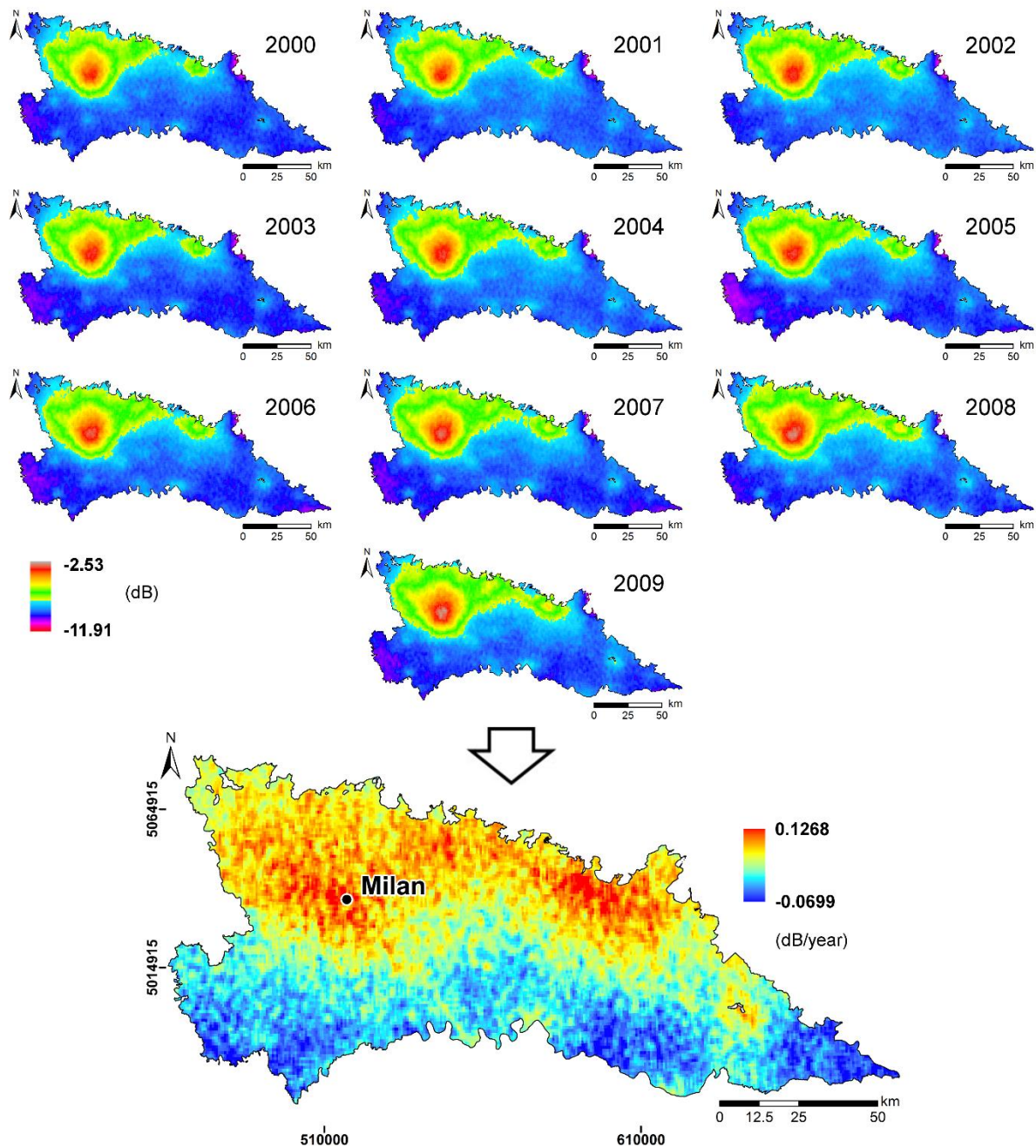
309 In DSM, backscatter signature of an area is characterized by the composition of a spatially-dependent  
310 mean part and a fluctuation part that is a function of location, azimuth angle (buildings are different on  
311 different sides; roads have preferential directions; hilly surfaces in a city, etc.), and any changes in time  
312 (vehicles and people move in a city; there can be rain, snow, hail, etc. at different times in different  
313 sections of a city). Thereby, DSM allows azimuthal and temporal changes to occur, and high-resolution  
314 results from DSM include information from both the mean value and the variability of backscatter at  
315 each location where the Rosette Transform is applied on an ensemble of backscatter data whose  
316 centroids are collocated in each unit area. At the expense of the daily temporal resolution, DSM is a  
317 breakthrough method to increase the spatial resolution in urban areas, where the inherent azimuth and  
318 motion changes invalidate the use of the traditional deconvolution method to enhance the resolution of  
319 satellite remote sensing data.

320 Moreover, advantages of QSCAT-DSM (Nghiem et al. 2009) include the delineation of urban and  
321 suburban contours both in metropolitan and rural areas, and the identification of urban development  
322 both fast and expansive or slow and restrained. Some limitations are due to complex mountainous  
323 topography, persistent snow cover on cold land at high latitudes (e.g., tundra and taiga), or extensive  
324 water surfaces, which affect backscatter signatures, but such factors are ineffective in the study area.  
325 The pointing accuracy of DSM was verified precisely with an accurate overlay of the Príncipe Island  
326 (Gulf of Guinea) on its true geographic location (Nghiem et al. 2009). DSM was validated and used to  
327 accurately delineate urban extent for a number of cities in different countries such as Dallas-Fort Worth  
328 and Phoenix in the United States, Bogotá in Colombia, Dhaka in Bangladesh, Guangzhou and Beijing  
329 in China, and Quito in Ecuador (Nghiem et al. 2009; Nghiem et al. 2014b).

330 The rate of land cover change, including both urban and rural areas, is determined by the slope of the  
331 linear regression with QSCAT-DSM data obtained for each year in 2000–2009, expressed in decibel per  
332 year (dB/year). Positive slope values represent increasing or growth of urban areas, while non-positive  
333 and shallow slope values indicate steady rural areas or natural environments. QSCAT-DSM slope varies  
334 within the range of  $-0.0699$  to  $+0.1268$  dB/year, or equivalently  $-16.0$  to  $+29.6$  %/decade (Fig. 6), as



335 the slope in dB/year can be converted to the 10-year percentage change given by 10 times of  
 336  $100 \times (10^{\text{dB/year}/10} - 1)$ .



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339 In this study's approach to assess impacts on groundwater contamination, the focal method is adapted  
 340 particularly for applications to QSCAT-DSM and hydrogeological data, and to DUSAF as well. The  
 341 algorithm in the focal method considers both the value of each cell and the values of the surrounding  
 342 cells with a deterministic mathematic function. It can account for groundwater flow direction: among

343 the surrounding cells of each cell, only the cells located upstream are considered in the calculation. The  
344 extent of the area of calculation is 9 km<sup>2</sup> for a 3×3 window above each pixel of 1 km<sup>2</sup>.

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### 346 *3.3.2 Urban nitrate sources: natural factors*

347 Natural factors, characterizing geological and hydrogeological conditions of the study area, are  
348 considered to be static in this study. While groundwater depth has a seasonal variability, it has not  
349 significantly changed over the Po Plain in the decade 2000–2009.

350 Soil protective capacity is obtained from existing data (Fig. 7a). It was produced by the Agency of  
351 Services of Agriculture and Forest. This soil variable has been mapped at a 1:250,000 scale to assign  
352 soil in three protective capacity classes: high, moderate and low. The variable describes soil capacity to  
353 reduce water-soluble polluting substances leaching from the surface. It is related to filtering and  
354 buffering capacity because of both mechanical and biological/microbiological activities contributing to  
355 degradation (Masetti et al. 2007).

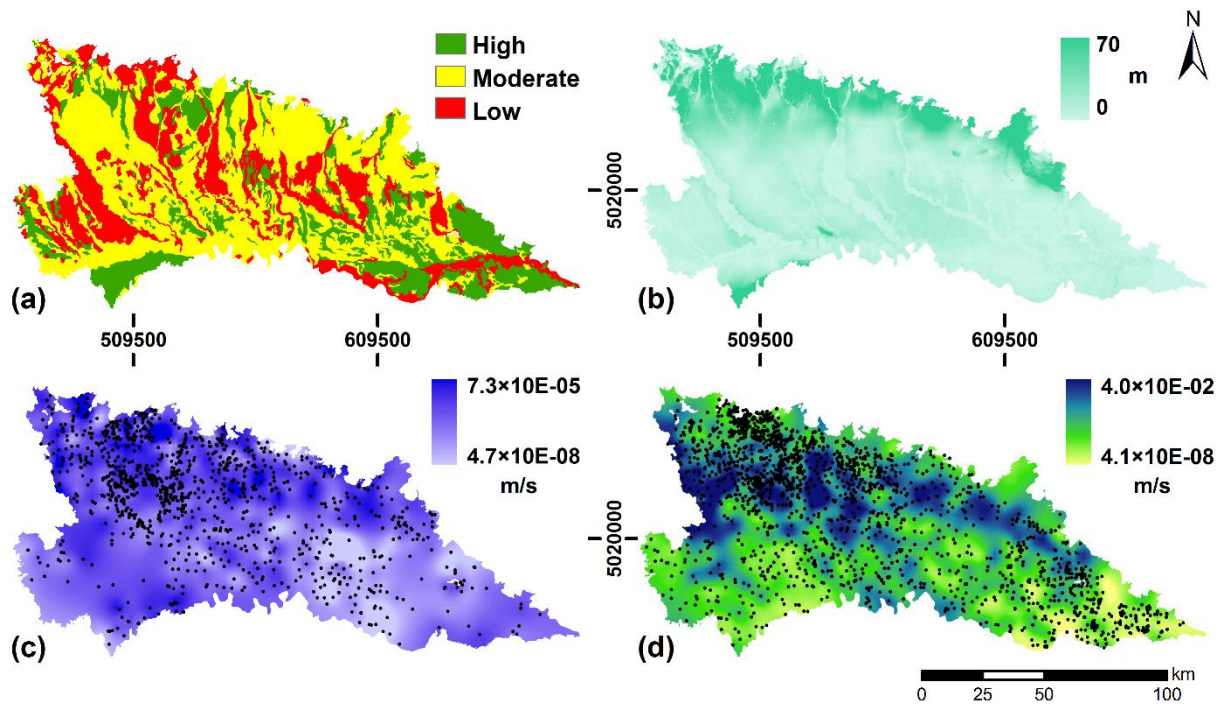
356 The other three hydrogeological variables, characterizing the shallow unconfined aquifer, were obtained  
357 for this study.

358 Groundwater depth was derived from the difference between the topographic level and groundwater  
359 piezometric levels (regional survey in 2003; Fig. 7b). The groundwater depth decreases from north to  
360 south, ranging from values higher than 70 m to less than 2 m. At some local areas, there are groundwater-  
361 fed streams where groundwater depth is reduced to zero.

362 Groundwater velocity was estimated from 1263 wells where pumping tests were available to determine  
363 hydraulic conductivity (Fig. 7c). These values were used together with the local hydraulic gradient to  
364 obtain groundwater velocity. Field data were interpolated through the kriging methodology to obtain a  
365 map of the distribution of groundwater velocity. In the study area, groundwater velocity ranges from  
366  $4.7 \times 10^{-8}$  to  $7.3 \times 10^{-5}$  m/s. Higher values are located in the northern sector and in some areas of the  
367 southwestern sector, while lower values are mainly found in the southeastern sector.

368 Hydraulic conductivity of the vadose zone was determined from 1597 well stratigraphy records (Fig.  
369 7d). For each well, the hydraulic conductivity was calculated with the equivalent vertical permeability  
370 method (Anderson and Woessner 1992), considering the thickness of the layers in the vadose zone in

371 the calculation of the hydraulic conductivity. Data were then interpolated through kriging methodology  
 372 to obtain the map of the distribution of hydraulic conductivity of the vadose zone in the study area.  
 373 Hydraulic conductivity of the vadose zone ranges from  $4.1 \times 10^{-8}$  to  $4.0 \times 10^{-2}$  m/s. Higher values are  
 374 located in the northern sector, especially along the belt of the heads of groundwater-fed streams and  
 375 along the main rivers (Ticino and Adda rivers).



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## 378 4. Results and discussion

### 379 4.1. Impacts observed from the independent variables

380 The contrasts of statistically significant evidential themes enable an assessment of the influence of the  
 381 variables under consideration on groundwater contamination. Contrast values, both for anthropogenic  
 382 and natural factors, are presented in Fig. 8.

383 All three variables, representing urban nitrate sources and their evolution, show a positive correlation  
 384 between the increase of urban areas or population growth and the occurrence of increasing nitrate  
 385 concentration trends in groundwater.

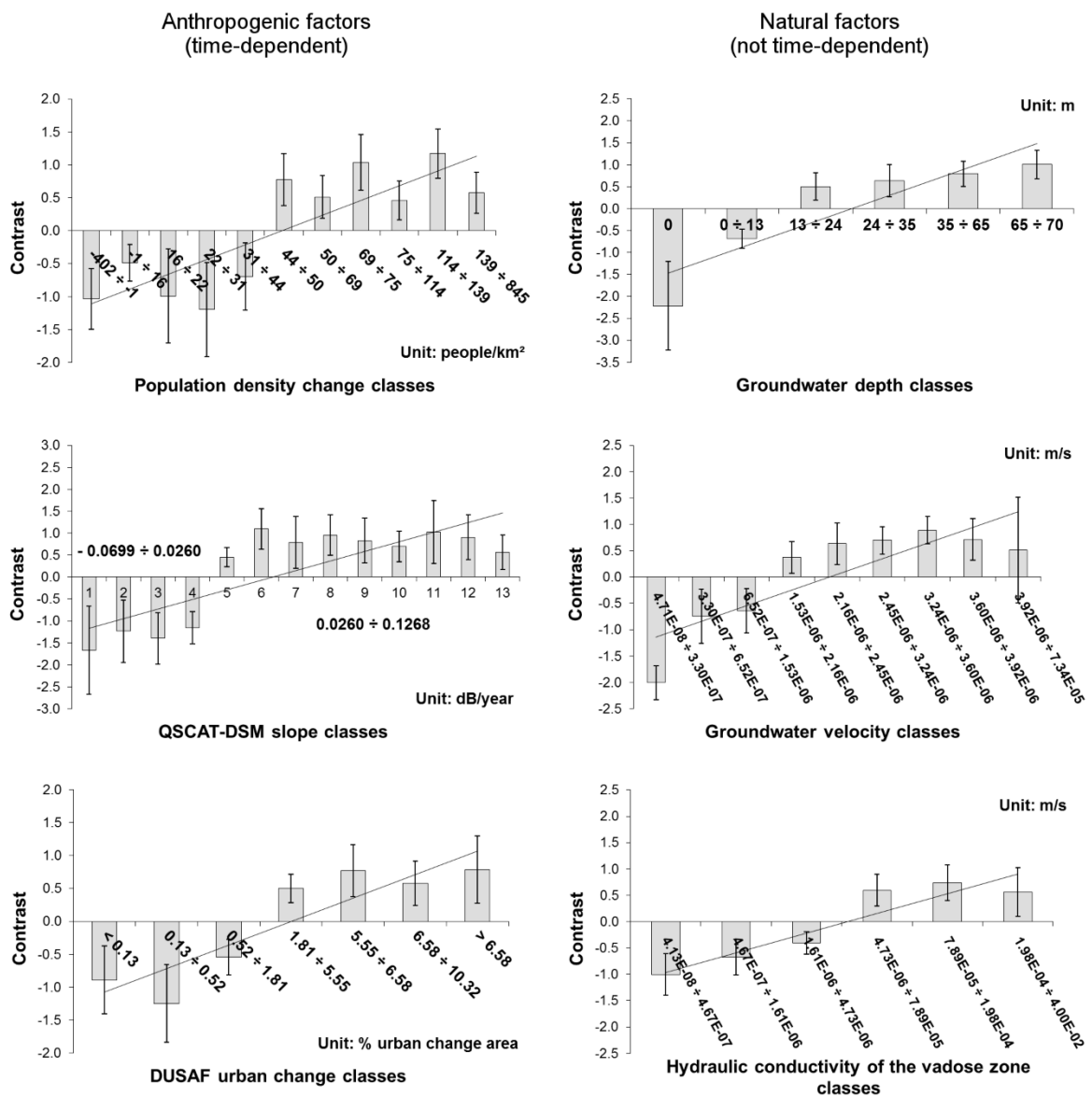
386 Threshold values, corresponding to the transition from negative to positive contrasts, are +44  
 387 people/km<sup>2</sup>, +0.0260 dB/year and +1.81 % over the study period, respectively for population density

388 change, QSCAT-DSM slope and DUSAF urban extent change. Observed from these variables, classes  
 389 with positive contrasts are clustered in the northern sector, while classes with negative contrasts are  
 390 mainly in the southern sector.

391 Soil protective capacity is not statistically significant. Also, it does not show a discernable correlation,  
 392 with negative contrasts for low and high classes and a positive contrast for moderate class.

393 Groundwater depth reveals that large values of water table depth are positively related to increasing  
 394 concentration trends, while low values (close to surface or less than 13 m) are negatively associated.

395 Groundwater velocity and hydraulic conductivity of the vadose zone show positive correlations and the  
 396 threshold values are about  $1.5 \times 10^{-6}$  m/s and  $4.7 \times 10^{-6}$  m/s, respectively.



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**4.2. Response themes and vulnerability maps**

In order to evaluate the reliability of each variable as a proxy of urban nitrate sources, three response themes were obtained and compared (Fig. 9). Each response theme considers one of the three urban variables, and the three statistically significant evidential themes represent the associated natural factors (Table 2).

Each response theme was 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). Five classes were identified with the degree of groundwater vulnerability increasing from 1 to 5. This number was selected based on the general criteria used to identify vulnerability classes (Sorichetta et al. 2011) and on visual analytic techniques (Cowan 2001).

It is important to note that these response themes are time dependent. This means that groundwater vulnerability classes reflect the tendency toward a deterioration of the quality of the aquifer rather than the absolute severity of the aquifer contamination in a static condition.

**4.3. Reliability and validation of the maps**

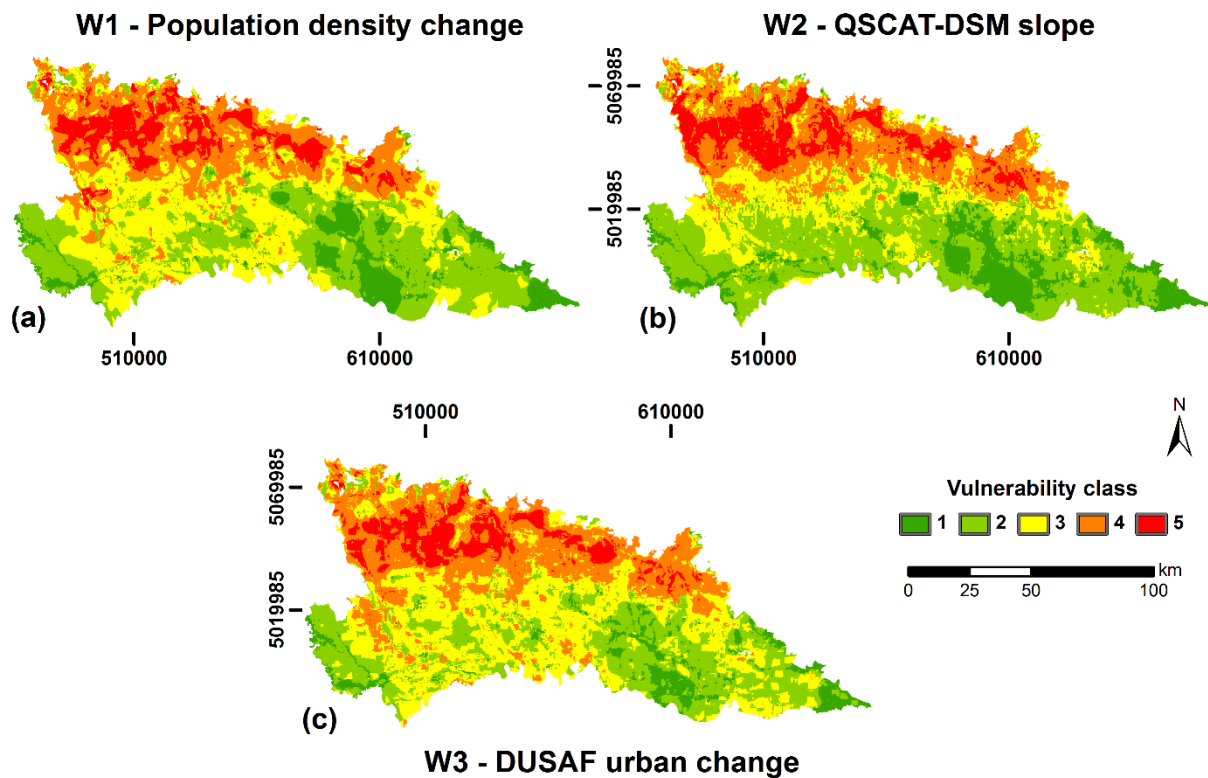
The general quality of each response theme (i.e., post probability map) can be evaluated with the Area-Under-the-Curve (AUC) value. AUC is a direct measure of the performance of the statistical approach, and is given by the area under the curve (integral) for cumulated area/cumulated training points expressed in percentage. The calculated AUC values are presented in Table 2 showing the consistent quality of the different maps.

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426 **Table 2** Combination of evidential themes used to obtained response themes and AUC values (pop =  
427 population density change, gwd = groundwater depth, gvw = groundwater velocity, hcv = hydraulic  
428 conductivity of the vadose zone, QSCAT-DSM = land use changes derived from satellite data, DUSAF  
429 = land use changes derived from aerial photographs)

Response theme	Combination of evidential themes	AUC value
W1	pop, gwd, gvw, hcv	74.4 %
W2	QSCAT-DSM, gwd, gvw, hcv	74.3 %
W3	DUSAF, gwd, gvw, hcv	73.7 %

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433 Then, the reliability of each classified map was evaluated again by considering its overall performance  
434 in classifying the occurrences. Two statistical validation procedures were used: (1) frequency of training  
435 set, and (2) average nitrate concentration trend of all wells in each vulnerability class.

436 The evaluation of the frequency,  $F$ , is expressed by the ratio:

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$$F = (N_{Wj}/T_{Wj}) \quad (5)$$

438

439 where  $N_{Wj}$  is the number of “increasing” wells in a vulnerability class  $j$  and  $T_{Wj}$  is the total number of  
440 wells in the same class  $j$ . This technique adds new information to the validation process because it  
441 includes also the wells not used in the modeling. Frequency is expected to increase monotonically as the  
442 degree of vulnerability increase. The expected trend is verified. In fact, for all the three vulnerability  
443 maps, there are no “increasing” wells in the lowest vulnerability class and the highest frequency of  
444 “increasing” wells is in the highest vulnerability class (Fig. 10).

445 The evaluation of the average nitrate concentration trend of all wells,  $C_{AVG}$ , is expressed as:

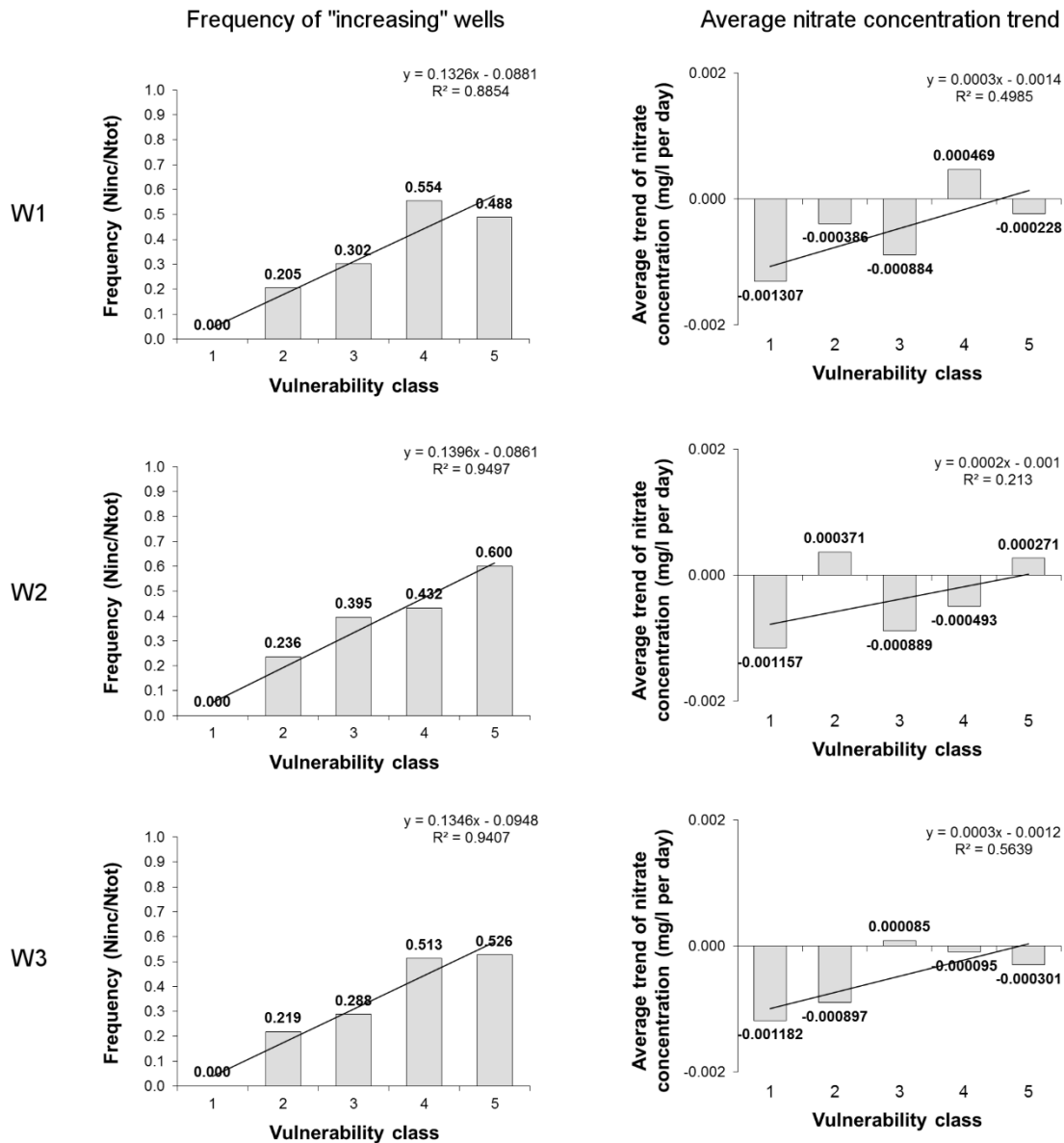
446

$$C_{AVG} = \frac{\sum_{i=1}^{T_{Wj}} C_{ij}}{T_{Wj}} \quad (6)$$

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448 where  $C_{ij}$  is the nitrate concentration trend of well  $i$  in the vulnerability class  $j$ , and  $T_{Wj}$  is the total number  
449 of wells in the same class  $j$ . This analysis was carried out using all wells stored in the database. The  
450 concentration should monotonically increase as the degree of vulnerability increases and the central  
451 vulnerability class should give a value close to the overall mean value. Despite some anomalies, all three  
452 histograms show a direct correlation between average nitrate concentration trend and the degree of  
453 vulnerability (Fig. 10).

454 With these two techniques, the quality of each vulnerability map was evaluated based on the slope  
455 coefficient of the regression line and the regression coefficient, so that a map should be deemed reliable  
456 if it passes these tests.



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460 **4.4. Spatial agreement between maps**

461 A spatial agreement is quantitatively evaluated through a pixel-by-pixel analysis representing the

462 difference, expressed as percentage, in the unit-cell classification for the three vulnerability maps (Fig.

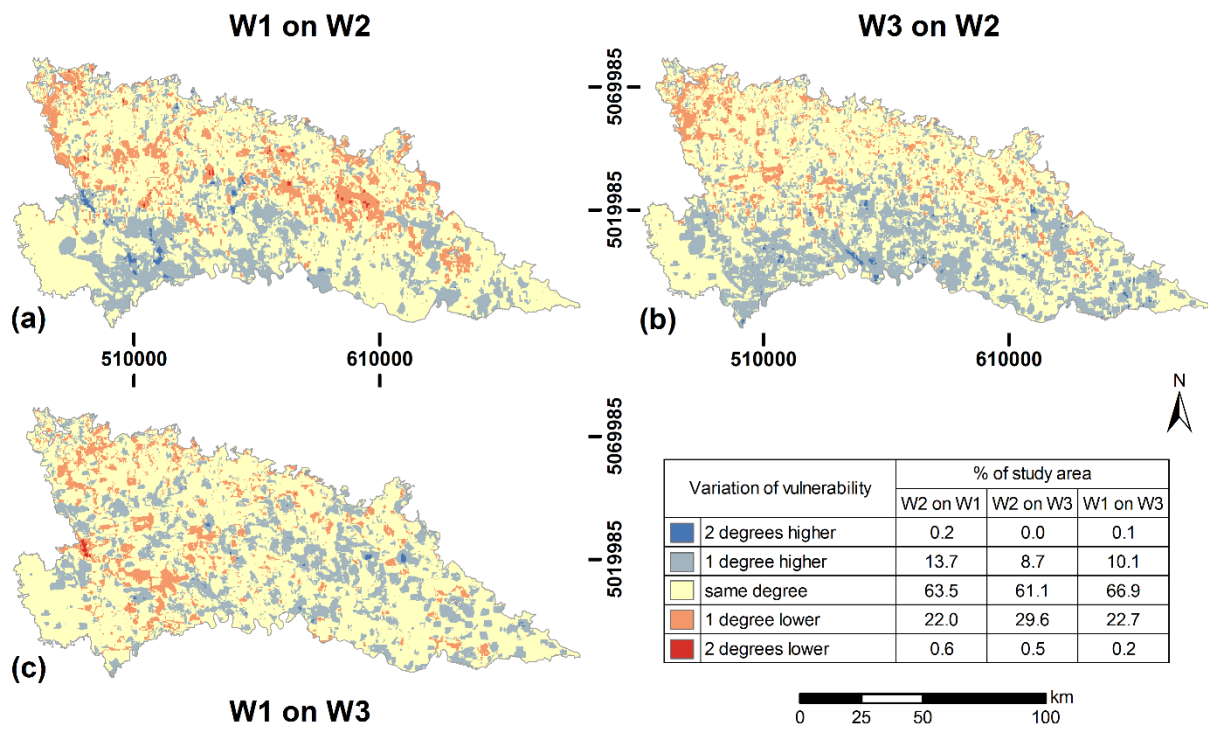
463 11). Results from this analysis show a high level of agreement between the maps in the paired map-to-

464 map comparison: almost 61–67 % of the study area is classified with the same degree of vulnerability,

465 33–38 % is classified within a difference of one degree of vulnerability, while only 0.3–0.8 % within a

466 difference of two degrees of vulnerability.





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469 Another method to evaluate the reliability of each vulnerability map is overlaying each map with the  
 470 classes of its urban change variable with positive contrast values to examine their consistency (Fig. 12).

471 Map W2, obtained using QSCAT-DSM slope, is the only one where the highest vulnerability classes  
 472 are consistently overlain by the classes of urban extent change variable with positive contrast values.

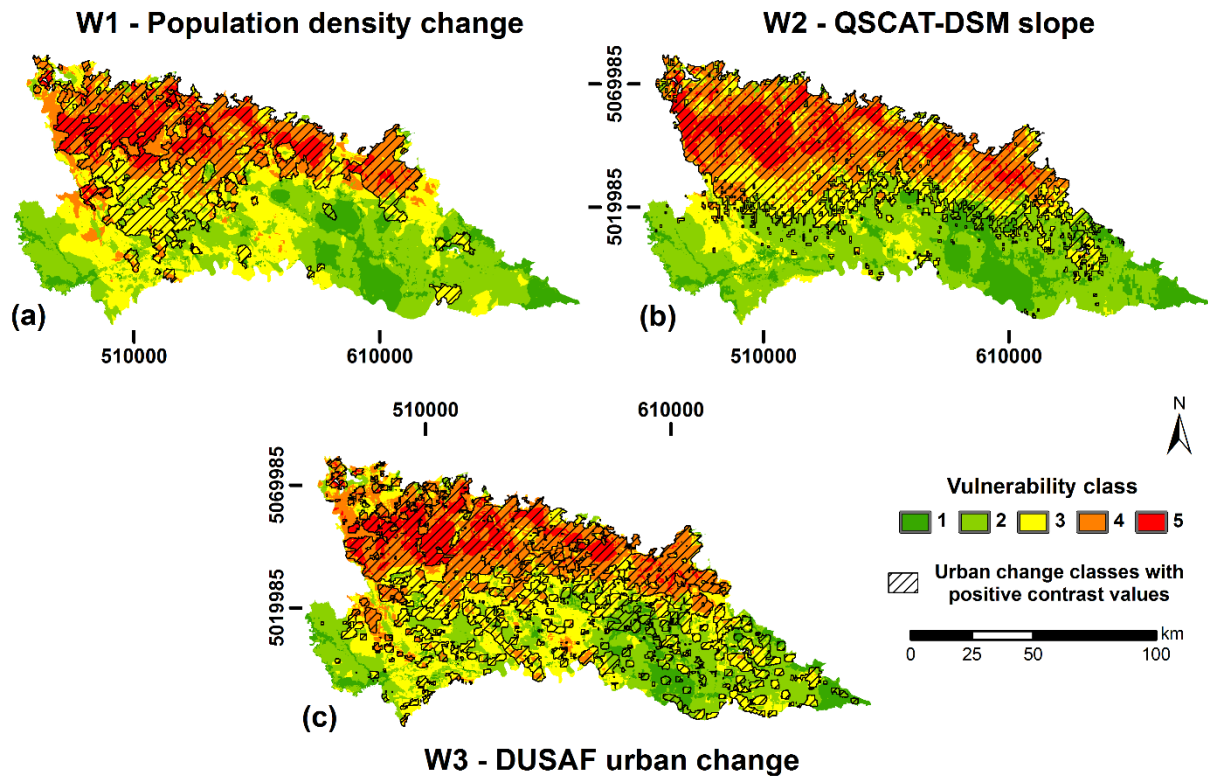
473 Instead, in the other two cases there are anomalous mismatches. In Map W1, obtained using population  
 474 density changes, some cities (like Monza or Brescia) show negative contrast values, meaning that their

475 population density change is lower than +44 people/km<sup>2</sup>. In Map W3, obtained using DUSAF maps,  
 476 some areas in the northern sector show negative contrast values (like Milan), while agricultural areas of

477 the southern sector are characterized by positive contrast values (e.g., Provinces of Cremona and  
 478 Mantua). The first anomaly could be explained by the urban sprawl phenomenon, with residential

479 citizens moving from the largest cities to the smallest cities, while the rate of urbanization is increasing  
 480 almost everywhere. The second anomaly could be caused by the focal application applied to the binary

481 land-use categorization in the DUSAF maps: small changes in urban extent cannot be accurately  
 482 detected in large urban areas.



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485 **4.5 Discussion**

486 The direct correlation for all of the three anthropogenic evidential themes means that increasing nitrate  
 487 concentration is related to areas of urban development or population increase, in agreement with  
 488 Stevenazzi et al. (2014).

489 The three urban variables consistently identify that the most important changes are clustered around the  
 490 biggest cities or in the northern sector where cities and industries in the Lombardy region are mostly  
 491 located, while the southern sector primarily consists of agricultural fields. There are anomalies in  
 492 population density changes because some sectors in large cities have decreasing trends while small  
 493 towns show significantly increasing trends. These changes indicate a tendency that people like to move  
 494 away from over-crowded urban areas and sprawl to more open suburban areas with natural or  
 495 agricultural surroundings (EEA 2006).

496 The direct relationship between groundwater depth and increasing concentration trends is consistent  
 497 with earlier static observations for shallow aquifer in USA (Nolan 2001; Nolan et al. 2002) and in the

498 Province of Milan (Sorichetta et al. 2013). The explanation can be found in bio-geochemical conditions  
499 of the vadose zone. In fact, very shallow water table leads to waterlogged conditions conducive to  
500 denitrification processes, in which denitrification rates tend to decrease as water table depth increases.  
501 The result in this study supports this hypothesis and indicates that nitrate concentration changes are  
502 related to bio-geochemical activities in the vadose zone.

503 Groundwater velocity and hydraulic conductivity of the vadose zone are two hydrogeological variables  
504 that influence the movements of contaminants from surface to aquifers and within aquifers. The first  
505 controls transport and dilution of contaminants within aquifers, and the latter controls the rate at which  
506 a contaminant can reach groundwater. In terms of increasing concentration trends in the study area,  
507 positive correlations mean that the transport process is generally prevalent over the dilution one, both in  
508 groundwater and in the vadose zone. Static analyses (Sorichetta et al. 2013) have found that these  
509 variables have positive correlations with the occurrence of high nitrate concentrations. From this study,  
510 increasing concentration trends are shown to relate to increasing groundwater velocity or increasing  
511 hydraulic conductivity in the vadose zone. Thus, both static and time-dependent analyses confirm the  
512 impacts of these hydrogeological factors on the distribution of contaminants, which are necessary to  
513 include in groundwater vulnerability assessment.

514 Vulnerability maps are calibrated and validated. The similarity in calculated high AUC values for Maps  
515 W1, W2 and W3 asserts the consistent quality of the maps. Histograms of frequency are excellent for  
516 all the maps, with a monotonic increase corresponding to higher vulnerability. Nevertheless, according  
517 to the criteria used in evaluating the frequency histograms, Map W2 can be considered the one that  
518 performs best. In fact, it has the highest regression coefficients and the one with the highest frequency  
519 of impacted wells in the highest vulnerability class.

520 Histograms of average nitrate concentration trends show a general positive trend, although with low  
521 values. Maps W1 and W3 have the highest angular and regression coefficients, but only Map W2 shows  
522 the mean positive value in the higher vulnerability class. No map presents the mean or median value of  
523 the whole distribution as average concentration trend in the central vulnerability class.

524 In summary, QSCAT-DSM can be successfully used as a proxy for nitrate contamination from urban  
525 sources and, among the three obtained vulnerability maps, the map that uses QSCAT-DSM slope to  
526 characterize the evolution of urban nitrate sources (Map W2) appears to be the best.

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## 529 **5. Conclusions**

530 Introducing the time variable to monitor trends in groundwater vulnerability assessment is an innovative  
531 approach to study the evolution of non-point-source pollution in an area and to forecast future changes.

532 With the application of a Bayesian spatial statistical approach, it is found that:

- 533 – Natural factors, such as groundwater depth, groundwater velocity and hydraulic conductivity of the  
534 vadose zone, influence groundwater vulnerability, confirming results from previous studies on  
535 nitrate contamination (Nolan 2001; Sorichetta et al. 2013);
- 536 – The innovative use of QSCAT-DSM satellite data (Nghiem et al. 2009) in the analysis enables the  
537 production of a time-dependent vulnerability map, which is compared with two other vulnerability  
538 maps obtained using different time-dependent factors related to urban changes (i.e., population  
539 density from census and changes in land use derived from the DUSAF database);
- 540 – All of the time-dependent factors indicate that increasing nitrate concentration occurs in areas  
541 related to urban development or population increase;
- 542 – The calibration and validation procedures affirm all of the three vulnerability maps have a high  
543 reliability, while the one obtained with QSCAT-DSM is the better one.

544 The latter result is remarkable for those areas where there are insufficient or inaccurate data for  
545 population or land use and their changes, and thus satellite observations of urban change become  
546 particularly useful. Moreover, QSCAT-DSM data have the advantages of a worldwide coverage, a  
547 continuous data collection and an adequate resolution without spatial gaps.

548 In conclusion, the approach developed in this study for the first time allows the inclusion of the time  
549 variable in groundwater vulnerability assessment with the use of innovative remote sensing data to carry  
550 out a quantitative statistical analysis of groundwater quality changes.

551 New approaches to combine groundwater vulnerability maps obtained by explicitly accounting for the  
552 time variable with traditional vulnerability maps should be advanced for better intervention strategies  
553 and for more efficient policy measures. Indeed, their combined use would allow one to not only identify  
554 already highly contaminated areas where expensive reactive remediation measures need to be  
555 implemented, but also to detect areas where pro-active interventions need to be planned.

556 With the method demonstrated in this study, existing and future satellite scatterometer data can be used  
557 to make and update maps of groundwater vulnerability as urbanization accelerates across the world.

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706 FIGURE CAPTIONS:

707

708 **Fig. 1** (a) Location of the study area; (b) well-monitoring network; (c) examples of well locations in  
709 urban (1) and rural (2) environments as marked by *1 and 2 next to the square boxes* on the map in panel  
710 (b) where the photographs were taken during the POPLEX field campaign in May 2014. Coordinates  
711 refer to WGS 1984 – UTM Zone 32N projection

712

713 **Fig. 2** Hydrogeological scheme along the N-S section marked by the *grey line* on the map in Fig. 1b  
714 (modified from Regione Lombardia and ENI 2001)

715

716 **Fig. 3** Frequency histogram of nitrate concentration trend

717

718 **Fig. 4** Population density maps, at municipality level, in (a) 2011 and (b) 2001, and (c) the final map  
719 obtained as the difference between the two maps (a) and (b). Coordinates refer to WGS 1984 – UTM  
720 Zone 32N projection

721

722 **Fig. 5** DUSAF urban area extent maps in (a) 2007/09 and (b) 2000, and (c) the final map obtained by  
723 calculating the percentage change of urban areas between the two maps (a) and (b) at a resolution of 1  
724 km<sup>2</sup>. Coordinates refer to WGS 1984 – UTM Zone 32N projection

725

726 **Fig. 6** QSCAT-DSM backscatter maps, at a posting of 1 km<sup>2</sup>, from 2000 to 2009, and the final map of  
727 the linear regression slope. Coordinates refer to WGS 1984 – UTM Zone 32N projection

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731 **Fig. 7** Natural factors maps: (a) soil protective capacity; (b) groundwater depth; (c) groundwater  
732 velocity; (d) hydraulic conductivity of the vadose zone. *Dots* in panels (c) and (d) represent the locations  
733 of pumping test sites and well stratigraphies used to map the spatial distribution of groundwater velocity  
734 and hydraulic conductivity of the vadose zone, respectively. Coordinates refer to WGS 1984 – UTM  
735 Zone 32N projection

736

737 **Fig. 8** Contrasts and error bars of the statistically significant classes of each evidential theme used to  
738 generate the maps in Fig. 9

739

740 **Fig. 9** Vulnerability maps obtained using static variables, representing natural factors, with (a)  
741 population density change, (b) QSCAT-DSM slope, and (c) DUSAF-based urban extent change as time-  
742 dependent variables. Coordinates refer to WGS 1984 – UTM Zone 32N projection

743

744 **Fig. 10** Histograms of the frequency of the “increasing” wells (left) and of the average nitrate  
745 concentration trend (right) in each vulnerability classes of the maps in Fig. 9. The degree of vulnerability  
746 increases from class 1 to class 5

747

748 **Fig. 11** Variation of vulnerability from map-to-map: (a) map W1 on map W2; (b) map W2 on map W3;  
749 (c) map W1 on map W3. The variation is expressed as the agreement in percentage between the  
750 vulnerability depicted in the first map with respect to the one depicted in the second map. Coordinates  
751 refer to WGS 1984 – UTM Zone 32N projection

752

753 **Fig. 12** Vulnerability maps obtained using (a) population density change, (b) QSCAT-DSM slope and  
754 (c) DUSAF urban extent change as time-dependent variables, overlain by the corresponding evidential  
755 theme classes with positive contrast values. Coordinates refer to WGS 1984 – UTM Zone 32N projection

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