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THEMATIC ISSUE

Evaluating upward trends in groundwater nitrate concentrations: an example in an alluvial plain of the Campania region (Southern Italy)

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Abstract

To achieve the objectives of the Water Framework Directive's, the identification and reversal of significant upward trends in pollutant concentrations in groundwater is crucial. A statistically significant increase in a pollutant, groups of pollutants or pollution indicators in groundwater bodies, means an upward trend in concentration values, calculated using a recognized statistical method, at least 90% significant. Following this aim, we focused on a groundwater body of southern Italy, with high concentration of nitrate and, therefore, defined "at risk", on the basis of its bad quality status. In this area, we calculated the trends of the time series of nitrate concentrations in groundwater. We described the adopted procedure for trend analysis, following the guidelines proposed by the Italian Institute for Environmental Protection and Research (ISPRA). We used the Mann–Kendall method for calculating the statistical significance of the upward trend of time series and, successively, the Sen method for estimating the value of the trend (angular coefficient). We applied these methods also to forecast nitrate concentrations in groundwater in 2021 and in 2027. We found differences in the sampling stations in terms of groundwater quality and trends, as a result of different environmental factors. Peculiarly, the location of the wells presenting upward trends seems to be in areas with high population density and intensive agriculture.

Keywords Groundwater quality · Nitrate pollution · Nitrate trends · Italy

Introduction

The assessment of long-term groundwater quality trends is a subject of growing interest. The 2000/60/EC directive of the European Parliament and of the Council (WFD EU 2000) sets the goal of achieving a ''good status'' for all Europe's surface waters and groundwater by 2015. Where this objective was not reached, the achievement of good status has been extended to 2021 or, at the latest, 2027, as part of specific "River Basin Management Plans—(RBMPs)". According to the water framework directives, member states must establish surveillance monitoring programs to provide information for the assessment of long-term changes in natural conditions and those resulting from widespread anthropogenic activity. Land-use practices can influence groundwater quality. In the case of nitrate, background concentrations are low, typically less than 1 mg/L (Burow et al. 2010; Daughney and Reeves 2005; Morgenstern and Daughney 2012). High concentrations and their evolution are generally related to anthropogenic sources as agricultural practices (Hansen et al. 2017), animal manures, inefficient sewerage systems, etc. In this framework, the detection and evaluation of the

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trends due to anthropogenic activity is a primary issue. The identification of periods and locations, where increasing pollution trends occur, would allow water management authorities to take adequate measures.

This is not an easy problem, since those trends may be hidden by the effect of external factors, such as river flow, seasonality, water temperature and precipitation.

Many researchers utilised time series statistical approaches to evaluate trends using different parametric (distribution-dependent) and non-parametric (distribution-free) methods. The nonparametric Mann–Kendall test (Mann 1945; Kendall 1975) is probably the most often used method in trend estimation of water quality and hydroclimatic time series. The Mann–Kendall test should be preferred to parametric trend tests when using multiple data sets, because it does not require a priori assumptions related to underlying distributions (Hirsch et al. 1982; Hirsch and Slack 1984) and it does not specify whether a trend is linear or non-linear (Yue et al. 2002).

Following this aim, in the present study the trends of the time series of nitrate concentrations were calculated for a groundwater body of the Campania region classified having a bad quality status due to nitrate concentration and, therefore, defined “at risk”, applying the guidelines (Procedure A) proposed by ISPRA (Italian Institute for Environmental Protection and Research). The study especially focuses on a large coastal plain, the “Volturno-Regi Lagni” plain (P-VLTR), with intensive agriculture and high nitrate levels in groundwater since the 1990s, and it also represents a follow-up of a previous study carried out with a different method and using a smaller data set (Ducci et al. 2019a, b).

The trend analysis procedure is based on the Mann–Kendall method for the statistical significance calculation of the nitrate trends and the Sen method for the trend’s value estimation.

Study area and data sources

Study area

The Campania region (southern Italy) is divided in 80 significant groundwater bodies (here and after GWB).

The Volturno-Regi Lagni plain (P-VLTR) GWB is the largest GWB of Campania, with an area of more than 1000 km^2, crossed by the Volturno river (Fig. 1). This GWB includes two porous aquifers: (1) the first is shallow and phreatic, constituted by 10–20 m of alluvial-pyroclastic deposits with grain size varying from silt to sand and with low–moderate permeability and (2) the second is the main aquifer, composed by coarse-grained sandy sediments (thickness: 60–70 m), of alluvial, pyroclastic and marine origin, with permeability from moderate to high. The first aquifer is not present everywhere and it is absent in drought years. The two aquifers are separated by a tuff, the Campanian Ignimbrite (39 ky B.P.), with a maximum thickness of 50–60 m, which plays the role of a semi-confining or confining layer, depending on the thickness (very thin near the Volturno river and along the coast), and on the welding status. Figure 1 shows the very low potentiometric slope of this main aquifer. Clayey-sandy deposits, prevalently impermeable, form the base of this aquifer.

The hydrogeochemistry of the GWB depends on the volcanic origin of the deposits and on the groundwater flow in the plain (Fig. 1). The alkaline content increases from the limestone mountains (feeding the aquifers of the plain) towards the coastal areas. The volcanic origin causes high fluoride content (almost everywhere > 1.5 mg/L) and, in the southern part, high arsenic content (As > 10 μg/L). In some parts of the GWB, prevalently along the Volturno river, there is a reducing environment with high levels of manganese and iron (Corniello and Ducci 2014; Ducci et al. 2016).

Since the previous century, a severe contamination by nitrate (more than 50 mg/L) has been recognized in the P-VLTR GWB, also in the main, protected aquifer. Nitrate contamination in groundwater is present everywhere, prevalently due to manure spreading and/or sewage leaking from collectors or septic tanks, as revealed by isotopic studies (Ducci et al. 2019c).

The CORINE Land Cover inventory (CLC 2012) indicates in this plain with high population density (often > 250 inhabitant/km^2), these main land-use units: arable land, permanent crops and urban areas (Fig. 2).

Data sources

For the trend analysis, the present study uses the GWB quality monitoring network of the Environmental Protection Agency of Campania (ARPAC). At least biannually, groundwater of the monitoring network points (wells and springs) are analysed with the aim to establish the chemical status classification, according to the European and Italian regulations (Directive 2006/118/EC and Italian D.lgs. 30/2009).

In the examined GWB, the nitrate concentration measurements were selected from 16 monitoring wells (Fig. 1), collected between 2003 and 2017 and with approximately two nitrate analyses per year. Monitoring well Bvr12 had the least number of samples (13), while well Bvr23 had the maximum number of samples (25). On average, there were about 20 samples per monitoring well.

Nitrate values below the laboratory’s analytical detection limit were replaced in the data set with values equal to one-half the value of the detection limit (Scarsbrook and McBride 2007; ISPRA 2017). Only 3.3% of the values used for spatial and trend analysis have been replaced.

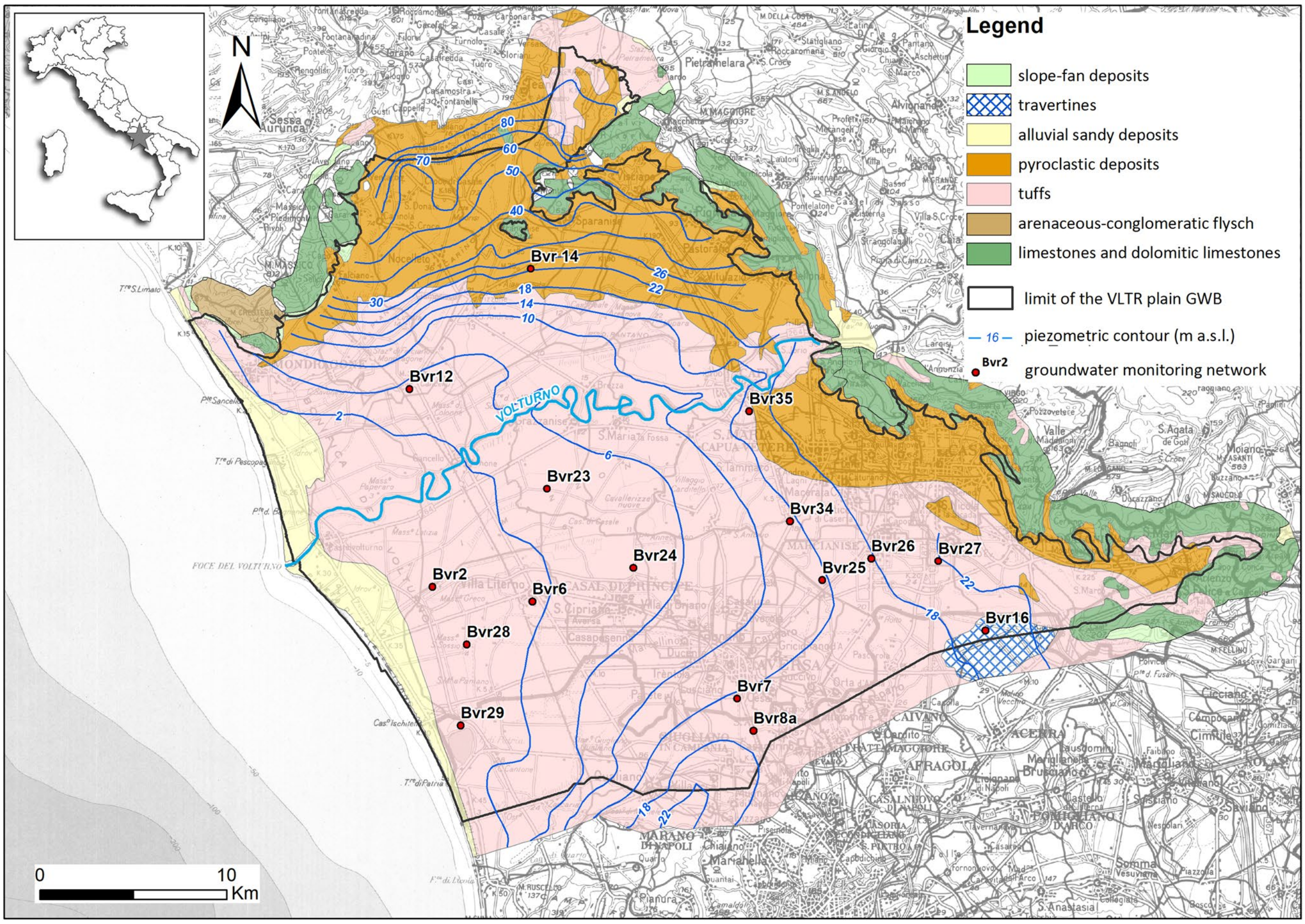

Fig. 1 Hydrogeological map of the groundwater body of the "Volturno-Regi Lagni" plain (P-VLTR) and the quality monitoring network of the Environmental Protection Agency of Campania (ARPAC)

All the examined monitoring wells draw water from the main, deeper aquifer, as testified by the depths reported in Table A in the Supplementary material. The depth below the Campanian Ignimbrite (about 25–30 m b.g.l.) has been considered to select the wells drawing water from the deeper aquifer, due to the lack of information about the construction scheme of these wells and especially about the position of the well screen.

For the spatial analysis, the present study uses data already published (Corniello and Ducci 2014) and web resources of the Campania region (http://www.campaniatrasparente.it/gis.php).

It is essential to highlights the differences between this two kinds of data: data used per trends are referred to few single wells with many measures over time, while spatial data used for building the nitrate contour surfaces are based on a groundwater wells monitoring network not including the points used for the trend analysis; moreover, the monitoring networks in 2004 (about 200 wells) and 2017 (132 wells) are different, because they are based on different groundwater wells.

Methods

Chemical methods

Water sampling for groundwater quality monitoring is done by ARPAC, according to ISO 5667-11 (2009) standard.

In the laboratory, ion chromatography for the determination of dissolved ions follows the ISO 10304 (1995, 2007) standards and its specifications suggested by APAT-IRSA, C. N. R. (2003). The Dionex (now Thermo Scientific™ Dionex™) ion chromatography is the instrument used for performing the analytical part, supported by the CHROMELEON software ver. 7.0 for control, automation, and data processing. The methods are deeper described in Ducci et al. (2019a).

Several nitrate values derive from chemical laboratory analyses of the main ions with an error in the ion balance < 5% (see Table A in the Supplementary material), while in some cases, only specific parameters have been analysed. The temperature and EC were measured on site by multi-parametric sensors.

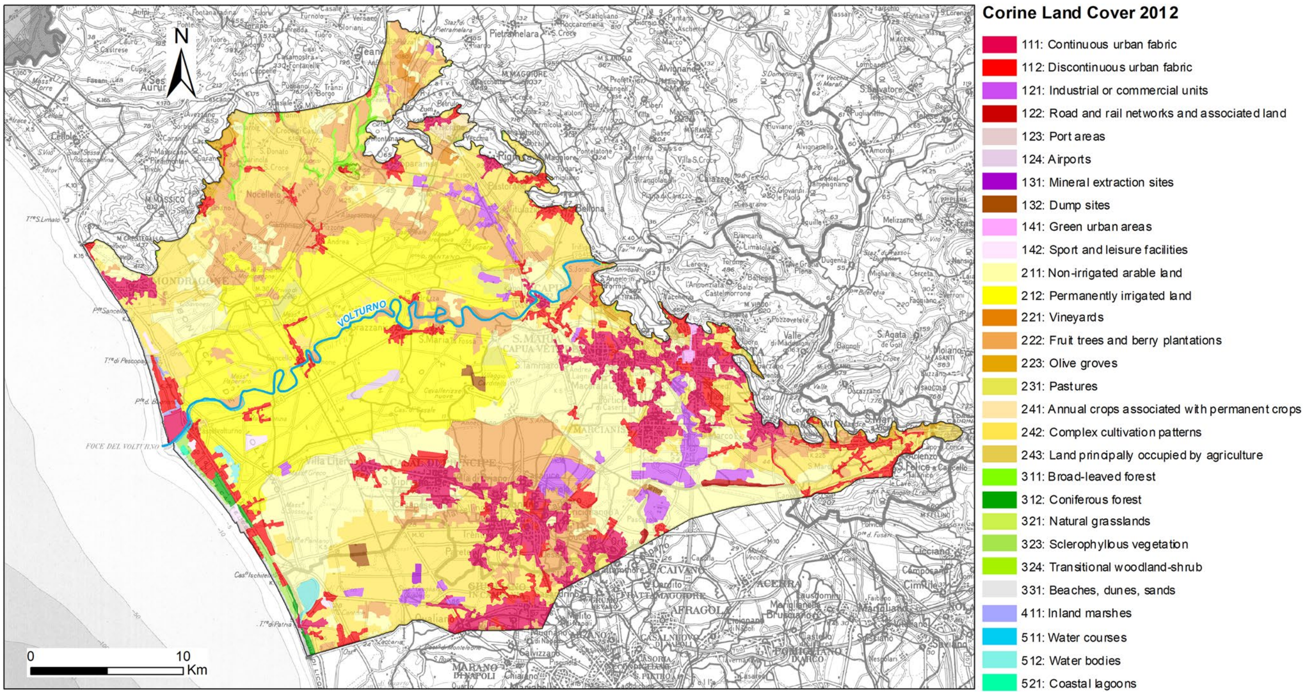

Fig. 2 Corine Land Cover (CLC), Level 3 of the year 2012 (http://land.copernicus.eu/pan-european/corine-land-cover/clc-2012)

Data analysis

To investigate the spatial variation of nitrate contamination, a comparison between nitrate distribution in 2004 and 2017 were done comparing the raster maps of the interpolations and classifying the differences as unchanged (from − 10 to 10 mg/L), decreasing (from − 50 to − 10 mg/L), increasing (from 10 to 50 mg/L) and highly increasing (from 50 to 126 mg/L).

To estimate the temporal variation in nitrate concentrations, the nitrate trends in each monitoring point were calculated by adopting the guidelines suggested by the Italian Institute for Environmental Protection and Research (ISPRA 2017) on the basis of the 2016 Decree of the Italian Ministry of the Environment (D.M. Env. 2016). A flow chart of the procedure is shown in Fig. 3:

1. Data set: the consistency of the data set for each monitoring point (minimum 8 years of sampling and the last measurement not older than 3 years) is indicated in the first box.
2. Evaluation of the increasing trend statistically significant (*p* value = 10%) for each monitoring point and estimation of the area covered by the monitoring points showing statistically significant upward trend.
3. Estimation of the effective value of the trend (angular coefficient) for the monitoring points showing statistically significant upward trend.
4. Evaluation of nitrate concentrations in the future (2021 and 2027 scenarios) and comparison with TV (Threshold Values).

In Italy the recommended TV to achieve the good standard of groundwater chemical quality for nitrate is 50 mg/L (Decreto Legislativo 16 marzo 2009, n. 30).

The nitrate trends of the 16 P-VLTR groundwater monitoring points were identified using the non-parametric Mann–Kendall test (MK).

The test compares the relative magnitudes of sample data rather than the data values themselves (Gilbert 1987). One benefit of this test is that the data need not conform to any particular distribution, but there must be no serial correlation in the data for the resulting p values to be correct (Lee and Lee 2003; Kahya and Kalayci 2004; Zhang et al. 2005; AquaTerra 2004). The procedure assumes that there exists only one data value per time period. When multiple data points exist for a single time period, the mean value was used in this analysis (ISPRA 2017).

The trend analysis was conducted by first examining each variable for seasonality (i.e., two seasons) computing the lag-one autocorrelation coefficient r_1 at 10% significance level. If seasonality was evident trend analysis was carried out using a Two-Season Seasonal Kendall test (Hirsch et al. 1982; Hirsch and Slack 1984) with the seasons classified as June to November (summer/autumn) and December to May (winter/spring). Where no seasonality

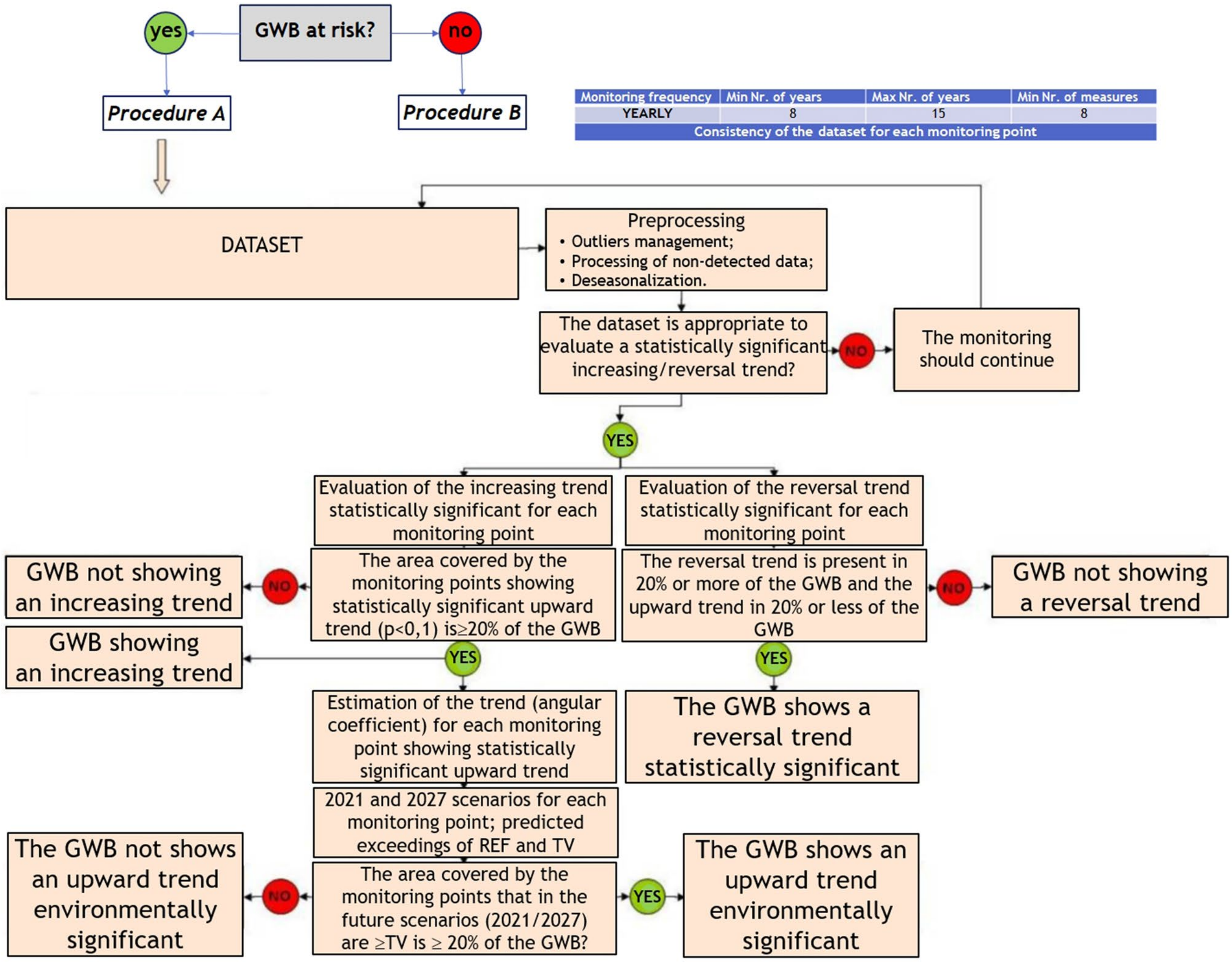

Fig. 3 Procedure for the evaluation of the increasing and reversal trends of contaminants for the groundwater bodies at risk (modified from ISPRA 2017)

was evident trend analysis was performed using the MK test.

The MK test statistic S is calculated in the following:

$$\operatorname{sgn}\left(x_j - x_i\right) = \begin{cases} 1; & If \quad x_j > x_i \\ 0; & If \quad x_j = x_i \\ -1; & If \quad x_j < x_i \end{cases} \tag{1}$$

$$S = \sum_{i=1}^{n-1} \sum_{j=i+1}^{n} \operatorname{sgn}\left(x_j - x_i\right), \tag{2}$$

where *xi* and *xj* are the data values at times *i* and *j*, *n* indicates the length of the data set.

A very high positive value of *S* is an indicator of an increasing trend, and a very low negative value indicates a decreasing trend. However, it is necessary to compute the probability associated with *S* and the sample size, n, to statistically quantify the significance of the trend. According to the guidelines of ISPRA it has been assumed a probability level of significance of 90% (p value = 10%). If the computed probability is less than the level of significance, there is no trend.

For a time series of more than equal to 10 years (i.e., $n \geq 10$), data are approximately normally distributed (variance $\sigma^2 = 1$ and mean $\mu = 0$) and another statistical parameter is the standard *Z* value calculated as

$$Z = \begin{cases} \frac{S-1}{\sqrt{Var(S)}}; & if \quad S > 0 \\ 0; \quad if \quad S = 0 & \\ \frac{S+1}{\sqrt{Var(S)}}; & if \quad S < 0 \end{cases} \tag{3}$$

in the expression Var (S) is variance and it is given as

$$Var(S) = \frac{n(n-1)(2n+5) - \sum_{i=1}^{p} t_i\left(t_i - 1\right)\left(2t_i + 5\right)}{18}, \qquad (4)$$

where n is the number of data points, p is the number of tied groups (a tied group is a set of sample data having the same value), and t_p is the number of data points in the pth group. If there are not tied groups, this summary process can be ignored.

The trend is considered statistically insignificant when $-Z_{(1-\alpha/2)} \leq Z \leq Z_{(1+\alpha/2)}$, where $\pm Z_{(1-\alpha/2)}$ are the $1-\alpha/2$ standard normal distribution quantiles.

In case of presence of an increasing trend, the effective value of the trend was estimated using the non-parametric Sen approach (1968). The procedure that was followed is:

- All the possible slope d_k were calculated for each couple of (x_i, x_j), where ($1 \leq i < j \leq n$):

$$d_k = \frac{x_j - x_i}{j - i}. \qquad (5)$$

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- The slope of the linear trend (Sen's slope) was calculated as the median of all the slopes. Similarly can be calculated the intercept. The calculation of the confidence intervals ($\alpha = 5\%$) for the Sen's slope estimate requires at least 10 values in a time series.
- The calculation of the slope was made on annual data and the value of the slope was expressed as a variation of the nitrate concentration per year.
- Using the Sen's slope it is possible to estimate nitrate concentrations in the future, i.e., nitrate concentrations at year N, starting from the last monitoring datum (ISPRA 2017).

Results

The nitrate concentration data in the 16 P-VLTR monitoring points were analysed before performing the spatial nitrate distribution and the MK and Sen tests.

A summary statistics (minimum, maximum and mean values, standard deviation, skewness coefficient) is shown in Table 1. The values show that the data depart from a normal distribution in skewness, which motivates the choice of the non parametric tests for trend analysis.

At groundwater body scale, nitrate distribution in 2017 shows (Fig. 4) the higher values in the southern part of the GWB, where the nitrate content is increased with respect to the distribution in 2004. On the contrary, in the northern part, there are large sectors with unchanged or decreasing nitrate content.

At groundwater well scale, in the period 2003–2017, in P-VLTR the nitrate concentration ranges from 0.1 to 130 mg/L and the average value was 40 mg/L.

The results were compared with the recommended level value of EU's Nitrate Directive (Council Directive 1991), considering in addition the class 37.5 mg/L indicated in the European Union's Groundwater Daughter Directive (Directive 2006/118/EC 2006) as the starting points for trend reversal initiatives. Table 1 shows that in seven wells the nitrate concentration (mean value) is higher than TV (50 mg/L) and in two wells the values are higher than 37.5 mg/L.

Table 1 Summary statistics in 2003-2017 in P-VLTR (nitrate in mg/L)

Sampling points	Number of data point	Minimum value	Maximum value	Mean, μ	Standard deviation	Skewness coefficient
Bvr 2	24	0.3	105	22.6	39.71	1.78
Bvr 6	23	38	115	63.4	23.07	1.11
Bvr 7	23	9.8	103	57.9	20.76	0.44
Bvr 8a	14	11.5	102	64.8	28.17	−0.083
Bvr 12	13	11	30.2	13.8	5.013	3.39
Bvr 14	14	0.1	79	11.8	25.67	2.33
Bvr 16	21	11	130,3	60.9	33.73	1.26
Bvr 23	25	0.1	111	8.60	22.69	4.19
Bvr 24	16	40	73.6	48.2	7.96	2.42
Bvr 25	24	0.1	89	11.4	23.50	2.68
Bvr 26	22	0.8	112	68.6	31.40	−0.37
Bvr 27	23	2.0	79.1	42.1	19.38	−0.21
Bvr 28	22	28.6	97	51.7	19.30	0.92
Bvr 29	20	0.8	106	9.10	23.22	4.22
Bvr 34	24	39.3	121	87.8	20.51	−0.58
Bvr 35	22	12	44	32.0	6.87	−0.88

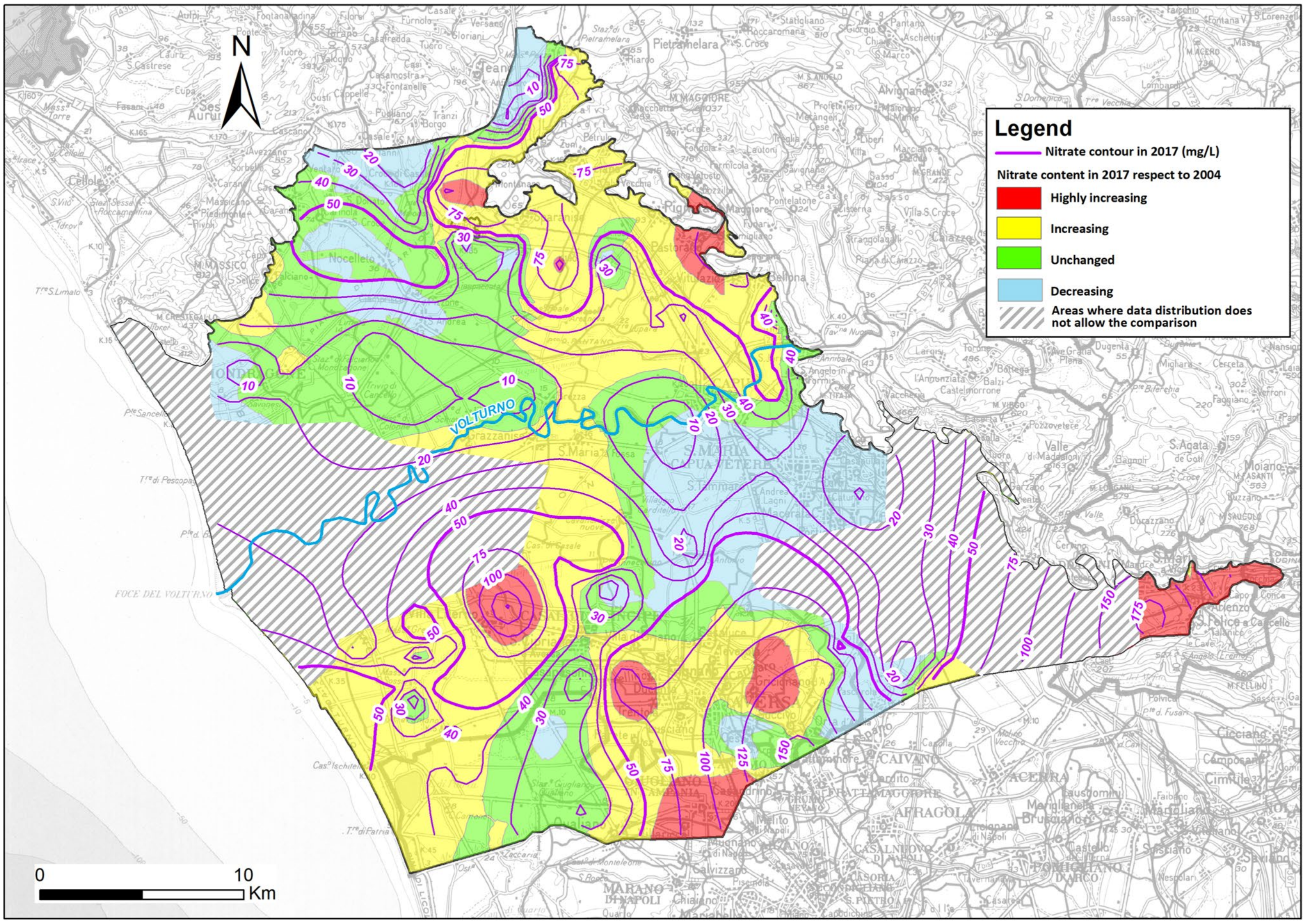

Fig. 4 Nitrate distribution in 2017 in mg/L and differences with the nitrate distribution in 2004

The nitrate concentration time series in the 16 wells were analysed using a MATLAB script file. Four sites showing seasonality (Bvr6, Bvr27, Bvr28 and Bvr35) were analysed using the Two-Season Seasonal Kendall test and the remaining 12 sites were analysed using the Mann–Kendall test.

Table 2 shows the results of the MK trend test for the monitoring wells. The *Z* value of each parameter was calculated and compared with normal distribution critical Z values at the 90% for two-tailed confidence levels.

The results showed that nitrate not changed in the majority of wells (56%). Statistically significant increasing trends were observed at four wells (Bvr7, Bvr26, Bvr28 and Bvr34), corresponding to a percentage of 25%. Three wells had a statistically significant decreasing trend (Bvr2, Bvr12 and Bvr35). An example of a decreasing trend (well Bvr35) is shown in Fig. 5. No reversal trends have been individuated. It is important to note that wells with increasing trends for nitrate concentrations are located in the southern part of the GWB (Fig. 4), where the mean nitrate concentrations are above 50 mg/L (Ducci et al. 2019a).

For the wells Bvr7, Bvr26, Bvr28 and Bvr34 the non-parametric Sen's method for estimating the slope of a linear trend was used using a MATLAB sub-routine. The nitrate concentrations at 2021 and 2027 (extended terms for the achievement of good status—see "Introduction") were calculated using the Sen's slope starting from the last monitoring datum with a 95% confidence intervals. The results are presented in Fig. 6. In the four wells the nitrate concentrations at the future scenarios 2021 and 2027 are higher than 50 mg/L, for the well Bvr26 the nitrate concentrations in 2021 and 2027 are the highest values.

The corresponding area covered by the four monitoring points is 25% of the total area of the GWB. This percentage was estimated as 100/n with $n = 4$ (ISPRA 2017), so the P-VLTR shows an upward trend environmentally significant (ISPRA 2017).

Table 2 Trend test results in P-VLTR using Mann–Kendall test

Sampling points	MK test statistic S	Calculated Z value	Cαritical Z value = 10%	Trend
Bvr 2	−26	−1.97	±1.645	Decreasing
Bvr 6	23	1.09	±1.645	No trend
Bvr 7	84	1.77	±1.645	Increasing
Bvr 8a	8	1.02	±1.645	No trend
Bvr 12	−6	−1.94	±1.645	Decreasing
Bvr 14	−3	−0.34	±1.645	No trend
Bvr 16	8	0.21	±1.645	No trend
Bvr 23	37	0.01	±1.645	No trend
Bvr 24	−7	−0.09	±1.645	No trend
Bvr 25	14	0.61	±1.645	No trend
Bvr 26	35	1.69	±1.645	Increasing
Bvr 27	4	0.77	±1.645	No trend
Bvr 28	27	1.79	±1.645	Increasing
Bvr 29	−17	−1.25	±1.645	No trend
Bvr 34	33	1.75	±1.645	Increasing
Bvr 35	−37	−2.80	±1.645	Decreasing

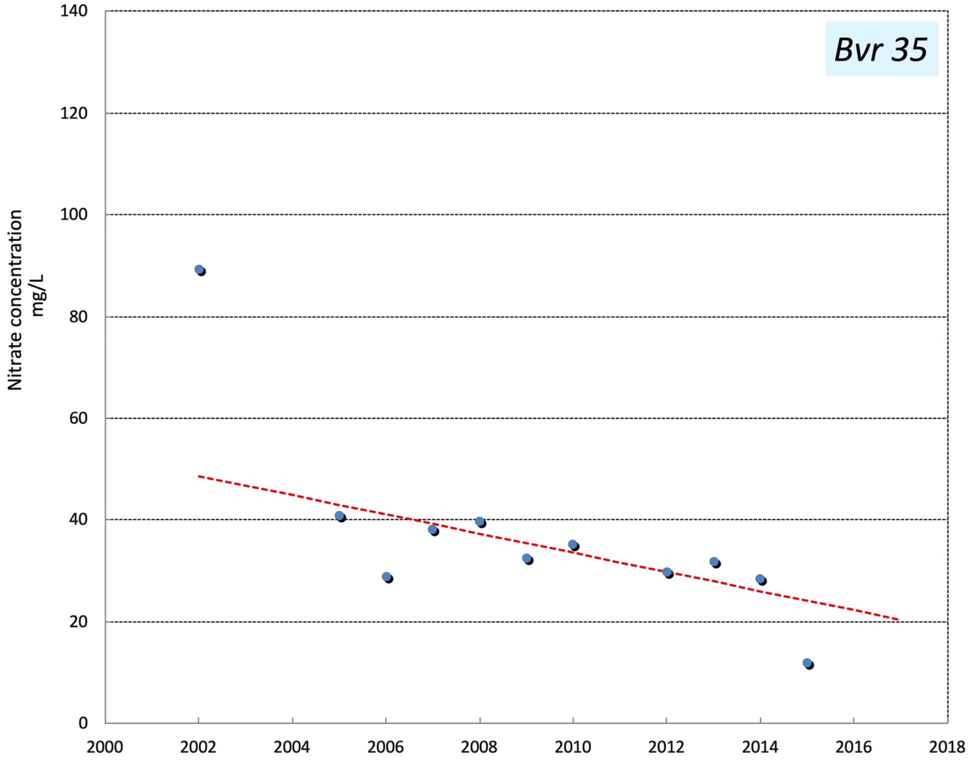

Fig. 5 Example of decreasing trend (well Bvr35)

Conclusions and discussion

To identify the evolution of nitrate concentrations in groundwater, we evaluated the variations at temporal and spatial scale, in an alluvial-pyroclastic groundwater body defined "at risk", for the bad quality status. At temporal scale, we used the data set deriving from the groundwater monitoring network of the Environmental Protection Agency of the Campania region (southern Italy). In the examined GWB, there is nitrate pollution in groundwater. The nitrate trends in each monitoring point were calculated by adopting the guidelines suggested by ISPRA (2017). The results of the trend analysis of nitrate concentration

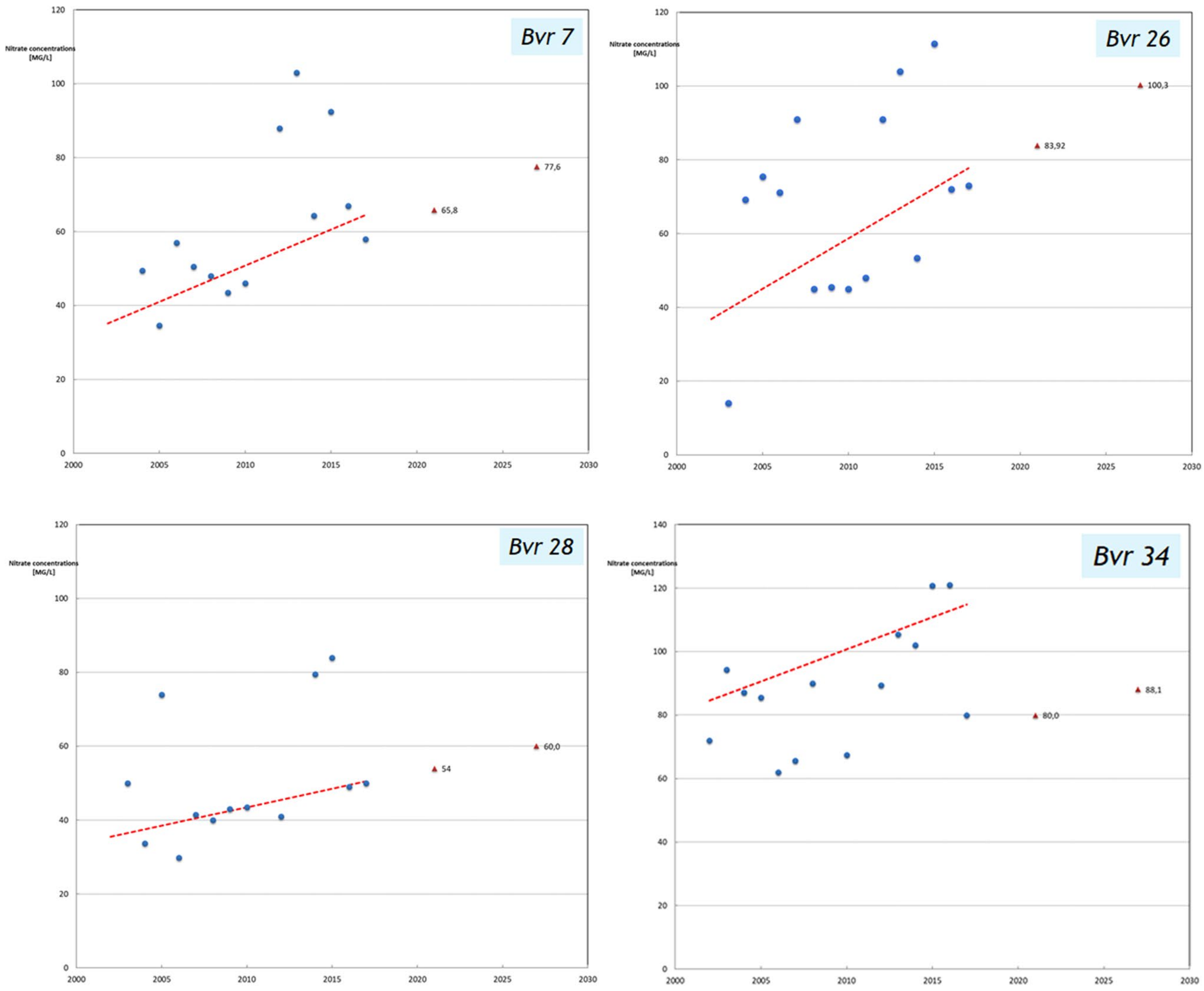

Fig. 6 Linear trends for the wells Bvr7, Bvr26, Bvr28 and Bvr34 estimated using the non-parametric Sen's method

demonstrate that the ISPRA approach is very effective. The southern sector of the study area, affected by high values of nitrate, present upward trends in some points, confirming the results underlined in Ducci et al. (2019a, b). This sector has a high population density and intensive agricultural land-use (arable lands and permanent crops), as described in the Corine Land Cover, 2012.

In detail, four wells show statistically significant increasing trends. Three wells have a significant decreasing trend and 9 wells do not show trend. No reversal trends have been identified and that confirms the absence of strong changes in the land-use in the examined years (2003–2017).

In the four wells showing the increasing trend, the nitrate concentrations in 2021 and in 2027 will be higher than 50 mg/L, if the land-use management does not clearly change toward a sustainable development taking into account an effective N mitigation. Moreover, the wells presenting upward trends seems to be strictly related to the agricultural practices, population density and efficiency of the sewer system. Indeed, this sector is characterised by small agricultural land parcels coexisting with urban settlements.

At groundwater body scale, the groundwater quality is very low, due to the high nitrate content; indeed, in 2017 more than 40% of the wells are above the regulatory limits. These data, compared with the percentage in 2004 (44%) appear in slight decrease (the comparison is not based on the same wells monitoring networks). In 2017, the higher nitrate values are in the southern sector of the GWB, and this is the same sector, where the nitrate content, compared with 2004, is increasing. In the northern sector of the GWB, is prevailing the unchanged or decreasing nitrate content.

In summary, the temporal and spatial analysis, notwithstanding they are based on different approaches and on different data, are in according, highlighting the sector with more anthropogenic pressure, as the sector with higher nitrate content and increasing trend.

Finally, our study highlights that groundwater wells presenting upward trends in nitrate content are probably related to the anthropogenic pressures. Therefore, the study stresses the importance of a consistent groundwater monitoring network at regional scale and the need to plan future measures of land-use management to protect groundwater resources against pollution.

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