

Article Vulnerability and Risk of Contamination of the Varaždin Aquifer System, NW Croatia

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Abstract: This paper presents the first study that assesses the vulnerability and risk of contamination of groundwater in the Varaždin aquifer system. The alluvial aquifer system is mostly unconfined with favorable hydrogeological features. Out of three wellfields, two still operate at full capacity, while the Varaždin wellfield, once the major source of drinking water, has been abandoned due to high concentrations of nitrates in the groundwater. Index-based methods are employed to assess groundwater vulnerability-two DRASTIC-based methods, standard and P-DRASTIC; two SINTACSbased methods, with normal and severe weighting strings; and the GOD method. Hazard is evaluated according to recommendations from the EU COST 620 action, while the risk intensity of the resource is calculated using the results of vulnerability and hazard assessments. The results reveal that for all vulnerability models, the resulting maps have a similar distribution pattern of vulnerability classes, with the high vulnerability class prevailing. However, notwithstanding the generally high groundwater vulnerability, a moderate resource contamination risk prevails as a consequence of a dominantly low hazard index. The validation of the groundwater vulnerability models demonstrates a weak correlation between the vulnerability indices and mean nitrate concentrations in groundwater. Conversely, a significantly higher correlation coefficient (0.58) is obtained when the groundwater vulnerability index is replaced by the resource risk intensity index, indicating that the results of resource risk intensity assessments are superior to groundwater vulnerability results in predicting the level of groundwater contamination.

Keywords: groundwater vulnerability; nitrates; hazard; risk; DRASTIC method; SINTACS method; GOD method; model sensitivity

1. Introduction

Groundwater is an extremely important natural resource, providing the basis of the water supply for the populations of many countries. However, its sustainability is being constantly threatened by increasing pollution, mismanagement, and ever-growing development activities [1–4]. In Croatia, over 90% of drinking water demand is fulfilled with groundwater exploitation. Considering the socioeconomic importance of groundwater, the need for the efficient protection of aquifers from pollution is obvious. This need is stressed in the national legislation governing water management, as well as in two European Union documents: the Water Framework Directive (WFD, 2000/60/EC) and the Directive on the Protection of Groundwater against Pollution and Deterioration (2006/118/EC).

Groundwater vulnerability maps have been extensively used as a tool for dividing a geographic area into subareas that reflect their susceptibility to groundwater contamination (e.g., [5,6]). Despite the long history of the concept, however, a standard definition of aquifer vulnerability still does not exist. In the broadest terms, aquifer vulnerability can be described as the possibility of aquifer pollution as a consequence of activities on the land surface [7]. Two types of aquifer vulnerability are differentiated: intrinsic vulnerability and specific vulnerability [8]. In terms of complexity of groundwater vulnerability models, they



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Copyright: © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). range from simple qualitative, more complex qualitative or semi-quantitative to statistical and fully quantitative models. However, due to simplicity of use, semi-quantitative—index-based methods—prevail. Most studies use a single groundwater vulnerability model (e.g., [9–15]), while those using two or multiple models are less common (e.g., [16–20]). Despite their widespread use, there are only a few groundwater vulnerability studies in Croatia, all of which are focused on karst aquifers, with the application of a single [12,15,21] or several groundwater vulnerability models [22].

There is also no uniform viewpoint on the concepts of the hazard and risk of aquifer pollution in global hydrogeological practice. In this regard, the EU COST Action 620 [23] made a significant contribution with the development of a methodological framework for the assessment of vulnerability, hazard, and risk of aquifer pollution. Although focused on the protection of carbonate aquifers, the concepts established in the EU COST Action 620, such as the "European approach" that was based on the origin–pathway–target model (Figure 1), can be also used in other groundwater environments [24]. The origin, or source of contamination, is the assumed place of release of a contaminant. The pathway is the flow path of a potential contaminant from its point of release (origin) through the system to the point that has to be protected (target). For resource protection, the pathway consists of a mostly vertical passage within a protective cover, while for source protection, it also includes horizontal flow in the aquifer. For resource protection, the target is the groundwater surface in the aquifer under consideration, and for source protection, it is the water in the well or spring [25].



Figure 1. Origin-pathway-target model [26].

Several studies have recently stressed the importance of assessing risk to groundwater contamination for formulating efficient measures for risk reduction [27–29]. However, in contrast to the frequent application of groundwater vulnerability models, the studies of the risk of groundwater contamination are less common. Particularly in Croatia, there is no such study that evaluates the risk of aquifer contamination resulting from a combination of aquifer vulnerability and hazard.

The Varaždin aquifer system represents the main source of drinking, industrial, and irrigation water for the town of Varaždin and its surroundings. Over the years, inadequate land use practices have caused deterioration of groundwater quality, with high concentrations of nitrates being a major concern [30,31]. As a consequence, the aquifer system is classified as a groundwater body of poor chemical status under the Water Framework Directive (WFD). Considering the regional significance of the Varažding aquifer system, it is of

utmost importance to attain a good qualitative status of the aquifer system as a prerequisite for sustainable management of groundwater resources. Therefore, the establishment of appropriate pollution control strategies and the application of adequate measures aimed at improving the quality of groundwater are necessary. To this end, reliable information on the susceptibility of the aquifer system to pollution and on the risk status is crucial. Therefore, the objective of this study is to assess the vulnerability of groundwater in the Varaždin aquifer system by employing several vulnerability models of varying complexity and the risk of groundwater contamination using the results of the groundwater vulnerability and hazard assessments. To achieve these objectives, (1) groundwater vulnerability assessment is performed using five models (Section 2.3.1), (2) the independency of model parameters and single parameter sensitivity are analyzed (Section 2.3.2), (3) hazard assessment is carried out (Section 2.3.3), (4) resource risk intensity evaluation is performed using the outputs from groundwater vulnerability and hazard assessments (Section 2.3.4), (5) validation of groundwater vulnerability and resource risk intensity models is carried out (Section 2.3.5).

2. Materials and Methods

2.1. Study Area

The study area is located in the north-eastern part of the Republic of Croatia along the state borders with Slovenia and Hungary (Figure 2). It covers 264 km² and encompasses the western part of the Drava river valley in Croatia. The valley extends in a northwest-southeast direction and is filled with deposits of Neogene and Quaternary age. The altitude varies between 156 m a.s.l. and 243 m a.s.l.



Figure 2. Location map, equipotential lines, and groundwater flow directions.

According to Köppen's classification, the study area belongs to the Cfb climate type [32], with a moderately warm, humid climate and warm summers. The mean annual air temperatures for the periods 1960–1991 and 1971–2000 were 9.9 °C and 10.2 °C, [33]. The average annual rainfalls for the same periods were 879.2 and 843.1 mm, respectively.

The Varaždin aquifer system was formed during the Pleistocene and Holocene as a result of accumulation processes of the Drava River [34]. It is composed of gravel and sand, with variable portions of silt [35–37]. The aquifer thickness varies from 5 to over 100 m (Figure 3). In the central part, near the town of Varaždin, a tiny aquitard divides the aquifer into two hydrogeological units, the first and the second aquifer. The aquitard has regional significance and can be tracked even downstream of the investigated area. It is composed of clay and silt, with thickness rarely exceeding 5 m. A covering layer is not continuously developed; in the central part and near the Drava River, it rarely exceeds two meters and very often completely disappears. Impermeable clays and marls underlie the aquifer. Hydraulic conductivity values of the aquifer generally range from 100 to 300 m/day in the central part, although these values are significantly lower in the southern and western boundary belt.



Figure 3. Lithological cross section [31].

The direction of groundwater flow is generally NW-SE, parallel to the Drava River (Figure 2). The aquifer recharges through the infiltration of precipitation and the seepage of water from reservoirs and the Drava River.

There is a large number of potential and actual groundwater polluters across the study area. The region is fairly developed, with agriculture and poultry farming among its most important industries. Industrial activity is relatively less important and is almost completely situated in the town of Varaždin, which is located in the centre of the region. The natural quality of groundwater had previously complied with Croatian drinking water standards; however, over time, inadequate land use management took its toll, and high nitrate concentrations in the groundwater were first noticed in the 1970s [30]. The sources of pollution are intensive agricultural production, an abundance of poultry farms, and a lack of sewerage network in the settlements surrounding the town of Varaždin. Out of three wellfields in the region, two still operate at full capacity—Bartolovec and Vinokovšćak. The Varaždin wellfield, once the major source of drinking water, has been abandoned due to high concentrations of nitrates in the groundwater. Recent study identified the poultry dumps as the main source of nitrate pollution in the catchment area of the wellfield [38]. However, a negative trend of nitrate concentrations in the shallow aquifer system has recently been observed. It is expected that this trend will continue under the assumption of maintaining the current dynamics of nitrate leaching from the surface into groundwater, as demonstrated by numerical modelling of nitrate transport [39]. Furthermore, another study showed that the future evolution of nitrate concentration in groundwater in the shallow aquifer system is strongly affected by land use practices, i.e., the amounts of nitrates leaching into groundwater [40].

2.2. Data Sources

The establishment of vulnerability and risk models relies on a wide range of information that is obtained from different sources. Lithological information is obtained from borehole records stored in the database of Croatian Geological Survey and reports from geotechnical and civil engineering companies (e.g., Geofizika, Geotehnika). In total, 136 borehole logs are used for interpretation of the aquifer media and impact of vadose zone vulnerability parameters. The monitoring of groundwater levels and quality has been carried out by Croatian Meteorological and Hydrological Service (DHMZ). Time series of groundwater levels from 93 observation wells for the period from 1995 to 2004 are used for the development of the groundwater levels equipotential map for average water levels. Furthermore, time series of nitrate concentrations in groundwater from eight observation wells are used to calculate mean nitrate concentrations for the period from 2007 to 2017. Digital orthophoto layouts and a digital elevation model (DEM) are acquired from the State Geodetic Administration. Information on soil media properties is obtained from the Hydropedological Map of the Republic of Croatia [41], while the net recharge and hydraulic conductivity of the aquifer are obtained from the calibrated groundwater flow model [31]. Land use information for hazard assessment is obtained from digital orthophoto layouts, topographic maps, and field surveys.

2.3. Methods

Groundwater vulnerability is assessed with 5 models—2 DRASTIC-based models, 2 SINTACS-based models, and a GOD model. Furthermore, the sensitivity of vulnerability models and the independence of the DRASTIC and SINTACS model parameters are evaluated. Finally, hazard and risk intensity of the resource are calculated, followed by the validation of the results of groundwater vulnerability and risk intensity assessments using linear correlation analysis.

Spatial analysis using thematic rasters is performed with the Spatial Analyst tools in the ArcGIS 10.2.1 modelling environment. All rasters are created using the appropriate ArcGIS tools depending on the geometry of the input data. For example, features with polygonal geometry are converted to rasters using the Polygon to Raster tool, while for line features, e.g., hazard types with linear geometry, lines are firstly transformed to polygons using the Buffer tool and the range of the impact of the feature (Table S1) as the input parameter, and secondly, polygons are converted to rasters using the Polygon to Raster tool. Raster overlay operations are performed using the Raster Calculator tool. All rasters have a resolution of 50 m.

2.3.1. Groundwater Vulnerability Methods

DRASTIC is one of the most widely used methods for groundwater vulnerability assessment. It was developed by the U.S. Environmental Protection Agency (EPA) to assess groundwater pollution potential for the entire United States [42]. Seven parameters are used to calculate vulnerability index (Equation (1)). Each parameter involved in calculation is assigned a rating between 1 and 10 based on their respective impact on the groundwater vulnerability.

$$DRASTIC Index = D_r \cdot D_w + R_r \cdot R_w + A_r \cdot A_w + S_r \cdot S_w + T_r \cdot T_w + I_r \cdot I_w + C_r \cdot C_w$$
(1)

where D is the depth to groundwater, R is the net aquifer recharge, A is aquifer media, S is soil media, T is topographic slope, I is vadose zone, and C is hydraulic conductivity, and r_i and w_i stand for ratings and weights, respectively.

Pesticide DRASTIC (P-DRASTIC) is a special type of DRASTIC method developed for assessment of specific groundwater vulnerability in regions with intensive agricultural activity. The only difference between the methods is in the weights assigned to the seven model parameters (Table 1). As a consequence, the maximum theoretical vulnerability index differs between the methods—226 and 256 for DRASTIC and P-DRASTIC, respectively.

Parameter	DRASTIC	P-DRASTIC	SINTACS-N	SINTACS-S
Depth to groundwater	5	5	5	5
Net recharge	4	4	4	5
Aquifer media	3	3	3	3
Soil media	2	5	4	5
Topography	1	3	2	2
Vadose zone	5	4	5	4
Hydraulic conductivity	3	3	3	2

Table 1. Weights for DRATIC-based and SINTACS-based models.

There is no single classification system for the vulnerability indices calculated using DRASTIC-based models. In this study, division into six vulnerability classes is used. It is performed according to [43] for the DRASTIC model, while the approach similar to [44] is applied for the P-DRASTIC model.

The SINTACS vulnerability model is based on the DRASTIC model and, therefore, uses the same 7 model parameters and the same Equation (1) to calculate the vulnerability index. The groundwater vulnerability indices are grouped into 6 classes: very low ($26 < I_v < 80$), low (80 < I_v < 105), moderate (105 < I_v < 140), elevated (140 < I_v < 186), high (186 < I_v < 210), and very high $(210 < I_v < 260)$. The SINTACS model was developed in Italy in an attempt to provide greater flexibility and correct the shortcomings of DRASTIC regarding the range of values of individual parameters and the distribution of vulnerability indices into appropriate categories [45]. There are different groups of weighting factors that are applied in an effort to gain more realistic insight into the impact of each individual parameter on aquifer vulnerability. In this way, the model may be adapted to different hydrogeological conditions and land uses. Five groups of weighting factors were anticipated, although the possibility of creating new ones was left open if justified by the conditions; however, the sum of these factors must equal 26. In the current study, 2 groups of weighting factors are used—normal impact areas (SINTACS-N) and severe impact areas (SINTACS-S) (Table 1). The former is suitable for application in lowland areas with aquifers made of unconsolidated rocks, with a relatively shallow groundwater and thick soil cover. Areas are uncultivated, or, if cultivated, use minimal fertilizers and pesticides and are unirrigated. Areas are mostly unpopulated or contain smaller settlements. The latter corresponds

to the same hydrogeological conditions as those in the normal impact area, but land is cultivated with intensive use of agrochemicals and breeding, and urban settlements and industrialization are also present.

The GOD model [46] is suitable for a relatively quick assessment of groundwater vulnerability due to its simple structure. It was developed for application in regions with a relatively small amount of available data on the natural system. The following key parameters are used: (a) the groundwater occurrence (G), (b) the overlying lithology of the aquifer (O), and (c) the depth to the groundwater (D). The resulting vulnerability index represents the product of the values of the individual parameters (Equation (2)). In total, there are 5 vulnerability classes: negligible ($I_v < 0.1$), low ($0.1 < I_v < 0.3$), medium ($0.3 < I_v < 0.5$), high ($0.5 < I_v < 0.7$), and extreme ($0.7 < I_v < 1$).

$$I_v = G \cdot O \cdot D \tag{2}$$

2.3.2. Sensitivity Analysis

Inclusion of a large number of model parameters is considered as one of the major advantages of the DRASTIC and SINTACS vulnerability models [47] due to the belief that they limit the effects of errors and uncertainties of the individual model parameters on the final result [48]. However, a larger number of model parameters increases the likelihood that they are correlated. Therefore, the independency of DRASTIC and SINTACS model parameters is examined using rank-order correlation analysis. Unlike the Pearson correlation analysis which assumes a normal distribution of the variables, the Spearman rank-order correlation coefficient (Equation (3)) is a nonparametric measure of the strength and direction of association that exists between two variables [49]—in this study, between individual model parameters.

$$\rho = \frac{6\sum_{i=1}^{N} d_i^2}{N(N^2 - 1)} \cdot 100 \tag{3}$$

where *N* is sample size and *d* is the rank difference. The greater the absolute value of the correlation coefficient ρ is, the stronger the correlation is.

Furthermore, summary statistics are used to evaluate the contribution of individual model parameters to the resulting vulnerability index. Finally, a single parameter sensitivity measure is employed (Equation (4)) to compare the effective weight of each model parameter with the theoretical weight from the corresponding groundwater vulnerability model.

$$W = \frac{P_r \cdot P_w}{V} \cdot 100 \tag{4}$$

where W stands for the effective weight of each model parameter, P_r and P_w are the rating and weight of each model parameter, respectively, and V is the groundwater vulnerability index.

2.3.3. Hazard Assessment

There are different hazard assessment methods that generate either absolute or relative results. Due to the complexity of the investigation and the difficulty in quantifying the hazard arising from the number of processes to be considered and the fact that the features of these processes change over time [50], an absolute hazard assessment is difficult to achieve. Therefore, relative hazard assessment provides a simpler approach based on pre-defined criteria. According to the concepts proposed in COST Action 620 [51], a relative hazard assessment involves 4 steps: (1) hazard identification, categorization and the establishment of the weighting factor system that reflects the toxicity of the substance, and characteristics that influence its solubility and mobility (Table S2), (2) ranking of hazards that belong to the same category, (3) assessment of the probability of contaminant discharge into environment, and (4) definition of a mathematical algorithm for the calculation of the

potential harm level for each individual hazard, taking into account the weighting factors, ranking, and hazard probability.

The ranking of hazards that belong to the same category is conducted by considering the quantity of the potentially harmful substances that could be discharged into the environment. However, this ranking should lead neither to a drastic minimization nor to an excess overvaluation of substances within the same hazard category. To maintain a fair balance with the average weighting values, these values are adjusted only slightly by multiplying them with a ranking factor (Q_n) between 0.8 and 1.2, to indicate low or high amounts, respectively, of a toxic substance in comparison with the general average.

The probability of contaminant discharge is expressed by the reduction factor (R_f) , whose value ranges from 0 to 1. Estimating this value requires an analysis of a number of processes and conditions related to the area in which potentially harmful substances are produced, stored, or applied, e.g., technical state, maintenance level, safety measures, etc. Such analysis requires caution because there is a possibility of minimizing the total hazard from very toxic substances due to the overestimation of safety procedures for their handling and storage. If relevant data are unknown, the reduction factor is given a value of 1.

Finally, the hazard index is calculated and classified according to Equation (5) and Table 2, respectively.

$$\mathbf{H} = \mathbf{H} \cdot \mathbf{Q}_{\mathbf{n}} \cdot \mathbf{R}_{\mathbf{f}} \tag{5}$$

where HI is the hazard index, H is the weighting factor, Q_n is the ranking factor, and R_f is the reduction factor.

Hazard Index	Hazard Index Class	Hazard Level	Colour	
 0–20	1	no or very low	dark green	
>20-40	2	low	green	
>40-60	3	moderate	light green	
>60-80	4	elevated	yellow	
>80-100	5	high	orange	
>100-120	6	very high	red	

Table 2. Hazard index and hazard index classes (modified according to [51]).

F

The raster representing hazard index is generated in such a way that each raster cell has a value corresponding to the HI value of the hazard(s) at that location. First, one raster is created for hazard types of polygonal geometry and the other for hazard types of linear geometry. After that, a hazard index raster is created by adding both rasters using the Raster Calculator tool.

2.3.4. Risk Assessment

According to the origin–pathway–target model (Figure 1), a risk assessment is performed separately for the resource and for the source [52]. In the current study, the risk is evaluated only for the resource using Equation (6) and the classification of the risk intensity index is performed according to Table 3.

$$RII = I_v \cdot HI \tag{6}$$

where RII is the risk intensity index, I_v is the vulnerability index and HI is the hazard index.

Out of the five groundwater vulnerability models used in the study area, the one with an approximately average correlation coefficient (R) between groundwater vulnerability index and nitrate concentration in groundwater is selected to perform the risk assessment of the resource.

Vulnerability Index	Hazard Index	Risk Intensity Index	Risk Level	Colour
26-80	0–20	0-1600	no or very low	dark green
>80-105	>20-40	>1600-4200	low	green
>105-140	>40-60	>4200-8400	moderate	light green
>140-186	>60-80	>8400-14,880	elevated	yellow
>186-210	>80-100	>14,880-21,000	high	orange
>210-260	>100-120	>21,000-31,200	very high	red

Table 3. Classification of the risk intensity of the resource.

2.3.5. Validation of Groundwater Vulnerability and Risk Assessment

Validation of the results of groundwater vulnerability and risk intensity assessment are performed using the concentrations of nitrates in groundwater as an indicator of anthropogenic impact on groundwater. To this end, the time series of nitrate concentrations in groundwater for the period 2007–2017 from 8 observation wells included in the national monitoring network are used to calculate the mean nitrate concentrations per each well (Table S3). Linear correlation analysis is performed to measure and interpret the strength of a linear relationship between the variables—groundwater vulnerability or risk intensity index in the raster cells corresponding to the locations of observation wells and nitrate concentration. The Pearson correlation coefficient (R) is used to interpret the strength of correlation, while the sign of the correlation coefficient, positive or negative, defines the direction of the relationship.

3. Results and Discussion

3.1. Vulnerability Assessment

Examples of rated rasters in the case of the depth to groundwater parameter are shown in Figure 4. Depths to groundwater in the central part of the study area vary from <0.5 m to 12 m. Greater depths are found within the depression cone of the pumping site Vinokovšćak and in the impact area of the HPP Varaždin derivative channel, which has a strong drainage impact (Figure 2). The impact areas of the reservoirs Varaždin and Čakovec, however, are characterized by smaller depths to groundwater. The rating scores range from 1 to 10 and from 2.75 to 10 for DRASTIC and SINTACS, respectively, with the DRASTIC model having generally higher scores compared to the SINTACS model. As for the GOD model, the rating scores are in the range from 0.6 to 1.

According to the results of groundwater flow modelling, the effective infiltration of precipitation is 270 mm/year in the largest part of the study area, while only in areas where the thickness of a silty–clayey covering layer is above 2.5 m, the effective infiltration is lower—155 mm/year [31]. Consequently, the parameter rating scores are 6 and 9 (Figure S1a), and 6.6 and 9.3 (Figure S2a) for the DRASTIC and SINTACS models, respectively, with higher scores corresponding to higher effective infiltration rates.

The aquifer characteristics layer is uniform as a consequence of the aquifer composition dominated by gravel and sand particles in different mutual ratios, with only subordinate interlayers of fine-grained sediments. Consequently, the score is eight for both DRASTIC and SINTACS (Figures S1b and S2b).

In the soil layer, the rating is performed using the predominant lithological composition of individual hydropedological units. The values range from 6 to 8 (Figure S1c) and from 4.5 to 6.5 (Figure S2c) for DRASTIC and SINTACS, respectively. Higher values correspond to the higher content of coarse-grained particles in the lithological composition. Thus, the highest score corresponds to the automorphic, well-drained soil located in the southern boundary part of the aquifer, while the lowest score corresponds to the very shallow and moderately shallow hypogley soil types found in the vicinity of the pumping sites Bartolovec and Vinokovšćak. 2.5

10[°]km



2.5



(a)

Figure 4. The rating scores for depth to groundwater layers used to calculate vulnerability indices: (a) DRASTIC, (b) SINTACS, and (c) GOD.

10 km

The study area is characterized by a flat relief of the Drava river valley; consequently, the predominant value for topography layer is 10 (Figures S1d and S2d). Only in the western and southern boundary parts of the aquifer do the slopes gradually increase, leading to lower DRASTIC and SINTAC rating scores.

The rating scores for the impact of the vadose zone range from 3 to 10 for both DRASTIC and SINTACS (Figures S1e and S2e). The lower values are assigned to the southern boundary area of the aquifer, the vicinity of the pumping sites Vinokovšćak and Bartolovec, and several smaller areas in the central parts of the aquifer. A common feature among these areas is the larger thickness of the unsaturated zone, dominated by fine-grained, clayey–silty particles.

Calibrated values of the hydraulic conductivity of the aquifer using the numerical model of groundwater flow range from 10 to 330 m/day [31]. The lower values are found along the western and southern aquifer boundaries. In the central part of the aquifer, they gradually decrease from 330 m/day in the north-westernmost part to 250 m/day downstream of the pumping site Bartolovec. Accordingly, the rating scores range from 2 to 10 (Figure S1f) and from 7 to 9.4 (Figure S2f) for DRASTIC and SINTACS, respectively, with higher scores corresponding to higher aquifer conductivity.

Regarding the GOD model, apart from the depth to groundwater parameter described above, there are two additional parameters used for groundwater vulnerability assessment—G and O. Unconfined aquifer spreads over the most of the study area, followed by unconfined–covered and semi-confined aquifers and, accordingly, the G parameter scores are 1, 0.5, and 0.3, respectively (Figure S3a). As for the O parameter, the rating scores are 0.5 and 0.7 (Figure S3b), with a lower score corresponding to a greater thickness and higher fraction of clay and silt in the lithological composition of the vadose zone.

For all methods used in the current study, the vulnerability maps generally have a similar distribution pattern of vulnerability classes (Figure 5), with the high vulnerability

(b)

class dominating the central part of the considered area. Furthermore, it is found that for the DRASTIC- and SINTACS-based models, only small areas belong to a very high vulnerability class, and they are spread mostly along the Drava river banks and in the eastern part of the study area, while in the vulnerability map created using the GOD model, there are no such areas with very high vulnerability. The resulting maps also clearly demonstrate that there are, overall, only small differences between the vulnerability maps created using SINTACS-based and DRASTIC-based models. The biggest difference is observed in the southern marginal area where there is a belt of low and moderate vulnerability for the DRASTIC model, while in the same area, DRASTIC-P- and SINTACS-based models produce an elevated vulnerability. As for the GOD model, the results indicate a lower vulnerability compared to other models, not only over the isolated areas in the central and western areas, but also along the southern marginal belt and at the utmost eastern part of the study area. These are areas with a greater thickness of the covering layer of the aquifer, which also contains a higher fraction of silt and clay. Thus, the rating scores are relatively low—0.3 and 0.5 for G, and 0.5 for O—which together with D rating scores leads to low vulnerability in these areas.

From Figure 6, it is seen that more than 55% of the study area belongs to the high vulnerability class in all vulnerability maps. The distribution of other classes is similar for DRASTIC-based and SINTACS-based models. However, it is interesting that the very high vulnerability class for both P-DRASTIC and SINTACS-S covers a smaller fraction of the total area compared to DRASTIC and SINTACS-N. This is due to the fact that, compared to DRASTIC and SINTACS-N, the smaller DRASTIC-P and SINTACS-S weighting factors are applied to the layers with overall higher scores (e.g., hydraulic conductivity) in the central area and along the Sava river banks. In addition, the higher DRASTIC-P and SINTACS-S weighting factors are employed for the layers with generally lower scores (e.g., soil media). Furthermore, it is also evident that a very high vulnerability class covers a greater area in SINTACS-based vulnerability maps than in the DRASTIC counterparts. Finally, larger areas (37%) with low vulnerability are only found in the map created using the GOD model.

3.2. Sensitivity of the Vulnerability Models

Of the seven parameters used for aquifer vulnerability assessment in both DRASTICbased and SINTACS-based models, the topography parameter, due to the flat relief, contributes the most to the resulting vulnerability index (mean value 9.9), followed by hydraulic conductivity and net recharge (Table 4). The least risk of contamination originates from the vadose zone and soil media parameters for DRASTIC-based and SINTACS-based models, respectively. Furthermore, all model parameters exhibit relatively low variability, with the vadose zone being the most variable parameter for both models.

Regarding the GOD model, the highest risk of aquifer contamination arises from the depth to groundwater parameter, followed by the groundwater occurrence, and finally, overlying lithology (Table 4). Of those three parameters, groundwater occurrence is the most variable one.

The results of the rank-order correlation analysis show that there are only a few significant correlations at 95% confidence level (Table 5), indicating the prevailing independence of the DRASTIC and SINTACS parameters, which is consistent with results from other studies [13,14,53]. The independency of vulnerability model parameters decreases the probability of misjudgment [48]. A relatively strong correlation (r > 0.5) for both DRASTIC and SINTACS models is achieved between the impact of the vadose zone and net recharge, and net recharge and hydraulic conductivity. The former correlation is due to the fact that the same geological dataset is used to rate the impact of the vadose zone and to delineate groundwater recharge zones and then rate the net recharge parameter. The latter correlation can be attributed to the fact that the areas with higher hydraulic conductivity values mostly coincide with higher net recharge and vice versa. Furthermore, strong and moderate correlations are achieved for the impact of the vadose zone and hydraulic conductivity parameters for the DRASTIC and SINTACS models, respectively, due to similar lithological

composition of the vadose and saturated zones of the aquifer. The remaining parameter combinations have either moderate or low correlations.

Figure 5. Aquifer vulnerability maps: (a) DRASTIC, (b) P-DRASTIC, (c) SINTACS-N, (d) SINTACS-S, (e) GOD.

The comparison between effective and theoretical parameter weights across the DRASTIC- and SINTACS-based vulnerability models is performed using single-parameter sensitivity analysis. Unlike some other studies where larger deviations are obtained [13,14], the results in the current study (Table 6) reveal that there is relatively good agreement between theoretical and effective weights. The largest difference is achieved for the soil media parameter, for which theoretical weight exceeds the effective weight by 5.4% in the SINTACS-S model. Furthermore, the depth to groundwater parameter is the most effective parameter, followed by the net recharge parameter for all methods except SINTACS-S, where the net recharge tops the list of the most effective parameters. Considering their significance, it is important to collect accurate information about the most effective parameters. In the current study, this is achieved for the net recharge parameter by extracting it from a calibrated groundwater flow model and for the depth to groundwater parameter, with geostatistical interpolation of groundwater depth measurements from 93 observation wells

(Figure 2). On the other side of the spectrum, the least effective parameters are topography and hydraulic conductivity for DRASTIC, SINTACS-N, and P-DRASTIC, SINTACS-S models, respectively.

Figure 6. Vulnerability classes for SINTACS-based, DRASTIC-based, and GOD models.

	Depth to Groundwater	Net Recharge	Aquifer Media	Soil Media	Topography	Vadose Zone	Hydraulic Conductivity		
DRASTIC									
Min	2.0	6.0	8.0	6.0	1.0	3.0	2.0		
Max	10.0	9.0	8.0	8.0	10.0	10.0	10.0		
Mean	8.2	8.3	8.0	6.7	9.9	6.6	9.0		
SD	1.3	1.3	0.0	0.6	0.4	2.1	2.6		
CV (%)	15.7	15.6	0.0	8.7	4.3	31.6	28.8		
	SINTACS								
Min	2.8	6.6	8.0	4.5	1.0	3.0	7.0		
Max	10.0	9.3	8.0	6.5	10.0	10.0	9.4		
Mean	7.4	8.6	8.0	5.3	9.9	6.6	9.0		
SD	1.2	1.2	0.0	0.5	0.4	2.1	0.8		
CV (%)	16.4	13.4	0.0	9.2	4.3	31.5	8.5		
			C	GOD					
	Groundwater	occurrence	Overlying	lithology	Γ	Dept to groundwa	iter		
Min	0.3		0.5		0.6				
Max	1.0		0.7		1.0				
Mean	0.8		0.6		0.9				
SD	0.3	0.3		0.1		0.1			
CV (%)	40.1	1	14	.8	8.7				

Table 4. Summary statistics of the vulnerability model parameters.

Correlated Parameters	Correlation Coefficient, r	Significance Level, p			
DRASTIC					
Impact of vadose zone and net recharge	0.67	< 0.0001			
Net recharge and hydraulic conductivity	0.62	< 0.0001			
Impact of vadose zone and hydraulic conductivity	0.52	< 0.0001			
Topography and hydraulic conductivity	0.36	< 0.0001			
Net recharge and topography	0.27	< 0.0001			
Topography and depth to water	0.24	< 0.0001			
SINTACS					
Impact of vadose zone and net recharge	0.65	< 0.0001			
Net recharge and hydraulic conductivity	0.50	< 0.0001			
Impact of vadose zone and hydraulic conductivity	0.49	< 0.0001			
Soil media and depth to water	0.35	< 0.0001			
Net recharge and topography	0.27	< 0.0001			
Topography and depth to water	0.23	< 0.0001			

Table 5. Summary of rank-order correlation analysis results between seven DRASTIC and SIN-TACS parameters.

Table 6. Theoretical and effective parameter weights.

	DRASTIC		P-DRASTIC		SINTACS-N		SINTACS-S	
Parameter	Theoretical Weight (%)	Mean Effective Weight (%)						
Depth to groundwater	21.7	22.9	19.2	20.1	19.2	18.9	19.2	19.0
Net recharge	17.4	18.2	15.4	16.0	15.4	17.6	19.2	22.1
Aquifer media	13.0	13.5	11.5	11.8	11.5	12.3	11.5	12.4
Soil media	8.7	7.5	19.2	16.4	15.4	11.0	19.2	13.8
Topography	4.3	5.6	11.5	14.6	7.7	10.1	7.7	10.2
Vadose zone	21.7	17.8	15.4	12.6	19.2	16.4	15.4	13.3
Hydraulic conductivity	13.0	14.5	7.7	8.6	11.5	13.7	7.7	9.2

3.3. Hazard Assessment

The different types of land use are grouped, according to their size, into the hazard types with polygonal and linear geometry. In the group of the polygonal hazard type, seven potential pollution sources are identified (Table S4), among which intensive agricultural areas with high demand of fertilizers and pesticides cover the largest area (200.5 km²), followed by forest (17.7 km²) and detached houses without a sewer system (20.8 km²).

The town of Varaždin is assigned the highest hazard index value (75), followed by sanitary landfill (50), detached houses without a sewer system (45), and poultry farms (45). The hazard index assigned to the town of Varaždin is higher than what is specified for urbanization (Tables S2 and S4) and the reason for that is a presence of different industrial entities in the town that, due to the scale of investigation, cannot be isolated and assessed individually. Therefore, the town as a whole has a higher hazard index that approximately corresponds to the interaction of different hazards.

The ranking factor 1.2 is assigned to only two polygonal hazard types—agricultural areas, due to intensive use of fertilizers and manure on arable land, and poultry farms, as a consequence of large amounts of poultry excreta disposed on-site (Table S4).

Assessment of all the factors that affect the potential reduction of the probability of releasing harmful substances into the environment is a complex task and beyond the scope of the current study. Therefore, all hazard types are assigned a reduction factor of one (Table S4).

In total, three hazard types are identified in the hazard group with linear geometry (Table S1), with a hazard index ranging from 40 for highways and main roads, to 30 for railway lines. The range of impact is the largest for highways (120), followed by main roads and railway lines (80).

The hazard index ranges from 0 to 106. Low and very low hazard levels dominate the areas with forests and fields (Figure 7). Settlements without sewerage systems and the town of Varaždin are in the moderate and elevated hazard category, respectively. The smallest areas are covered by high and very high hazard categories and include places with combined impacts from different types of land use, e.g., main roads, poultry farms, agricultural surfaces, etc.

Figure 7. Hazard map.

3.4. Risk Intensity Assessment

The risk intensity map (Figure 8) is created according to Equation (6) and using the SINTACS-S vulnerability index. It clearly reveals that the moderate risk class prevails, occupying 75% of the study area. It is followed by very low, elevated, and high risk classes, with 11%, 10%, and 5% of the study area, respectively. The smallest fraction belongs to low and very high vulnerability classes—0.1%.

The areas with moderate risk are predominantly characterized by elevated to very high aquifer vulnerability and low hazard, originating mostly from agricultural activities that involve the intensive use of manure and fertilizers. Forest areas are in the very low risk class regardless of the aquifer vulnerability level due to negligible hazard. The low risk class covers small areas with meadows along the western and southern boundary parts of the aquifer where aquifer vulnerability is elevated. The elevated risk class covers the areas with poultry farms and settlements in the central part of the study area, as well as a section of the town of Varaždin. The other section of the town of Varaždin and the areas that integrate the impacts of the main roads and agriculture belong to the high risk

class. Identical hazard types belonging to different risk categories are the result of different aquifer vulnerability levels.

Figure 8. Risk intensity map.

The lack of a standardized methodology for assessing the risk of aquifer contamination makes it difficult to compare the results of studies performed in different regions. In this regard, it is worth pointing out that the studies that followed the COST 620 guidelines have generally comparable results. Thus, in similar studies of the risk of contamination of alluvial aquifers, both Boulabeiz et al. [54] and Gonzales Herrera et al. [55] reported low to moderate risk in agricultural areas with moderate and high aquifer vulnerability, the former using the GOD model and the latter using a modified DRASTIC model for vulnerability assessment. These results are generally consistent with the findings of the current study. In contrast, Campoverde-Munoz et al. [20], using DRASTIC and GOD aquifer vulnerability models together with the POSH method for the evaluation of contamination danger, and Ribeiro et al. [56] using the SI method, found that agricultural areas are at high risk of aquifer contamination. The differences in the resulting risk categories for the similar combinations of hazard and vulnerability classes in these studies are mainly due to different methods applied for hazard assessment.

3.5. Validation of Groundwater Vulnerability and Risk Intensity Maps

For the groundwater vulnerability index, correlation coefficients range from 0.22 to 0.38 (Table 7), indicating a weak correlation between the variables. The lowest one is obtained for the SINTACS-N model. Overall, the differences in results among different groundwater vulnerability models are relatively small. However, it is interesting that the second highest correlation coefficient is obtained for the GOD method (0.34), which includes the fewest model parameters. This aligns with the conclusions from other studies, e.g., [57,58], suggesting that similar or even better results can be achieved using models

with a lower number of model parameters. Furthermore, the benefit of combining the results of hazard and vulnerability assessments in the scope of the resource risk intensity assessment is clearly indicated by a significantly higher correlation coefficient (0.58), as compared to those obtained for the results of vulnerability assessments, which corresponds to a moderate correlation between variables.

Table 7. Correlation coefficient, R between groundwater vulnerability and risk intensity indices and mean nitrate concentration in groundwater.

Groundwater Vulnerability Model/Risk Intensity	DRASTIC	P-DRASTIC	SINTACS-N	SINTACS-S	GOD	Risk Intensity
Correlation coefficient, R	0.33	0.38	0.22	0.32	0.34	0.58

Furthermore, as a limitation of the study, the time series of nitrate concentrations in groundwater are available for only eight observation wells that are currently included in the national groundwater monitoring network. Although these preliminary results clearly indicate the advantage of performing a risk intensity assessment, further research including data from more observation wells across the study area is needed to validate the performance of the vulnerability and risk intensity models and confirm the results of the current study.

4. Conclusions

In this study, the vulnerability of an alluvial aquifer is assessed using five different index-based models. In addition, the risk intensity of the resource is evaluated by combining the results of vulnerability and hazard assessments. This is the first study that evaluates groundwater vulnerability and the risk of contamination in the intensive agricultural area surrounding the town of Varaždin. The key findings are:

- Similar groundwater vulnerability results are obtained using five models, with the high vulnerability class covering most of the study area.
- Both DRASTIC and SINTACS parameters are independent with only a few significant correlations at a 95% confidence level.
- The theoretical and effective parameter weights for DRASTIC- and SINTACS-based models are in relatively good agreement, with the depth to groundwater and net recharge the most effective parameters.
- Despite the generally high groundwater vulnerability, a moderate resource contamination risk prevails as a consequence of a dominantly low hazard index.
- The correlation between the vulnerability indices and mean nitrate concentrations in groundwater is weak for all used vulnerability models. The second highest correlation coefficient (0.34) is obtained for the GOD method, which includes the fewest model parameters.
- A significantly higher correlation coefficient (0.58) is obtained when the groundwater vulnerability index is replaced by the resource risk intensity index.

This study demonstrates that the results of a resource risk intensity assessment are superior to groundwater vulnerability results in predicting groundwater contamination levels. Therefore, resource risk intensity maps should be an integral part of the decision-making process in the domain of physical planning and identifying areas where a change of land use is needed to achieve a trend reversal in groundwater quality. Validation of the groundwater vulnerability and risk intensity models was performed using only eight observation wells. Therefore, further investigation, including more observation wells in the national monitoring network, is needed to provide additional evidence to support the results of the current study.

Supplementary Materials: The following supporting information can be downloaded at: https: //www.mdpi.com/article/10.3390/su152316502/s1, Table S1. Hazard types with linear geometry; Table S2. Hazard weighting values for several example hazards (modified according to [51]); Table S3. Mean concentrations of nitrates in groundwater in the period 2007–2017; Table S4. Hazard types with polygonal geometry; Figure S1. The rated maps used to compute the DRASTIC vulnerability index; Figure S2. The rated maps used to compute the SINTACS vulnerability index; Figure S3. The rated maps used to compute the GOD vulnerability index.

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