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Concepts and methods for assessment of the risk for chemical contamination of groundwater with arsenic in river floodplains (overview)

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Abstract: The article provides an overview of the concepts and methods applied for evaluation of the risk for chemical contamination of groundwater and their consistency with the concept of ecological risk assessment. The aim of this study is selection of indicators for estimation of the specific risk for arsenic pollution of groundwater in contaminated river floodplains on future work. A procedure for calculation of the specific risk is suggested.

Keywords: ecological risk assessment, groundwater pollution, indicators, index method

INTRODUCTION

River valleys offer probably the best conditions for development of the social and economic life of human beings since the dawn of mankind until nowadays. Certainly, the anthropogenic impact to the environment and threat of pollution are one of the highest in the valleys compared with other landscapes on the Earth. Together with lowlands, they are the most populated areas on the planet where resources of fresh water are often insufficient and protection of their good quality is of high importance.

Arsenic (As) is a pollutant of great concern for a number of contaminated sites worldwide, usually as a result of intensive mining and smelting activities (Byrne et al., 2012; Mandaliev et al., 2013). It is recognized by WHO within the frame of the International Programme on Chemical Safety as one of the 10 most dangerous substances regarding human health (WHO, 2010). It is also included in the Priority Pollutant List of US EPA (US EPA, 2014). Groundwater contamination with As is documented at least in 36 countries on six continents (Rahman et al., 2006; Mukherjee et al., 2006; Smedley, 2006). River floodplain soils are often polluted with arsenic and heavy metals via inundation when contaminated sediment is deposited in the riverine lands. Tailings dam failures and mine tailings spills cause some of the worst environmental catastrophes leaving behind thousands of tons of contaminated sediment spread on the soil in the river floodplain. About 218 of such accidents are documented for the last 100 years (Azam and Li, 2010) and probably more of them left out of the registers. Well known are the accidents at Aznalcóllar mine in the Guadiamar Valley in Spain in 1998 (Edward et al., 2005; Kraus and Wiegand, 2006; Madejon et al., 2002) and at Baia Mare in the Someş Valley of Romania in 2000 (Brewer et al., 2003) which caused tremendous environmental damages and wide spread contamination. This type of technological disasters are often triggered by torrential rain falls (Azam and Li, 2010) while the number of extreme climatic and hydrologic events is expected to increase as a result from the climate change (Easterling et al., 2000).

Groundwater accumulated in the alluvial sediment deposits presents important source of fresh water in the river valleys. At the same time it is highly vulnerable to chemical pollution. Shallow water table and unconfined character of alluvial aquifers favor infiltration of pollutants from the land surface to the groundwater bodies. Chemical peculiarities of As species in the environment favor their transport in the vadose zone under the usual neutral to slightly acidic or slightly alkaline active reaction of the alluvial sediment (Bhumbla and Keefer, 1994). Arsenic is even more soluble under anoxic conditions which are rather typical for certain parts of the riparian zones and special attention should be paid to arsenic contaminated areas in river floodplains. The fact of the high susceptibility of alluvial aquifers to chemical pollution requires actions for assessment of the risk for groundwater pollution with As in river valleys where As contamination of soil is likely or has been already documented.

Specific hydrogeological and geomorphological settings in the river floodplains combined with specific behavior of As in the weathering zone require specific methods to be applied for this kind of assessment. To our best knowledge there is no appropriate method for assessment of specific risk for groundwater contamination with arsenic in river floodplains described in the literature. In this regard, the goal of this review is to find the most complete and appropriate method that can be adopted or modified in order to be used for this particular task. In order to ensure better insight of the problem, the review will first present the main concepts of ecological and groundwater risk assessment and analyze their consistency. It will be followed by an overview of the existing methods for groundwater risk assessment and appropriate indicators will be selected or suggested.

CONCEPT FOR ECOLOGICAL RISK ASSESSMENT

Ecological risk assessment is intended to evaluate the level of risk associated with anthropogenic activities that threaten ecosystems (EEA, 2008). Concept of Ecological Risk Assessment is considered more general and is expected to provide theoretical basis for assessment of risks related with each environment component, e.g. groundwater. The same authors define environmental risk as a broader term which includes both ecological and human health risk. First guidance and procedures for environmental risk

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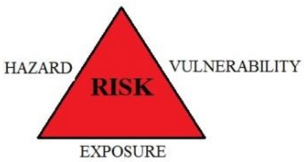
assessment has been probably developed and applied in the 70s and early 80s by US EPA. These were focused on health risk assessment, while surveys associated with ecological risk evaluations were launched by the Agency in 90s (US EPA, 2016). Our search of literature found most of the publications on ecological risk assessment to be issued in the period after the beginning of 90s. Risk related theoretical base and terminology is relatively new and developing, still being in process of unification for the various specific applications (Troldborg, 2010). Definitions of risk from a variety of literature sources are presented in Table 1. Most authors point at the probabilistic nature of the risk and express it by different combination between hazard, vulnerability and exposure. According to ISO 31010, risk is a combination of the consequences of an event (hazard) and the associated likelihood of its occurrence (EC, 2010). Similar to this view are the definitions of Varmes (1984) and Uricchio et al. (2004) for the term of ecological risk. US EPA (2016) provides the following explanation for ecological risk assessment: "…a process for evaluating how likely it is that the environment may be impacted as a result of exposure to one or more environmental stressors such as chemicals, land change, disease, invasive species and climate change". It should be noted that different authors consider different meaning for exposure with regard to risk assessment. In most cases it is related with the experience of the hazard impact by the receptor. Though the lack of complete unification of the risk related terminology, it can be stated that probability, hazard impact, exposure and vulnerability are mile stones in the concept of ecological risk assessment and they should be considered when the risk for groundwater pollution is evaluated.

Table 1. Definitions of risk, risk assessment and ecological risk assessment

Source	Definitions
Royal Society (1992)*	*Risk*: "The combination of the probability, or frequency, of occurrence of a defined hazard and the magnitude of the consequences of the occurrence".
Smith, 1996*	"*Risk* is the actual exposure of something of human value to a hazard and is often regarded as the combination of **probability and loss**".
Helm, 1996*	***Risk* = probability*consequences.**
Crichton, 1999*	"*Risk* is the probability of a loss, and depends on three elements, **hazard, vulnerability and exposure**".  The "risk triangle"
Varmes, 1984; Uricchio et al., 2004	In general terms, risk is defined as a combination of hazard and vulnerability, as follows: **risk=vulnerability*hazard**.
Schneiderbauer et al., 2004	*Risk* is "the probability of harmful consequences or expected losses resulting from a given hazard to a given element at danger or peril, over a specified time period".
Schmidt-Thomé et al., 2005	"*Risk***:** A combination of the probability (or frequency) of occurrence of a natural hazard and the extent of the consequences of the impacts. A risk is a function of the exposure of assets and the perception of potential impacts as perceived by a community or system".
ISO 31010 (EC, 2010)	*"Risk* is a combination of the consequences of an event (hazard) and the associated likelihood/probability of its occurrence".
EC, 2010	*Risk:* "When the extent of the impacts is independent of the probability of occurrence of the hazard, such as earthquakes or storms, risks are the combination of the consequences of an event or hazard and the associated likelihood of its occurrence: **risk = hazard impact * probability of occurrence**". *Risk:* "Where the size of the impact influences the likelihood of occurrence, risk is a function of the probability of occurrence of a hazard (p), the exposure (E -total value of all elements at risk), and the vulnerability (V - specific impact on exposure): **risk=ƒ(p*E*V)**".
Directive 2012/18/EU, 2012	"*Risk* means the likelihood of a specific effect occurring within a specified period or in specified circumstances".
Suter, 1993	"*Risk assessment* can be defined as the process of assigning magnitudes and probabilities to the adverse effects of human activities or natural catastrophes".
US EPA, 1992, 1998	*"Ecological risk assessment...*evaluates the likelihood that adverse ecological effects may occur or are occurring as a result of exposure to one or more stressors". "The ecological risk assessment process is based on two major elements: **characterization of effects** and **characterization of exposure**".
Australian Government, Department of the Environment,	"*Ecological risk assessment* is a term ascribed to the method(s) for determining risk posed by a stressor (contaminant or perceived threat) to the survival and health of ecosystems.

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2016	*Risk* is quantified as the probability of an adverse event, or the likelihood of exposure multiplied by the consequences or effects of that exposure: r**isk = exposure*effects**".

*The definition of Crichton (1999) is taken from a similar table in Brooks (2003); definitions of Smith (1996) and Helm (1996) are taken from a similar table in Kelman (2003); definition of Royal Society (1992) is cited by the European Environment Agency (EEA, 2008).

CONCEPT FOR GROUNDWATER POLLUTION RISK ASSESSMENT

The concept for groundwater pollution risk assessment was established in the late 1980s when Foster (1987) states that "groundwater pollution risk is to conceive the interaction between: a. the natural vulnerability of the aquifer; b. the pollution loading that is, or will be, applied on the subsurface environment as a result of human activity" (fig. 1A). Since 1988 the methodology is restricted to the assessment of groundwater pollution risk from man's activity at the land surface (Foster and Hirita 1988) (fig. 1B).

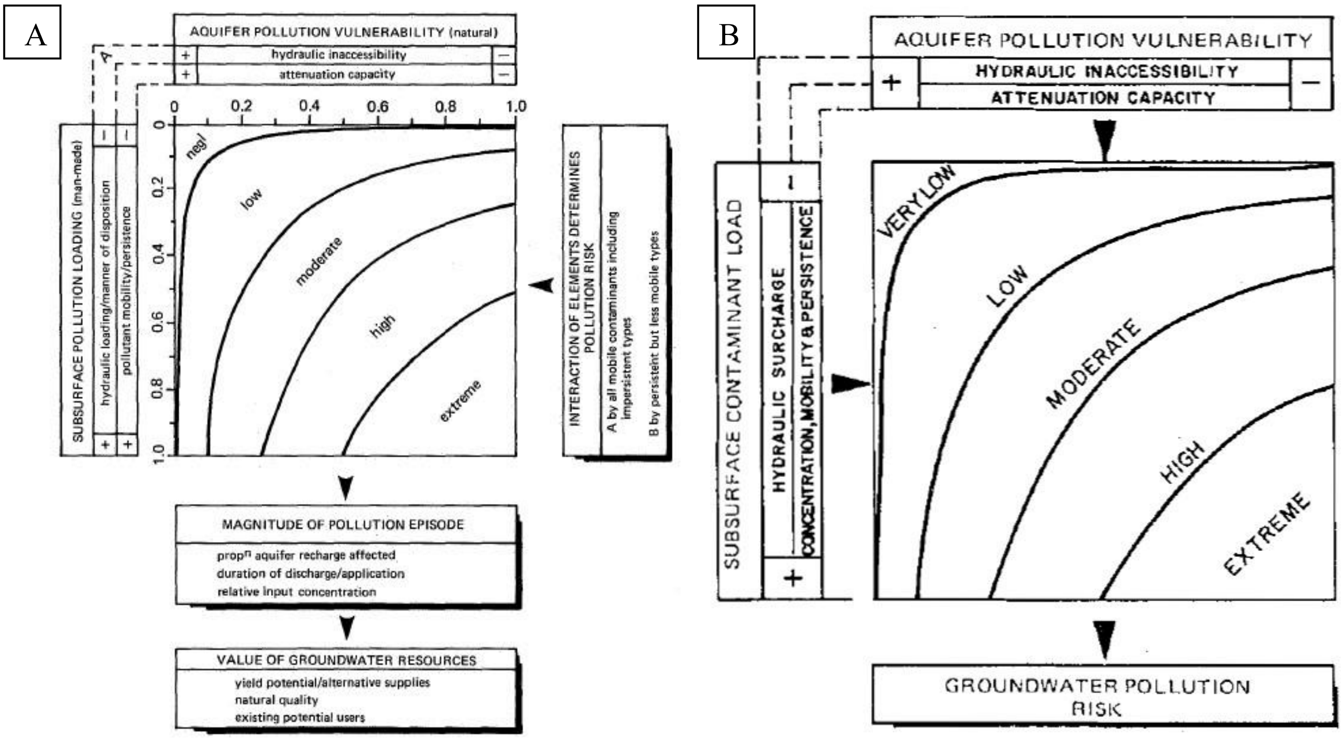

Figure 1. General conceptual scheme for groundwater pollution risk assessment: A -Foster (1987); B – Foster and Hirita (1988)

The concept for groundwater pollution risk assessment is focused mainly on two aspects: aquifer vulnerability and magnitude of contaminant load near the land surface. It is illustrated by the definitions listed in Table 2 and by Figure 2.

Table 2. Definitions of groundwater pollution risk

Source	Definitions
Foster and Hirata, 1988; Adams and Foster, 1992	"As the probability that the uppermost part of an aquifer will become contaminated to an unacceptable level by activities on the land surface, and will be the result of the interaction between **the intrinsic vulnerability** and **the contaminant loading** applied at the location concerned."
Diamantino et al., 2005	"As a general definition risk could be defined as the superimposition of two factors that can be characterized separately: **the vulnerability of the physical medium** and the pollutant load or **hazard applied on the subsurface environment** as a result of human activity."
Saidi et al., 2010, 2011	"The risk of pollution is determined by both the intrinsic characteristics of the aquifer, which are relatively static, and the existence of potentially polluting activities, which are dynamic factors that can be changed and controlled. The risk intensity map is based on the risk intensity index and calculated by multiplying the reciprocal value of **the hazard indices** with the **vulnerability factor**".
Pacheco et al., 2013	"A (potential) risk of groundwater being contaminated, described by the following relation: **potential risk = intrinsic vulnerability + specific vulnerability**, where intrinsic vulnerability is determined by the seven DRASTIC features (Depth to Groundwater, Net Rechrge, Aquifer Media, Soil Media, General Topography or Slope, Vadose Zone and Hydraulic Conductivity of the Aquifer) and specific vulnerability is set by land use"

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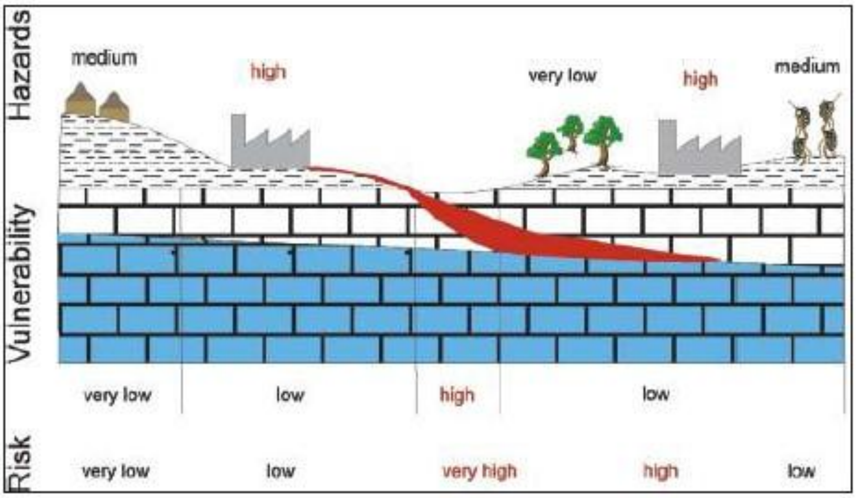
Two possible approaches for assessment of groundwater pollution risk are specified in the Technical report on groundwater risk assessment (2004) supporting the Common Implementation Strategy for the Water Framework Directive 2000/60/EC (WFD). The direct one is associated with chemical quality monitoring of the groundwater body, while the indirect method which is more relevant to WFD involves estimation of subsurface contaminant load and pollution vulnerability of the threatened aquifer.



Fiure 2. Risk assessment of groundwater considering the superimposed effects of hazards and vulnerability (Source: Hötzl et al, 2003)

Risk assessment of contaminated sites is often based on a source - pathway-receptor concept (Troldborg, 2010) as shown on Fig. 3. It is well developed in the guidelines elaborated by the US EPA.

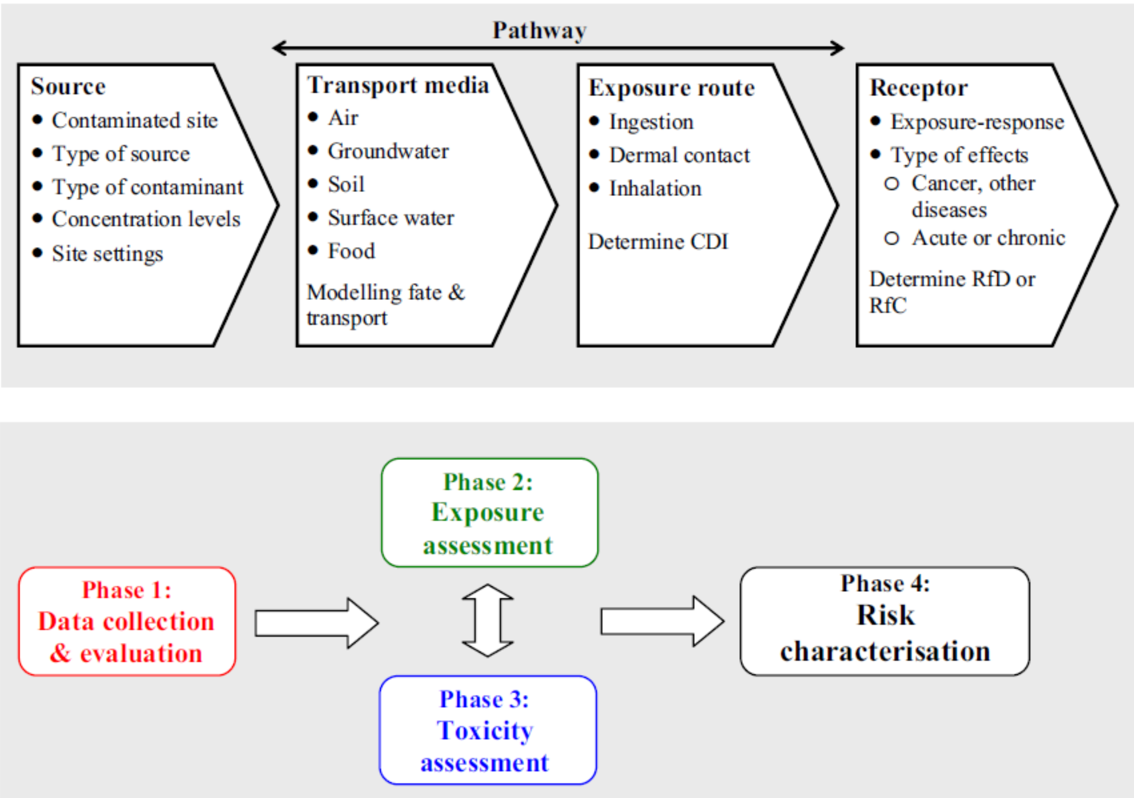


Figure 3. A concept of risk assessment of contaminated sites (after Troldborg, 2010)

Main differences between available approaches for estimation of groundwater pollution risk are due to their focus either on groundwater quality or on the threat to public health and ecosystems. In many cases assessment of the risk for exceeding certain groundwater quality thresholds is only required, for example when prioritization of areas for more detailed survey of aquifer contamination is needed. In such cases the first approach is preferred and aspects of toxicity and exposure may not be considered. The survey is then focused on the aquifer vulnerability and magnitude of contaminant load near the land surface and groundwater is considered an end-point of the hazard impact. This concept is well developed in the works of Foster and his co-authors (Foster, 1987; Foster and Hirita,1988; Adams and Foster, 1992; Foster et al., 2007) and used for the purpose of this study.

METHODS FOR GROUNDWATER POLLUTION RISK ASSESSMENT

Methods associated with the concept considering aquifers as end-points of contaminant transfer will be subject of the present review. They reflect the two main aspects of the assessment - vulnerability and hazard. The hazard is defined as contaminant load at the subsurface.

The concept of groundwater vulnerability was established in the late 1960s and was developed in the 1980s and the 1990s. The first definition of Margat (1968) states that vulnerability is "the degree of protection that the environment provides against the spread of pollution in groundwater". The definition has been amended through the years, but to the present day there is still no standard definition. It is our opinion that the term 'vulnerability of groundwater to chemical pollution' should mean the probability that chemicals of anthropogenic or natural origin to infiltrate from the surface to the water table and go beyond certain threshold concentration in groundwater.

Vulnerability can be assessed with several groups of methods including index methods, mathematical modeling, e.g. numerical and statistical models (Management…, Vol. 4, 2003), simulation (Liggett et al., 2009) and hybrid methods (Antonakos et al., 2007). The choice of particular method depends on data availability, as well as on the geomorphological and the hydrogeological settings of the study area.

Index methods are the most widely used ones for assessing the vulnerability of groundwater to chemical contamination. The advantage of these methods is that they use publicly available data and are cost effective. After analyzing the available information it can be organized in different layers using geographic information systems (GIS). Index methods are based on a combination of data on various environmental factors (depth of groundwater, lithology, topography, soil cover, etc.) where a score is given to each factor in order to evaluate its impact on the contaminant transport from surface to the aquifer. DRASTIC index (Aller et al., 1987) and its modifications are the most widely used for groundwater vulnerability assessment surveys (Foster et al., 1995; Adamat et al., 2003; Diamantino et al., 2005; Nobre et al., 2007; Saidi et al., 2011; Wang et al., 2012; Pacheco et al, 2013; Pisciotta et al., 2015; Bartzas et al., 2015). For areas with specific hydrogeological settings, e.g. karst territories, index methods like SINTACS (Civita et al, 1997; Uricchio et al., 2004; Pisciotta et al., 2015), GOD (Foster, 1987; Foster et al., 1988; Chilton et al., 1990; Foster et al., 2002), GLA (Hoelting et al., 1995), PI (Goldscheider, 2002) and EPIK (Doerfliger et al., 1999) have been developed. Guo et al. (2007) created a modification of DRASTIC index for evaluation of specific vulnerability of groundwater to arsenic contamination, named DRARCH. The new index leaves out some parameters of DRASTIC and adds new ones, which reflect the ability of geological horizons to retain arsenic (Stoyanova, 2013, 2015). However, the study did not combine vulnerability with hazard assessment in order to evaluate the risk of groundwater arsenic contamination. What is more, the used factors did not consider the specific chemical peculiarities of arsenic and the hydrogeological conditions in the riverine areas.

UNISDR defines hazard as "…a dangerous phenomenon, substance, human activity or condition that may cause loss of life, injury or other health impacts, property damage, loss of livelihoods and services, social and economic disruption, or environmental damage" (EC, 2010). In regard to groundwater pollution risk assessment hazard can be considered to be a potential source of contamination resulting from human activities and taking place mainly at the land surface, and that in particular circumstances could lead to harm in given area for a certain period of time (Passarella et al., 2002; Zwahlen, 2003, Uricchio et al., 2004; Saidi et al., 2011; Wang et al., 2012). Zwahlen (2003) suggests the following formula for hazard assessment: HI = H*Qn*Rf, where HI = the hazard index; H = the weighting value of each hazard; Qn = the ranking factor (0,8-1,2); and Rf = the reduction factor which provides an assessment of the probability for a contamination event to occur. In case of risk assessment for contaminated sites where the contamination event has already happened, the hazard can be associated with the subsurface contaminant load (Foster and Hirata, 1988; Adams and Foster, 1992; Diamantino et al., 2005). In this case the risk related probability is associated with the likelihood that the concentration of a certain pollutant in groundwater may exceed a given threshold (Uricchio et al., 2004).

Although significant studies worldwide have been recently focused on characterizing the spread of arsenic in groundwater and on the processes of its mobility in aquifers (Fazal et al., 2001) we were not able to find a method established particularly for assessment of the risk for chemical contamination of groundwater with arsenic in the hydrogeological settings of river floodplains.

PROCEDURE FOR CALCULATION OF THE RISK FOR GROUNDWATER CONTAMINATION WITH ARSENIC IN RIVER FLOODPLAINS

Proposed here procedure for calculation of the risk for groundwater contamination with arsenic in river floodplains is based on the concept which considers the groundwater as an end-point receptor of the hazard impact and presents the risk as superimposition of groundwater vulnerability and subsurface contaminant load:

Risk = Specific Vulnerability * Arsenic Load in soil.

It is developed particularly for the arsenic contaminated Ogosta Valley in the NW Bulgaria, but can be easily adapted for other river floodplains.

DRESPI index (Stoyanova, 2015, Stoyanova et al., 2016) is applied in order to assess the specific vulnerability of groundwater contamination with arsenic in river floodplains. It is a modification of the DRASTIC method. The acronym DRESPI is created from the first letters of the following six parameters integrated in the final assessment: Depth to groundwater table (D); net Recharge (R); Eh - redox state of soil (E); Soil texture (S); pH - active soil reaction (P); Impact of the soil thickness (I). The indices of DRESPI can be divided into two groups: (1) for evaluation of hydrogeological settings – D, R, S, I; (2) for assessment of As mobility in the vadose zone – E, P.

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The DRESPI index calculates the sum of product of ratings and weights assigned to each of the parameters.

$$Di = D_R*D_W + R_R*R_W + E_R*E_W + S_R*S_W + P_R*P_W + I_R*I_W$$

Where: Di – DRESPI index, R – rating, W – weight.

The minimum DRESPI index value is 23 and the maximum is 219 It resulted in dividing the whole range into the following five classes: 23-79 (negligible vulnerability), 80-119 (low vulnerability), 120-159 (moderate vulnerability), 160-199 (high vulnerability) and 200-219 (extreme vulnerability) (Table 3).

Table 3. Classes vulnerability to arsenic contamination in the floodplain

DRESPI				
Range	Color	RGB	Vulnerability	Rating
<79	Dark Green	38/118/0	Negligible	1
80-119	Green	10/204/0	Low	2
120-159	Yellow	255/255/0	Moderate	3
160-199	Red	255/0/0	High	4
>200	Violet	194/0/187	Extreme	5

The assessment of subsurface contaminant load was performed using the maximum admissible concentration of As for arable lands (25 mg/kg) and intervention level (90 mg/kg) set by Bulgarian legislation (Regulation 3/2008 for the thresholds of harmful substances in soil). The set thresholds take into account the impact of soil to aquifers and are intended also for risk assessment of groundwater pollution (Atanassov, 2007). If no appropriate thresholds are available, the estimation of contaminant load could be performed using the background concentration of pollutant in soil.

Three classes of hazard associated with the contaminant load are determined as follows: low hazard, moderate hazard and high hazard (Table 4).

Table 4. Classes of arsenic load in floodplain soil

Range mg/kg	Color	RGB	Hazard	Rating
<25	Green	10/204/0	Low	1
25-90	Yellow	255/255/0	Moderate	2
>90	Red	255/0/0	High	3

A public data set can present contaminant concentrations at various soil depths even for one and the same study area as it is the case with the study of Ogosta Valley. Subsurface contaminant load can be then determined considering several options: average concentration to a fixed depth available for all the sampling points in the data set; maximum levels of contaminant in the sampled soil profiles; pollutant content in the topsoil (0-20 cm).

A matrix is used for combining vulnerability and arsenic contamination of floodplain soil, thus calculating the risk level (Table 5).

Table 5. Risk matrix

Risk	**Vulnerability**				
Hazard	**1**	**2**	**3**	**4**	**5**
1	1	2	3	4	5
2	2	4	6	8	10
3	3	6	9	12	15

Table 6. Classes for assessment of the risk for chemical contamination of groundwater with arsenic in floodplains

Range	Color	RGB	Risk
1	Dark Green	38/118/0	Negligible
2-3	Green	10/204/0	Low
4-7	Yellow	255/255/0	Moderate
8-14	Red	255/0/0	High
15	Violet	194/0/187	Extreme

The minimum risk value is 1 and the maximum is 15 (Table 6). It resulted in dividing the whole range into the following five risk classes: negligible (1), low (2-3), moderate (4-7), high (8-14) and extreme (15). A green-yellow-red color pattern is used to distinguish the risk classes. Risk maps can be easily created and kept updated if the data is organized and processed in GIS environment.

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CONCLUSIONS

The concepts of ecological risk assessment are rather in good consistency with the concepts of the groundwater pollution risk assessment. Depending on the target end-point receptor of the hazard impact, the groundwater contamination risk assessment would consider or not the aspects of toxicity and exposure. In case of contaminated sites the risk related probability is associated with the likelihood that the concentration of a certain pollutant in groundwater may exceed a given threshold. Development of index methods for groundwater risk assessment has not provided specific procedures for certain contaminants of the group of inorganic persistent hazardous substances, e.g. arsenic and heavy metals, as well as for particular type of environment or landscape[1]. Thus, modification of the available methods and elaboration of new ones are needed for more accurate assessment of risk of environmental pollution under variety of chemical substances and environmental conditions. The proposed modified assessment procedure integrates new indices to provide more accurate evaluation of groundwater risk to As contamination in river floodplains.

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[1] The term is used according to the concept for the landscape as a specific complex of environmental components with spatial development and boundaries

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