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FLUX-BASED RISK ASSESSMENT OF THE IMPACT OF CONTAMINANTS ON WATER RESOURCES AND ECOSYSTEMS
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FRAC-WECO

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FLUX-BASED RISK ASSESSMENT OF THE IMPACT OF CONTAMINANTS ON WATER RESOURCES AND ECOSYSTEMS

"FRAC-WECO"
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ACRONYMS AND ABBREVIATIONS

Chemical substances

CAH .... Chlorinated Aliphatic Hydrocarbon
BTEX .... Benzene Toluene Ethylbenzene Xylene
DCE .... Dichloroethene
cDCE *cis-1,2*-dichloroethene
PAH .... Polycyclic Aromatic Hydrocarbons
TCE .... Trichloroethene
VC .... Vinyl Chloride

Others

ChemRI .... Chemical Risk Indice
EnvRI .... Environmental Risk Indice
EtoxRI .... Ecotoxicological Risk Indice
GwQT approach .... Groundwater Quality TRIAD-like approach
IA model .... Independent Action model
PE .... Percentage of Effect
PhysRI .... Physico-chemical Risk Indice
SEQ-ESO .... Système d'Evaluation de la Qualité des Eaux Souterraines
TU .... Toxic Unit
VITO .... Vlaamse Instelling voor Technologisch Onderzoek
DO .... Dissolved Oxygen
ORP .... Oxidation Reduction Potential
TOC .... Total Organic Carbon
DNA .... DeoxyriboNucleic Acid
k .... Degradation constant
$T_{1/2}$ .... Half life time of chemical organic species
WFD .... Water Framework Directive
TEV .... Total Economic Value
CEA .... Cost Effectiveness Analysis
CBA .... Cost Benefit Analysis
DGRNE .... Direction Générale des Ressources Naturelles et Environnement
WTP .... Willingness To Pay
OLS .... Ordinary Least Square
CILE .... Compagnie Intercommunale Liégeoise des Eaux
EPA .... Environmental Protection Agency
INERIS .... Institut Nationale de l'EnviRonnement industriel et des rISques

GIS .... Geographical Information Systems
VHR .... Very High Resolution
MLP .... Multiple Layer Perceptron
COSW .... Carte d’Occupation du Sol de Wallonie
FRAC-WECO .... Flux-based Risk Assessment of Contaminants on Water resources and ECOsystems
SPR .... Source Pathway Receptor
RRA .... Regional Risk Assessment
SEQ-ESO .... Système d’Evaluation de la Qualité des Eaux SOuterraines
PNEC .... Predictive No Effect Concentration
NOEC .... No-Observed Effect Concentration

SUMMARY

The FRAC-WECO project elaborates a framework for the assessment, at the regional scale, of the risk posed by contaminated sites on water resources and ecosystems. The general methodology developed in the project relies on several key research and tools developments, on the one hand, process studies contributing to a comprehensive assessment and modelling of water and contaminant fluxes at the scale of interest and of biogeochemical properties and toxicity of contaminants, on the other hand, regional indicators of water quality deterioration and ecotoxicity, so as to propose management tools and indicators for contaminated sites ranking in terms of risks and costs. It consists in evaluating the level of water degradation at the scale of the groundwater body as a referential for decision-making in terms of water resources management in the perspective of the EU Water Directive (trend assessment and reversal) and for an objective socio-economic analysis evaluating, in monetary terms, the damages related to water resources deterioration and the costs for restoration.

To do so, existing and potential sources of contamination are identified in the study zone (usually of an extent corresponding to that of the (ground)water body). Using analytical or numerical models for contaminant leaching in the unsaturated zone and dispersion in groundwater, contaminant fluxes to the receptors are calculated providing a way of estimating the level of exposure / degradation of these receptors (groundwater, surface water through groundwater discharge and possible specific receptors such as sensitive groundwater dependant ecosystems). The modelling application relies on a regional scale groundwater flow model developed using MODFLOW. Groundwater recharge, as identified as the main vector of contaminant leaching and dispersion, is obtained from maps based on the object-oriented land-use classification of the complex urbanized environment characterizing industrial and urban zones. Contaminant properties governing their behaviour in soil and groundwater (sorption, degradation constants ...) have been gathered for literature, previous studies and most importantly from degradation studies carried out specifically for this project on selected real test cases.

The regional risk for groundwater and surface water bodies is calculated as follows. The different contaminant plumes generated using the contaminant dispersion models are classified in zones of equivalent degradation using the SEQ-ESO system considered in the Walloon Region of Belgium, adapted for a grid-based application in the context of the project. An aggregated indicator of groundwater degradation is obtained as the mean of the SEQ-ESO indicators weighted by the corresponding

volumes of degraded groundwater to the total volume of water in the studied groundwater body. A second indicator is being developed to focus more specifically on the regional ecotoxicological risk. It is based on a modified TRIAD approach, considering both water quality and ecotoxicity data.

The final result of this research project is a regional scale risk assessment methodology for groundwater bodies, proposed as a flexible approach for evaluating the pressure exerted by various sources of contamination on groundwater resources. The RRA provides two type of groundwater quality indicators, (1) a map of spatially distributed indicator that can be used to identify most problematic sectors (hot contamination spots) and their evolution with time and (2) the spatially-aggregated indicator that can be used to report on the global status of groundwater quality in the groundwater body, for groundwater quality trend assessment and as a referential for site prioritization and for the evaluation of programs of measures aiming at restoring groundwater quality in the groundwater body, using cost-efficiency approaches. This RRA is compliant with the ongoing legislation in the Walloon region, based on the SEQ-ESO and it fits very well with the guidelines of the EU Water Directive which promotes the use of aggregated indicators able to reflect status and trends in groundwater quality.

The regional risk assessment approach proposed here is data demanding. However, this is the case for all these regional scale approaches that require relatively important preparatory works for data acquisition, organisation and processing. This drawback is partly overcome by the use of a geospatial database and better characterization of existing polluted site with good organization of data. There are many sources of uncertainties in the different data feeding the approach. In particular, the pollution sources are not always perfectly identified and characterized and hydrogeological parameters and contaminant properties remain difficult to identify and quantify at the regional scale. A key next step will be to implement a statistical approach for handling all the uncertainties that remain at regional scale.

Results of the RRA performed on a groundwater body can now be used as a referential for a cost - benefit assessment of total or partial remediation of the contaminated groundwater body, according to different management scenarios. This analysis starts from the actual groundwater body quality state for which groundwater restoration scenarios are suggested. The regional risk assessment method is then applied on these scenarios to evaluate the resulting improvement of the groundwater quality index when management plans are applied. This allows evaluating, in monetary terms, the costs for given improvements in the groundwater body quality status and the benefits that can be released.

1. INTRODUCTION

1.1 General context and objectives

The EU Water Framework Directive requires management plans to monitor, to maintain and, if necessary, to restore the quality of surface water and groundwater bodies. In very urbanised and industrialised regions, water resources, and particularly groundwater, are subject to many pollution pressures related to different kinds of socio-economic activities and contaminants (UNEP/ADEME 2005, European Environment Agency 2006). These plans cannot be defined without considering industrial sites potentially harmful to groundwater resources and, in this context, different questions arise. How can we take into consideration all these potential and actual sources of pollution in evaluating the risk of deterioration of groundwater quality and the efficiency of programmes of measures defined to restore this quality? And, as a consequence of this, how can we evaluate groundwater quality at the regional scale of the groundwater body and the evolution with time of groundwater quality?

Classical risk assessment and management concepts for contaminated sites are usually based on a univocal relationship between a source of pollution and a potentially exposed receptor, commonly referred to as the source – pathway – receptor approach, with an evaluation of the receptor's exposure level and a comparison to environmental and health regulations (e.g. Fairman et al., 1990; Ferguson et al., 1998). This conceptual approach is convenient for pollution sources and receptors well located in space, such as local pollution "hotspots" and pumping wells nearby. By contrast, in heavily industrialised and urbanised areas, because of the spatial extent of groundwater bodies, many point or diffuse pollution sources may need to be considered in the analysis, with complex groundwater vectors for contaminant dispersion, and a meaningful regional risk assessment approach has to be considered (Gay and Korre, 2006; Critto and Sutter, 2009).

Several projects have been dedicated recently to the development of methodologies for contaminated megasite management, such as WELCOME (2004), INCORE (Ptak et al., 2003; Jarsjö et al., 2005), DESYRE (Carlon et al., 2007; 2008), SAFIRA II (Morio et al., 2007; Schädler et al., 2007). These projects propose concepts and tools for a regional analysis of environmental issues related to contaminated sites, for regional risk assessment, for prioritization of investment and rehabilitation on industrial land parcels and brownfields or for cost-benefit socio-economic analyses. Beside these megasite-oriented projects, other decision support systems have been developed, based on GIS systems, e.g. SMARTe (Vega et al., 2009) or DECERNS

(Sullivan et al., 2009) for data management and cartography and for regional scale risk assessment for contaminated sites, SADA (Purucker et al., 2009) or ERAMANIA (Semenzin et al., 2009) for ecological risk assessment, BOS (Tait et al., 2004; 2008, Chisala et al., 2007) for the management of groundwater in urban areas and BASINS (Kinerson et al., 2009), RISKBASE (Brils and Harris, 2009) or CatchRisk (Troldborg et al., 2008) for water pollution risk assessment and management at catchment scale.

However, these projects do not really propose specific indicators for the quality of regional groundwater resource and associated ecosystems (Caterina et al., 2009). The general objective of the FRAC-WECO project is thus to fill this gap by proposing a flux-based pollution risk assessment approach that consider the cumulative effect of multiple, spatially-distributed, pollution sources with multiple types of contaminants on groundwater resources and groundwater-dependent ecosystems.

1.2 Challenges and research needs

In order to reach the general objective, different scientific aspects have to be addressed. Appropriate indicators have to be established as a referential and a support to decision making for groundwater quality/trend, for pollution risk (specific objective 1) and ecological risk (specific objective 2) assessment. Such indicators should reflect the level of groundwater quality deterioration at the regional scale, seen as a resource assuring different functions, including its contribution to the ecological quality of surface water bodies (through groundwater base flow). Because in urbanized / industrialized catchments, groundwater quality monitoring networks will never be sufficiently dense to intercept all the contaminant plumes at the regional scale, the indicators should be constructed using a forward approach, using appropriate regional groundwater flow and transport models to calculate contaminant dispersion in groundwater. This modelling includes the censuses of the leaking pollution sources (specific objective 3) and the compilation of the specific properties of contaminants coming from field and lab experiments and literature or databases (specific objective 4). In addition, because of the complexity and heterogeneity of land use with partial imperviousness, advanced approaches integrating remote-sensing and hydrological budgeting are required to estimate as accurately as possible the groundwater recharge patterns and related contaminant leaching (specific objective 5). In order to apply the concepts to real case studies, GIS-based platforms have to be developed and fed with relevant data and information on land use, geology, hydrogeology, industrial activities, leaching sources and released pollutants (specific objective 6). Finally, a framework for the definition of programmes of measures has to be proposed, based on a cost- benefit analysis of groundwater remediation at regional scale, with a quantification of the value of groundwater (market and non-market) and gross estimates for clean-up procedures (specific objective 7). These seven specific objectives were reflected, in a somehow different order, in the workpackages of the FRAC-WECO project.

Groundwater quality assessment or deterioration can be investigated using “status” (chemical analysis of water) and using “effects” (response of the ecosystem to the actual status).

The chemical status can be measured using groundwater analysis coming from monitoring wells network or either estimated using groundwater flow and transport modelling, based on the definition of sources of pollutants. The effects on the ecosystem can also be measured using traditional ecotoxicological test performed on groundwater samples but the estimation of these effects is more complicated. This estimation lies on relationships defined between the concentration of a particular chemical and the response of specific living specie. These relations can be found in literature but are very specific to the parameters applied for the ecotox test (physical conditions, tested living species …) and therefore quite difficult to generalize.

At site scale, the groundwater quality is nowadays well investigated using various techniques for site characterization, often combined with modelling for more details. On the other hand, measurement of the groundwater quality is hopeless at regional scale because it would suppose to dispose of an unrealistically large number of monitoring wells located in the area of study. The only way to avoid this is to estimate the groundwater quality by modelling. Regional scale groundwater flow and transport models are currently used but they are still currently a bit rough and suffer of lack of contaminant sources information.

At the site scale, the status aspect of the groundwater quality is well measured, it is equivalent to classical site characterization. At the regional scale, only few general approaches exists (see above) but not really one that integrate the effect of various sources of pollution, contaminant and pollution behaviour into a global indicator for the investigated area of interest. Investigation of groundwater quality by the effects is even more complicated. At the site scale some approaches exists but there is still difficulties concerning integration of ecotoxicological tests performed at different location on the site into an indicator for the whole site. At the regional scale it is nowadays impossible. It would imply that, from results of modelling (a grid with concentrations in different pollutants), it could be possible to define the ecotoxicological status of a groundwater body. It would mean to know the relation between concentration and ecotox effect (mortality, inhibition of reproduction, etc) for different living species, for each contaminant and, more difficult, for the mixture of the contaminant. That would mean infinity of relations between concentration and effects.

The objective of the FRAC-WECO project is to fill the gaps in the evaluation of groundwater quality by studying the status at the regional scale using groundwater flow and transport modelling and methodology of integration of various contaminants/contaminations and by studying the effects at the site scale with the integration of ecotoxicological tests for the complete characterization of a contaminated site.

1.3 Report organization

In the following chapter, the general methodology and related research components are described in details and illustrated using a real applications in Belgium ranging from the scale of a contaminated site (i.e. Chimeuse, Vilvoorde) to the scale of a deteriorated groundwater body (i.e. groundwater body RWM073 Alluvial gravels of the Meuse river between Engis and Herstal).

2. METHODOLOGY AND RESULTS

2.1 General methodology

The methodology for regional risk assessment is summarised in Figure 1.

In the groundwater body of interest, the various required information are identified and geo-referenced into a specific geospatial database (Figure 1a). This database is the basis for the entire regional risk assessment procedure and is organized according to the Source-Pathway-Receptor schema. The “Source” comprises information about contaminated sites (actual or potential) and the specific physico-chemical properties of the considered pollutants. Such specific properties can be assessed from laboratory or field experiments (available or specifically performed) or from literature surveys on similar hydrogeology and contamination context (Figure 1b). The “Pathway” gathers all the data about the physical environment, from the land surface to the aquifer and their related attribute properties (soil map, land-use, DEM, hydrogeology). The “Receptor” is here the groundwater body as a whole and its quality criteria for risk assessment that are characterized by the water use that it is made of it. The geodatabase also contains specific modules such as a matrix developed to define probabilistic values of expected concentrations of pollutants released in groundwater from potentially contaminated site knowing the prevailing industrial activities and their sizes. Infiltration from rainfall is the main driver for contaminant leaching from the pollution sources to the groundwater table. In this context, a land cover mapping with object-oriented classification methodology based on high resolution satellite data is developed and used in combination with hydrological modelling in order to distribute in space the recharge as a function of the complex industrial and urban land-use and soil imperviousness (Dujardin et al. 2009) (Figure 1c).

For each of the selected contaminant sources, the local infiltration leaching through the unsaturated zone is calculated using the data from the detailed spatially and temporally distributed recharge, and pollutant and unsaturated zone physico-chemical properties in order to define pollution entry in the saturated zone for groundwater modelling (Figure 1d).

The migration of contaminants within the aquifer is then calculated using a numerical groundwater flow and transport model developed and based on general data coming from the database and from case specific literature reviews or field studies performed on the groundwater body of interest. Repeating the same procedure for all identified sources of contamination provides maps of contaminants' plumes in the studied groundwater body (Figure 1e) at different times.

The generated plumes are classified in terms of groundwater quality classes on a normalized scale that accounts for threshold values defined specifically for each contaminant, using the SEQ-ESO evaluation system used in the Walloon Region (Rentier et al., 2006) for groundwater body chemical status. This gives a global picture of the quality status of the groundwater body (Figure 1f). Finally, a global quality index (Iglobal) is calculated for the groundwater body at each time step, using a weighting-average formula (Figure 1g).

Results of the Regional Risk Assessment are finally used in a socio economical study. From the evaluation of benefits gained from a good quality status groundwater body and the typical costs of the remediation of polluted sites, various management scenarios are tested to provide a cost-benefit analysis (Figure 1h). The cost of the remediation measures required for restoring the quality of the groundwater body by cleaning the contaminated site have been evaluated using standardized costs. This remediation will cause improvement of groundwater quality and thus increase its economic value. The concept of Total Economic Value (TEV) is here used to quantify the benefits of having a clean groundwater body by evaluating use (commercial use and ecosystem benefits) and non-use value (citizen willingness to pay for a preserved groundwater resource for future generations).

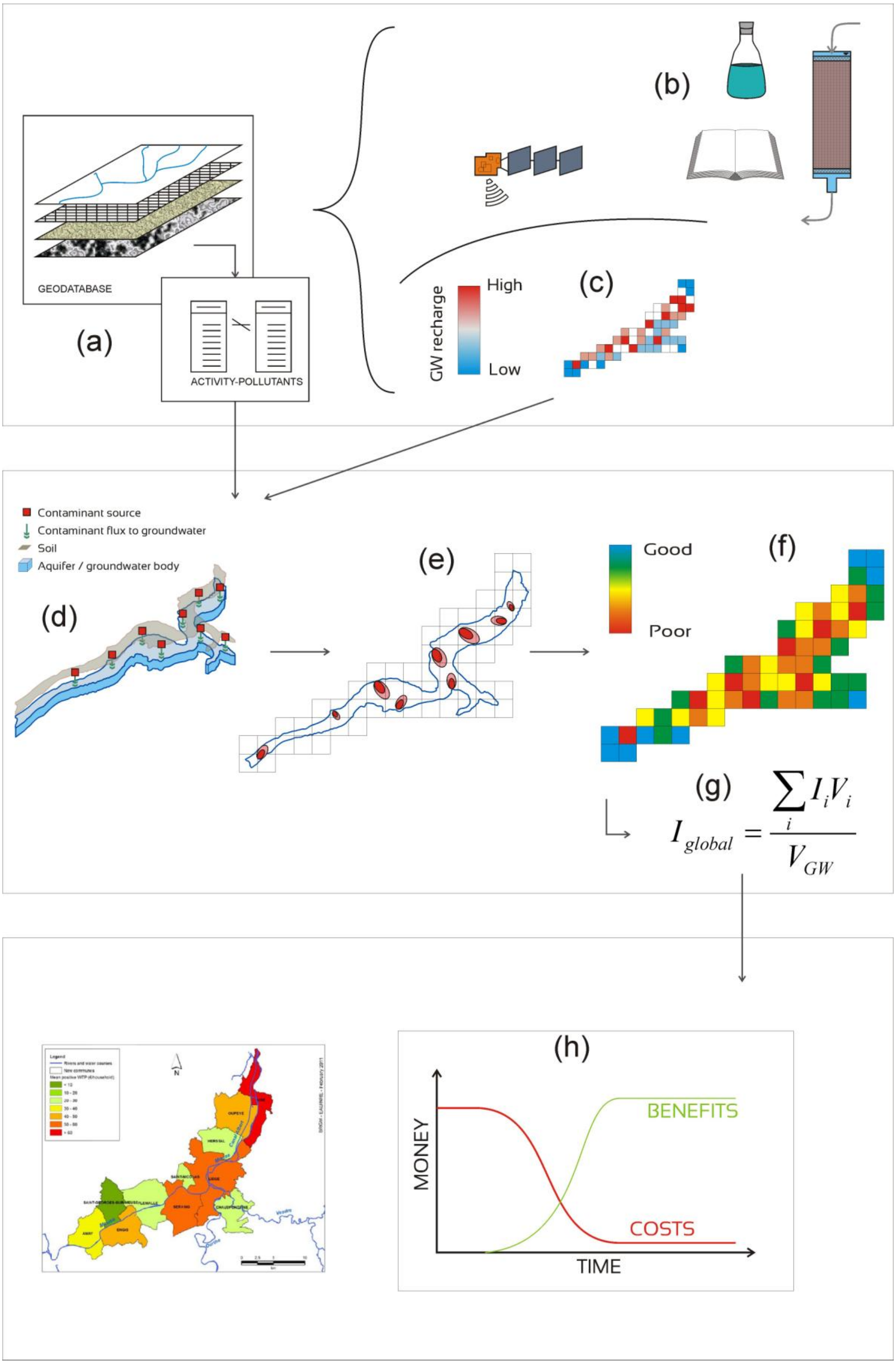


Figure 1: General Region Risk Assessment methodology

The different modules and research elements composing the regional risk assessment system are described in details in the following sections.

2.2 Project databases for data management and handling

2.2.1 Description of the geodatabase

A geodatabase, organized according to the Source-Pathway-Receptor schema, has been developed to store and manage the important quantity of spatially distributed data required by the regional approach. This geodatabase has been developed under Microsoft Access environment and uses shapefiles and attributes tables for handling environmental data. The conceptual schema of the geodatabase is shown in Figure 2. The “Source” module gathers the information on potential contaminated sites. It is used to characterize the nature, size and location of industrial sites. It also contains a dictionary of seventy-five selected pollutants with their main physico-chemical properties (e.g. solubility, Koc …), contaminant threshold values used in the Walloon regulation (Walloon Soil Decree, 2008) and ecotoxicological thresholds.The “Pathway” module organizes the information needed to characterize the physical environment, such as geology, hydrogeology, topography, soils or land-use. All the physical and chemical properties of the environment related to these parts and influencing the behaviour and fate of contaminant during its way from source to receptor are also listed in this module (organic mater content, porosity, hydraulic conductivity …). The "Receptor" module gathers the information on potentially exposed receptors, which are first the groundwater resource, the saturate zone of the aquifer and the use that is made of it and that will determine the quality criteria and risk thresholds, but possibly also other receptors such as streams, pumping wells or springs.

At the regional scale, it is unlikely to obtain all the information on existing pollution sources because all the contaminated sites are not necessarily characterised in details or even known. To overcome this difficulty, the list of considered contaminant at the pollution sources can be established using as "activity-pollutant” matrix based on registered industrial activities. The "activity-pollutant” matrix is presented as two Microsoft Access-type linked tables connected with a shapefile listing all industrial plants. The first table is the dictionary of pollutants previously mentioned. The second table relates classified industrial activities and associated pollutants, based on previous similar works (e.g. MATE, 2000; SITErem, 2002). The “Industry” shapefile and the "activity-pollutant” matrix are linked using an activity code defined according to the European industrial activity classification NACE (European Commission, 2008) to create a shapefile of potential contaminated sources.

A quantitative module would also been integrated to the "activity-pollutant” matrix. Knowing the size of the industrial plant, this module can deliver the mass or concentration of related contaminants likely to be present in the groundwater. This relation is based on the probabilistic interpretation of the French database ADES which lists all the groundwater analyses around industrial plants in France, as required by the French authorities. This module constitutes a further step to classical "activity-pollutant” matrices by giving a probabilistic quantification of the released mass of contaminants or expected concentration in the saturated zone as a function of the industrial plant characteristics (size, production quantity, involved industrial processes …). This task has still to be completed after the end of the project.

2.2.2 Specific degradation constants

Regional Risk Assessment requires the investigation of pollutant behaviour over the whole groundwater body. The most important processes governing the fate of the contaminants are sorption and degradation. Sorption coefficient for different pollutants can be found in the dictionary of pollutant and been modelled using K_{OC} and fraction of organic matter. Degradation is a more specific parameter that can also be taken from literature in the worst case from degradation test under similar conditions as the ones prevailing at the site of interest. But degradation constant is very site-specific and has often to be determined by performing degradation test for the case study in question.

To determine degradation constants of different compounds, batch or column degradation tests can be performed. In these tests, a certain amount of porous medium is brought in contact with the groundwater. In the batch test, the degradation is studied under static conditions while the columns better represent the *in situ* conditions since they are running under dynamic conditions. The pollutant can be present in the groundwater or it can be spiked in a certain amount to the batch or column set-up. From the graphs representing the decrease in pollutant concentration in function of time, degradation constants can be obtained. The degradation potential in an aquifer is being determined by the physico-chemical parameters (like correct pH, redox potential EC, Dissolved Organic Carbon, concentration of electron acceptor, etc...). Different compounds can be added to these set-ups to create optimal physico-chemical conditions to stimulate the degradation of pollutant. For example, oxygen can be added to stimulate the degradation of e.g. benzene or Methyl Ter Butyl Ether, or an electron donor can be added to stimulate the degradation of chlorinated aliphatic hydrocarbons (CAH). Next to physicochemical conditions, also the number of bacteria has a big influence on the degradation potential present in an aquifer. Molecular techniques can be used to study, in the aquifer, the diversity and the number of bacteria able to degrade a pollutant. In the FRAC-WECO project Quantitative PCR has been used to determine the number of bacteria involved in the degradation of CAHs. These bacteria can be determined by quantifying their 16S rRNA gene (present but different in all bacteria) or degradation genes. The obtained kinetic parameters and biomass numbers can then be used in the reactive transport model for simulating the contaminant plume behaviour in the groundwater body. This model can then predict how much CAHs will reach the receptor under different site conditions such as natural attenuation (corresponding to *in situ* conditions) or after the addition of a factor stimulating the degradation of the pollutant.

2.3 Description of indicators and risk assessment concepts

2.3.1 Regional Risk Assessment for groundwater quality

Evaluation of groundwater quality at the regional scale and related pollution risk assessment are made complex because of two major aspects: the spatially distributed nature of the contamination and the integration of the effect of various pollutants of different nature, with specific physico-chemical properties and harmfulness for health or environment.

To evaluate objectively the overall quality of groundwater and its level of derioration, it is necessary to normalize the concentrations of contaminants using a uniform classification procedure with classes that consistently reflect equivalent levels of deterioration for the different contaminants considered (e.g. drinking water limit). To reach that objective, the SEQ-ESO indicator used by the Walloon Region of Belgium to report on the Water Framework Directive groundwater quality monitoring network (Rentier et al., 2004) has been selected and adapted. It is based on the SEQ-Eau ranking system initially developed by the French Water agencies (Agences de l'Eau, 2002). The SEQ-ESO provides an interpretation grid for a complete protocol analysis related to a single water sampling point. Conversion from contaminant concentration to a normalised non-dimensional index is based on interpolation functions between different threshold values that depend on water uses. The final quality index, ranging from 100 (good quality) to 0 (very degraded) corresponds to the index of the most problematic compound (Rentier et al., 2006). The calculation the SEQ-ESO quality index is performed in two steps. First, contaminant concentrations are normalized on a [0 = poor, 100= good] scale considering different contaminant specific threshold values, as illustrated for benzene in Figure 2. The threshold values are defined for each kind of contaminant with respect to different water uses. Three different water uses have been defined originally for the SEQ-ESO: patrimonial status (PS), drinking water use (DWU) and, biological impact on water courses (BIO). A global groundwater quality status, called SEQW, is also evaluated using a combination of PS and DWU thresholds (see example for benzene in TABLE I). Four threshold values are defined for each pollutant with linear interpolation for the three first intervals (S0-S1, S1-S2 and S2-S3), antilogarithmic interpolation for the S3-S4 interval and negative exponential interpolation above the S4 threshold. In a second step, the quality index is set equal to the lowest quality index corresponding to the most degrading contaminant. In the regional risk assessment approach described here, the SEQ-ESO threshold values have been incorporated into the pollutant library of the database considering the different water uses (PS, DWU, BIO and SEQW). However, the SEQW indicator, which is assumed to better reflect the global quality of water, independently of any use, is effectively considered for the calculation of the regional indicator for groundwater quality.

TABLE I: Illustration of the combination of PS and DWU thresholds for the definition of SEQW threshold and origin of thresholds for benzene (example).

Benzene	Patrimonial Status (PS)		Drinking Water Use (DWU)		SEQW
µg/l	Threshold	Origin	Threshold	Origin	Threshold
S1	0.25	Reference value for natural groundwater quality including geochemical background.	0.5	Guidance value of the 80/778/CEE EU Directive concerning drinking water (CEU 1980).	0.25
S2	0.5	Linear interpolation between S1 and S3	1	Walloon drinking water standard	0.5
S3	0.75	Threshold value as defined in the Walloon Soil Decree		*Does not exist*	1
S4	4	Intervention value for cleaning as defined in the Walloon Soil Decree	10	Guidance value of the 75/440/CEE EU Directive concerning quality water before treatment of surface abstracted for drinking water (CEU 1975).	10

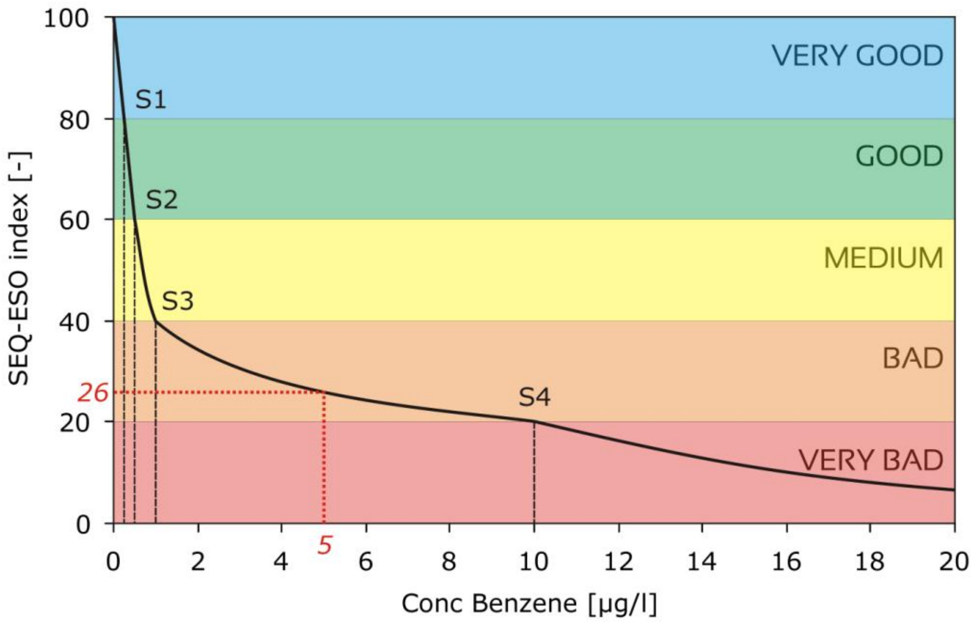


Figure 2: Normalization from benzene concentration in groundwater to groundwater quality index according to SEQ-ESO method and SEQW thresholds (for example, groundwater contaminated at 5µg/l of benzene corresponds to a quality index of 26.

The SEQ-ESO threshold values for each pollutant and for different water uses have been incorporated into the pollutant library of the database and a specific SEQ-ESO module has been developed in the ArcMAP environment for calculation of the regional indicator of groundwater quality based on the SEQ-ESO methodology.

Results imported from the groundwater flow and transport simulations provide concentrations for each contaminant in each grid cell and at each time step. The SEQ-ESO procedure is applied in each cell of the grid to normalize the concentrations of contaminants and to convert them into groundwater quality indexes by considering the specific threshold values of each contaminant. This provides, at each time step, a map of distributed groundwater quality indexes at the level of the grid. Finally, an overall indicator can be obtained at each time step, by spatial aggregation of the grid indicators weighted by the volume of groundwater present in each cell (Equation 1).

$$I_{global} = \frac{\sum_i I_i V_i}{V_{GW}} \qquad \text{(Equation 1)}$$

Where I_{global} is the global quality index for the whole groundwater body at time t [-], V_{GW} the volume of groundwater comprised in the zone where the risk is assessed [L^3], V_i the volume of groundwater into the cell i [L^3] and I_i the quality index for the cell i[-].

The results of the SEQ-ESO application can be used in different ways. The maps of indicators can be used to identify most problematic sectors (hot contamination spots) with heavily contaminated groundwater volumes within a more generalized contamination. At the grid cell level, one can examine the global evolution with time of groundwater quality. Finally, the aggregated indicator can be used to report on the groundwater quality status of the groundwater body and for groundwater quality trend assessment.

2.4 Water and contaminant fluxes through the catchment

The objective of this project is to study the impact of contaminated sites on the receiving water bodies being the river basin or the groundwater body. Since water is often the main vector of the mobility of contaminants, an accurate description of surface and subsurface water fluxes in the contaminated sites and water bodies is required. A modelling approach is developed for calculating accurately the water and contaminant fluxes on the contaminated sites, with the aim of describing the state and dynamics of the 'water-soil-subsoil-sediment' system. This approach used to determine groundwater recharge is then combined with a groundwater flow and transport model to route the contaminants through the groundwater body and calculated the extent of the contamination plumes and chemical status of the groundwater body.

2.4.1 High-resolution groundwater recharge simulation and surface run-off routing

Groundwater recharge, defined as the water arriving at the water table, is a key parameter determining the input towards a groundwater body. In urban and industrial areas, the sources and pathways for groundwater recharge are more numerous and complex than in rural environments (Lerner, 2002). Buildings, roads, and other surface infrastructure make the surface impervious or less permeable for water to infiltrate, resulting in higher run-off rates. An important factor determining this spatial and temporal variation in infiltration and groundwater recharge is the land cover. While information on land use and land cover might be extracted from existing data sets, like the land use map of Flanders or the CORINE land cover database, the problem with these data sets is that they are too strongly generalized, both spatially and thematically, to be useful for hydrological modeling applications within urbanized/industrialized watersheds.

In order to be able to describe the state and dynamics of the 'water-soil-subsoil-sediment' system we created a land-cover map of the study areas using high resolution satellite imagery to obtain enough detail from a hydrological point of view. Traditional land cover classification can not be used at regional scale because of the lower resolution of satellite imageries. A stratified classification approach has been adopted. The first step, the satellite image is classified on pixel level to distinguish land-cover classes and separate urban and natural pixel. Secondly the urban pixels are characterized at sub-pixel level to determine fraction of imperviousness and natural pixels are combined with administrative landuse maps. Finally, the result is a detailed map providing relevant hydrological classes and soil imperviousness.

The created map can be directly imported or interpolated under grid format and used as an input for runoff and groundwater recharge modeling. Groundwater recharge is varying in space and time, but is not directly measurable from precipitation. No fixed percentage of rainfall is infiltrating into the soil. Development of GIS, remote sensing, spatially variable land cover and soil data leads to an approach that directly takes into account the influence of the spatial variability of soil texture, land cover, slope and meteorological conditions in groundwater recharge estimation, and improves the spatial estimation of recharge.

The methodology followed for simulation of the recharge and run-off in this study is based on the distributed seasonal WetSpass model (Batelaan and De Smedt, 2007).

2.4.2 GW flow and transport modelling at the scale of the groundwater body

In order to evaluate the impact of pollution sources on the quality of the groundwater resource, it is necessary to evaluate, at regional scale, how much and to which extent groundwater is deteriorated from these sources. This requires determining the volume of deteriorated groundwater and the severity of this deterioration, *i.e.* the spatial extent of the contaminated plumes and concentrations of contaminants. Existing monitoring networks, usually based on piezometers, are unlikely to provide extensive and exhaustive information on groundwater pollution because they are very often limited to the immediate neighbourhood of identified polluted sites. The alternative is to use groundwater flow and transport models to calculate, based on reasonable assumptions, contaminant leaching trough the unsaturated zone and spreading of contaminant plumes in the saturated zone from the selected pollution sources. The objectives of the groundwater flow and transport model is first to calculate the dispersion plumes of contaminants in the saturated zone, but also to evaluate the relative importance of specific pollutant sources within a regional contamination (*i.e.* site prioritization) and to test programmes of measures defined for the restoration of groundwater quality for groundwater body management.

A comprehensive modelling approach requires calculating first the vertical leaching of contaminants from the pollution source to groundwater table, then the horizontal spreading of contaminants in the saturated zone. However, the pollution sources considered correspond mostly to historical contaminations having occurred when regulations were less restrictive. Accordingly, it is assumed, as a first approximation, that contaminants have already reached the saturated zone and that the leaching of contaminants to this zone can be modelled as a specified mass flux considering the spatially distributed recharge rate and the effective solubility of each contaminant.

The groundwater flow and transport model consists of a transient transport model based on a steady-state groundwater flow model. It serves to calculate groundwater fluxes and groundwater flow directions in order to route contaminants from the pollution sources through the groundwater body and, subsequently, to run transport simulations. Steady state simulations for groundwater flow are considered as sufficient because the objective is to delineate contamination plumes over long time periods, and not to consider the dynamics of groundwater variations on these plumes. At this development stage, aquifer hydraulic conductivity is assumed to be homogeneous over all the groundwater body. This can be assumed since, as a statement, groundwater body gathers lithological entities of same hydrogeological properties and that local variation can not be considered at a regional scale.

Practically, the MODFLOW (Harbaugh et al., 2000) and MT3D (Zheng and Wang, 1999) software are used. Numerical simulations are performed under GMS environment. Data and information exchanges between the modelling application and the regional risk assessment system are managed through specific communication modules developed in the GIS interface. The communication procedure is described in Figure 3. The different shapefiles required for the hydrogeological conceptual model are prepared within the GIS system using data from the geodatabase and imported in GMS (Fig. 3a) using the ‘GIS and Map Module’. Based on that, a regional finite differences grid is created and exported back to the GIS interface (Fig. 3b) where it is used to clip the pollution sources and related information (contaminant types and properties …) to be again further exported in the appropriate GMS grid format (Fig. 3c) for the contaminant transport simulations (Fig. 3d). Currently, the considered contaminant transport processes are advection, hydrodynamic dispersion, linear sorption and degradation (Equation 1).

$$R\frac{\partial C}{\partial t} = div(\underline{\underline{D}}\,\underline{gradC} - \underline{v_e}C) - \lambda RC + C^*\frac{q'}{n_e} \quad \text{(Equation 2)}$$

Where C is the concentration [M/L³], t the time [T], D the hydraulic dispersion [L²/T], v_e the effective velocity of water [L/T], λ the first order degradation kinetic constant, q' the incoming water flux [T^{-1}], C the concentration is the incoming water flux [M/L³] and equal to C if q'<0, equal to Cin if q'>0, n_e: the effective porosity, R the retardation factor equal to $1+\frac{\rho_b K_d}{n_e}$ with ρ_b the bulk density of the porous media [M/L³] and K_d the solute distribution coefficient between aqueous and solid phase [L³/M].*

Once simulations are performed, modelling results (piezometric heads and contaminant concentrations at different time steps, one data set per contaminant) are imported into the GIS interface (Fig. 3e) for the calculation of the groundwater quality indicator (Fig. 3f), as described in the next section.

2.5 Descritpion of the Ecotox Risk Assessment methodology

To assess the ecotoxicological risk at a more local scale, a conceptual groundwater risk assessment approach adapted from the original TRIAD approach (Long and Chapman, 1985), applied for sediment quality assessment has been developed. This approach, referred to as "Groundwater Quality TRIAD-like (GwQT)", combines chemical data with laboratory bioassays and key physico-chemical variables to evaluate the impacts of pollutants on selected organisms. The overall study of these three components provides an assessment of the environmental risk. To our knowledge, this study presents the first application of the TRIAD approach on the groundwater system. This new concept is a starting point for groundwater characterization and it is open for improvement and adjustment.

It has been validated at the local scale (Chimeuse and Morlanwelz sites) because more data are required at theregional scale.

The Toxic Unit approach (TU) has been used in combination with the TRIAD-like approach to identify micro-pollutants which contributed to effects observed in the bioassays.

2.5.1 Groundwater Quality TRIAD-like approach

The "Groundwater Quality TRIAD-like" (GwQT) approach is based on measurements of chemical concentrations (chemistry), laboratory toxicity tests (ecotoxicity) and physico-chemical analyses (pH, conductivity…). These components are combined in the GwQT using qualitative and quantitative integration approaches. The advantage of the qualitative methods is the simplicity of interpretation, although no quantification is provided. The TRIAD-like approach is explained in detail in Deliverable D.1.5 (Crévecoeur & Thomé, 2010a). It will allow the classification of sites into categories according to the degree of contaminant-induced deterioration.

The first component of the TRIAD is environmental chemistry. Specifically, identification and quantification of contaminants present in the groundwater samples must be undertaken.

Total concentration of pollutants is integrated into risk index (Chemical risk index, ChemRI) by using a sequence of successive steps (Jensen and Mesman, 2006). Concentrations of pollutants are compared to normative value and a risk number (between 1 and 0, the lower the score, the better the quality) is assessed for each class of pollutants and globally for each sampling point using Independent Action (IA) model.

All parameters (conductivity, pH, nutrients, …) used for water characterization are generally not integrated in the classical TRIAD approach. However, the physico-chemical component is part of GwQT as these parameters are critical to the survival and growth of aquatic life. Physico-chemical Risk Index (PhysRI) is assessed by applying the same sequence of successive steps as for ChemRI.

Application of biotests with different organisms can indicate general stress as a result of complex mixtures, and provide information about potential hazard to aquatic life. An Ecotoxicological Risk Index (EtoxRI) is assessed by dividing the results from bioassays (*e.g.* mortality in the pure sample), expressed as a percentage, by 100 (Jensen and Mesman, 2006). A global EtoxRI is estimated as the mean of the different RIs (Dagnino et al., 2008).

The quantitative integration method integrates all information in a single numerical value, the Environmental Risk Index (EnvRI). The difference between the final score and the theoretical score 1 gives an indication of the risks (and its magnitude) for ecosystem health at the studied site (Den Besten et al., 1995). In the integration procedures, arbitrarily assigned weighting factors (wf) are applied to the diverse indices on the basis of their ecosystem relevance (Dagnino et al., 2008). Chemistry is the more direct measure, but toxicity is given equal weight because of the possibility that unmeasured chemicals are present (Bay and Weisberg, 2008). The physico-chemical component is also an important parameter. In this study, unequal weighting among components was used (wf =1). The Environmental Risk Index is calculated using the following equation (adapted from Dagnino et al., 2008):

$$EnvRI = \frac{(wf_{CHEM} * ChemRI) + (wf_{PHYS} * PhysRI) + (wf_{ETOX} * EtoxRI)}{(wf_{CHEM} + wf_{PHYS} + wf_{ETOX})} \quad \text{(Equation 3)}$$

Qualitative methods have often been applied to determine the degree of deterioration of each studied site. Therefore, contamination, physico-chemistry and toxicity data are combined in the TRIAD-like approach using tabular matrix, pie charts and triangular representation.

In tabular matrix, the different responses are shown with positive or negative signs to indicate a possible risk (Chapman, 1990).
Different levels of probability in the similarities between stations can be represented using pie charts. It represents the integration of all the components using colours coming from the SEQ-ESO system (DGARNE, 2010) and risk indicators (TABLE II).

TABLE II: Representation of TRIAD-like components in classical pie-charts and risk classification according to SEQ-ESO colours (PE, Percentage of Effect; RI, Risk Indice), adapted from DGARNE (2010) and Persoone et al. (2003).

ChemRI & PhysRI	EtoxRI	Risk Indicator
$0.00 < RI \leq 0.20$	$PE \leq 10\%$	No risk (- -)
$0.20 < RI \leq 0.40$	$10\% < PE < 50\%$	Low risk (-)
$0.40 < RI \leq 0.60$	$50\% \leq PE < 100\%$	Moderate risk (±)
$0.60 < RI \leq 0.80$	PE = 100% (at least 1 test)	Relatively high risk (+)
$0.80 < RI \leq 1.0$	PE = 100% (all tests)	High risk (+ +)

The three indices can be displayed in graphical form as three segments (for Contamination, Toxicity and Physico-chemistry) with a common origin (“0”, no risk), where the lengths of each segment equals the values of the risk index for the three groups of determined parameters (Chapman, 1990).

The different methodologies used in the representation and interpretation of the TRIAD-like results have been shown to be complementary.

2.5.2 Toxic Unit approach

The Toxic Unit approach (TU) was used to identify micro-pollutants which contributed to effects observed in the bioassays.
This concept can be mathematically expressed as the following equation:

$$TU_i = C_i / L(E)C_{50,i}$$ (Equation 4)

Where C_i gives the concentration of the component i in a mixture which elicits 50% of effect, and $EC_{50,i}$ denotes the concentration of that substance which induces 50% of effect if applied singly (collected from literature and ecotox databases).

Assuming that the effects of pollutants with a similar mode of action are mainly additives, individual Toxic Unit values were added to estimate the toxicity of groups of compounds such as ΣTU values for metals, PAHs or organochlorine compounds (Lahr & al, 2003). In one approach to characterize joint effects, the sum of all TU_i can be used. $\sum TU_i$ values < 1, = 1, or > 1 were taken as proof of synergism (‘‘greater than additive”), simple additivity or antagonism (‘‘less than additive”), respectively (De Laender & al, 2009).

2.6 Socio-economic analysis

2.6.1 Applying the Water Framework Directive in the case of groundwater bodies contaminated by brownfields.

Following the requirements of the Water Framework Directive, a programme of measures selected on the basis of a cost-effectiveness analysis (CEA) should be proposed for each water body at risk of not reaching good status. In case of disproportionate costs derogation may be justified on the basis of a cost-benefit analysis (CBA).

This economic analysis is a challenging task in case of groundwater bodies located in heavily industrialised and urbanized area and historically contaminated by brownfields. Remediation measures are expected to be very expensive and time required for remediation may be very long. As groundwater resources are generally not used anymore as a resource it is difficult to anticipate (i) if market benefits could be expected from an improvement of the groundwater quality and (ii) if people would be willing to contribute to this quality improvement.

Changes in the state of the aquatic system may lead to positive impacts (benefits) or negative impacts (damage). Impacts are characterised as the change of goods and services provided by aquatic systems that lead to change in human welfare. The concept of total economic value (TEV) can be used to provide a measure of the economic value of an environmental asset (Pearce *et al.*, 2006). Any improvement in water status (for instance after the implementation of remediation measures) leads to *environmental benefits* (positive impacts) that may be defined as the TEV of the physical environmental improvement of the water system. The TEV of groundwater is calculated as the sum of use values (including direct and indirect use values) and non-use values (Brouwer, 2006).

Use values are associated with the direct or indirect, current or potential future use of the water resource:

- *Direct use values* involve human interaction with the resource itself. It may be consumptive or extractive use (e.g. domestic, public, commercial, agricultural and industrial supply, fisheries). Direct use value leads to activities that would not take place if that quantity and quality of water did not exist.
- *Indirect use values* derive from the ecological services provided by the groundwater resource as a support of living organisms or as water discharge to connected surface waters and associated ecosystems (*ecosystem benefits*).
- *Option values* represent the amount which certain users may be willing to pay to secure access to and use of groundwater at some time in the future and include the value stemming from future use of groundwater.

Non-use values are not related to any actual or potential future use but refer to values attached to the water resource conservation based on considerations that, for instance, the resource should be preserved for future generations.

Classical CBA often take into account only direct use values to assess the low range of benefits of groundwater quality improvement. These values may be obtained with the implementation of market-based methods (e.g. averting behaviour, replacement costs). However, in the case of groundwater currently contaminated and not much used, direct use values are expected to represent a small part of the TEV. Contingent valuation is the unique method enabling to integrate both use and non-use values in the TEV. However, its application to groundwater resource improvement remains scarce, especially in the case of groundwater resources with low expected direct use values.

2.6.2 Valuing the benefits

Classical Cost-benefit analysis often takes into account only direct use values to assess the low range of benefits of groundwater quality improvement. These values may be obtained with the implementation of market-based methods (e.g. averting behaviour, replacement costs). However, in the case of groundwater currently contaminated and not much used, direct use values are expected to represent a small part of the Total Economic Value (TEV). Contingent valuation is the unique method enabling to integrate both use and non-use values in the TEV. However, its application to groundwater resource improvement remains scarce, especially in the case of groundwater resources with low expected direct use values.

The general methodology proposed by the FRAC-WECO project is a valuation of market benefits through the averting behaviour approach and a valuation of the TEV expected from a groundwater quality improvement through a the implementation of the contingent valuation method.

Direct use/ option use values

Market benefits may occur to current and future potential water users i.e. those that could be impacted (positively or negatively) by a change in the groundwater quality. Types of water users concerned are those that are sensitive to water quality and more precisely that are sensitive to industrial micropollution, at least for one of their uses of water. Using the averting behaviour approach (Abdalla, 1994; Rinaudo et al., 2005), adaptation strategies that water users may undertake in case of groundwater quality improvement have been simulated.

Three main types of adaptations of the water supply strategies in case of groundwater quality improvement have been simulated and assessed:

- Water treatment may be reduced or abandoned (adaptation n°1).
- Water users currently supplied by the public network may be interested to turn to private groundwater supply (adaptation n°2).
- Water users with private wells may be interested to increase their use of the groundwater under study (adaptation n°3).

The understanding of the water supply strategies in place in the case study areas was made possible by the use of databases provided by the DGRNE and the CILE and by experts' consultation (representatives of drinking water sector and economic activities). Concerning the public water sector, data were gathered at the public water supply area (PWSA) scale.

Potential market benefits to be expected from an improvement of the RWM 073 groundwater quality to a 'good water status' may reach 3.3 to 4.4 million € per year from 2030 (TABLE III).

As these benefits are expected to appear from 2030, discounting can be used to express these future benefits and compare them to the cost that would be required today to implement a programme of measures. The discounted benefit expected for year 2030 is estimated between 1.3 and 1.8 million € (TABLE III). The sum of discounted benefits on the 2008-2060 period is estimated between 23.8 and 32.4 million € (TABLE III).

Benefits related to the effects 1 and 2 (19 million € on the 2008-2060 period) are expected to occur for the companies that currently use or may use the RWM073 as a resource i.e. located on the RWM073 aquifer. Benefits related to the effect 3 are expected to be redistributed to public water consumers at the sub-basin level through the water bill: from 19.4 to 26.4 million € for the domestic sector i.e. households (81%) and from 4.5 to 6.2 million € for the non-domestic sector (19%) on the 2008-2060 period.

TABLE III: Synthesis of expected benefits per effect

			Annual benefits (M€/year, from 2030)			**Discounted**(2) annual benefit in 2030 (M€/year)	Sum of **discounted**(2) benefits (M€, 2008-2060)
			Expected "direct impact" (1)	Expected "indirect impact" (1)	Expected annual benefit		
①	Decrease or abandonment of water treatment		0.7	n.a.	0.7	0.3	4.8
②	Turning from public network supply to private supply		2.1	-0.2	1.9	0.8	14.2
③	Increase in the use of RWM073 as a resource	Low increase	2.1	-1.4	0.7	0.2	4.7
		High increase	6.9	-5.2	1.8	0.7	13.4
① + ② + ③		Low	**4.9**	**-1.6**	**3.3**	**1.3**	**23.8**
		High	**9.7**	**-5.4**	**4.4**	**1.8**	**32.4**

(1) in reference to Figure 7

(2) discount rate r=4%

Total economic value

A contingent valuation was developed on the Meuse alluvial aquifer (between Engis and Herstal, namely the RWM073 groundwater body). This method consists in implementing a quantitative survey on the perception by the population of the Meuse alluvial plain. The aim is: (i) to characterise the knowledge and perception by the population of the groundwater body, its quality level, the relevance to improve its quality and (ii) to assess the value people would be willing to contribute to the improvement of the groundwater body quality.

The questionnaire is structured into four main sections, with a total of 33 questions (for more details, please refer to Annex 7): (i) Section 1 presents the groundwater body under study, (ii) Section 2 summarizes the groundwater quality problem today and in the future if no action is undertaken, (iii) Section 3 presents a scenario of groundwater quality improvement, (iv) Section 4 deals with socio-economic characteristics of the respondents.

The questionnaire was tested from the 9th to the 11th June 2010 by three BRGM researchers. 56 face-to-face interviews were completed in the cities of Liège, Flémalle, Amay and Seraing. The sampling area is composed of 11 communes located at least partly on the RWM073 groundwater body and those partly located on the RWM072 groundwater body at a maximum distance of 5 km upstream and downstream the groundwater body under study. A team of 6 students and 4 engineers/ researchers was constituted at the beginning of September. A half day training session was organised at the University of Liege the 9th September 2010. Quotas were defined by age and sex.

The questionnaire was administered from the 13th to the 17th September 2010 in the communes of Liège, Engis, Amay, Visé, Oupeye, Herstal and Seraing. 531 face to face interviews were completed.

98% of the respondents consider it is important to improve the RWM073 groundwater quality and 66% would be willing to contribute financially to this improvement by an increase in their water bill (Figure 3). The average willingness to pay (WTP) is 46.9 €/household/year when only positive WTP are considered. The average WTP is 40.1 €/household/year when true zeros are included. The Figure 4: Mean positive Willingness To Pay observed by former commune (old boundaries)Figure 4 depicted the geographical distribution of the mean positive WTP by former commune (old boundaries[1]).

A multivariate analysis is currently carried out to identify the factors explaining the variations in the stated willingness to pay. The statistical analysis is split into two steps. We first model the decision to participate financially to the scenario with logistic regression. We then conduct a series of OLS regressions to identify the factors explaining the variations of the stated WTP amount. First results of the logistic regression model show that location of the respondent, realism and importance levels attached to the scenario, perception of the risks related to groundwater quality deterioration, types of groundwater use, exposition to environmental problems and socio-economic characteristics (gender, children, education, income) are significant factors explaining the decision to contribute to the scenario. First results of the OLS regression models show that location of the respondent, perception of the risks related to groundwater quality deterioration, number of benefits attached to the scenario, main types of benefits attached to the scenario, exposition to environmental problems, socio-economic characteristics (gender, children, education, income) and difficulty to answer to the WTP question are significant factors explaining the variations in the stated WTP amount.

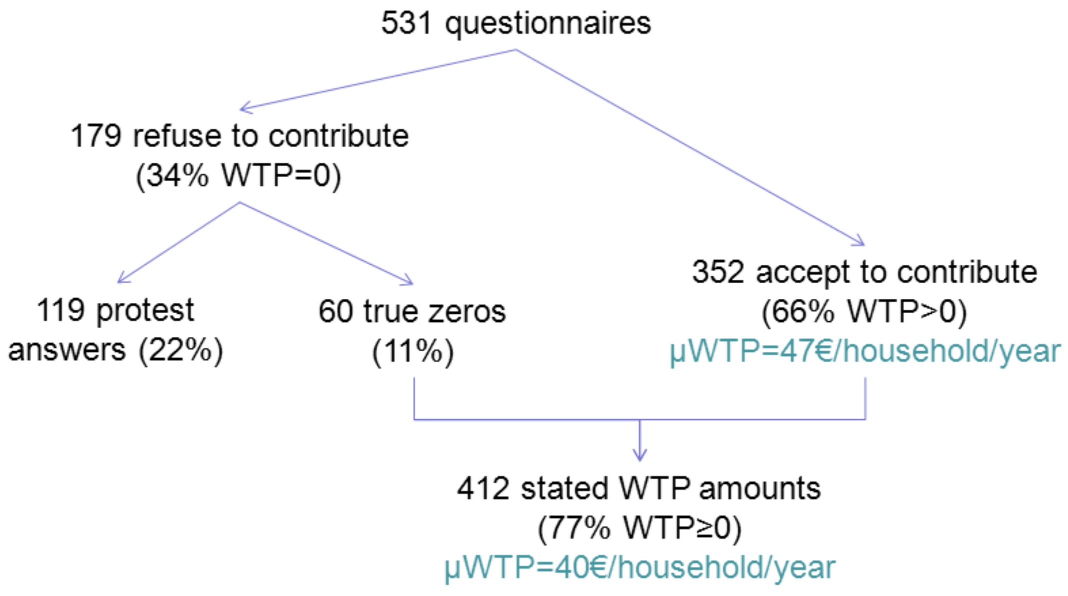


Figure 3: Proportion of different types of answers

[1] From 1961 to 1983 the number of Belgian communes has been reduced from 2663 to 589. In this report we use the term "former commune" to refer to a commune of 1961 and the term "commune" to refer to a commune of 1983.

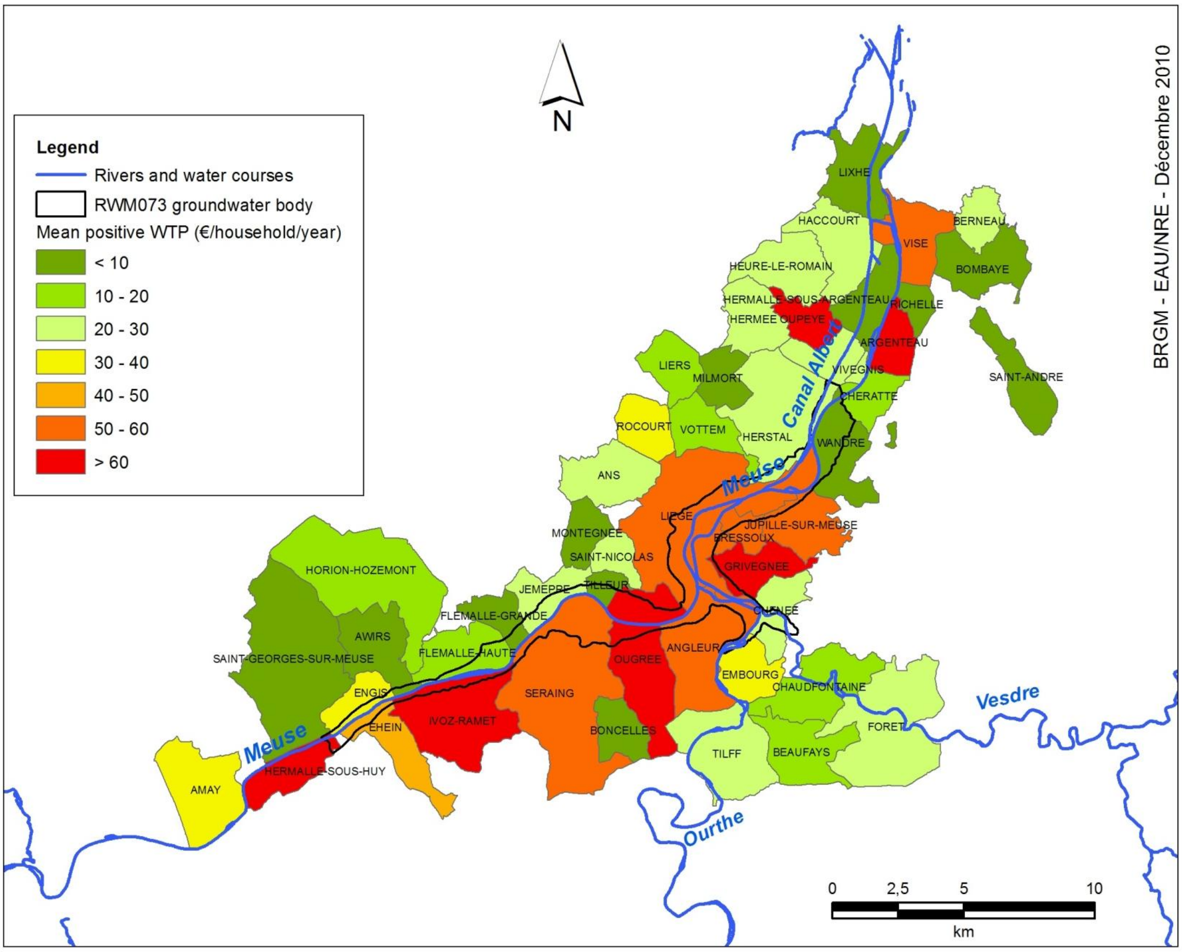


Figure 4: Mean positive Willingness To Pay observed by former commune (old boundaries) around the RWM073 groundwater body.

2.7 Applications

The Regional Risk Assessment methodology has been applied on the RWM073 groundwater body in Walloon Region (Belgium). But before this complete application on a real case, various tests have been performed on numerical synthetic test cases for the modeling concepts and on local contaminated sites for the rest of the RRA methodology. The different test cases are described hereafter to illustrate the applicability of the RRA.

2.7.1 Application of the RRA methodology on synthetic test cases

The simplified example reproduces the relatively frequent case of pollution of an alluvial aquifer by industrial contaminants emitted from an industrial plant located nearby a river. The studied domain corresponds to a polluted area of 500 by 400 meters. The alluvial aquifer has a thickness of 15 meters The mean hydraulic conductivity of the alluvial sediments is 1×10^{-5} m/s. Groundwater is recharged by infiltration from rainfall (135 mm/y = 4.3×10^{-9} m/s). No-flow boundaries conditions are assumed at the external lateral boundaries (north, south, west) of the model, except at the riverbank (east) where a draining river boundary condition is defined (river stage: 55 m, hydraulic conductance of the riverbed: 2.5×10^{-4} m²/s). The pollution sources defined at three different locations in the contaminated land parcel are assumed to emit three different pollutants with contrasted physico-chemical properties representative of common contaminants in such contexts: a PAH-like pollutant (low mobility, moderate degradation), a BTEX-like pollutant (high mobility, high degradation) and a VOCl-like pollutant (high mobility, low degradation) (TABLE IV). Contaminants are assumed to leach into the aquifer at their maximum solubility in the recharge water for a period of 15 years, after which the sources are assumed to be removed and contaminant leaching is stopped. The contaminant transport model is run for 45 years from the beginning of the leaching of contaminants.

TABLE IV: Contaminants parameters used for the synthetic test case.

Parameters	Units	Benzene-like species	PAH-like species	VOCl-like species
Concentration in recharge	mg/l	1830	10	1100
Kd	m³/mg	4.15×10^{-11}	4.77×10^{-7}	3.50×10^{-11}
Degradation	s^{-1}	1×10^{-7}	1×10^{-9}	1×10^{-9}

Figure 5a shows the three contaminants plumes in terms of concentrations after 15 years. As expected, these plumes reflect the specific properties of the contaminants (solubility, sorptivity and degradability). PAHs are almost immobile and do not spread into groundwater. VOCL are very mobile and BTEX are mobile but their plume extent is reduced because of degradation processes. However, these pictures do not give any clear insight on the actual deterioration level of groundwater in the area. Figure 5b shows the SEQ-ESO map of indicators for the three contaminants concentrations scaled into in a single groundwater quality index. This shows that most of the groundwater is heavily contaminated in the plumes. Finally, Figure 5c shows the evolution of the spatially aggregated groundwater quality index for each contaminant considered separately and the global SEQ-ESO indicator for the whole aquifer. During the period of contaminant leaching, groundwater quality is progressively degraded with time, the VOCL contaminant being the most significant contributor (approximately 40%) to this deterioration. The BTEX contaminant contributes to a subsequent deterioration of groundwater (approx. 10%) during the first 25 years. After 25 years, the contribution of BTEX to groundwater quality deterioration becomes negligible because of the progressive removal of the remaining quantity of BTEX by biodegradation in contrast with VOCL that does not degrade and keep a strong impact on groundwater quality after 45 years. The PAH contaminant is very sorptive and insoluble and it remains captured in the soil near the source from where it never effectively contaminates groundwater.

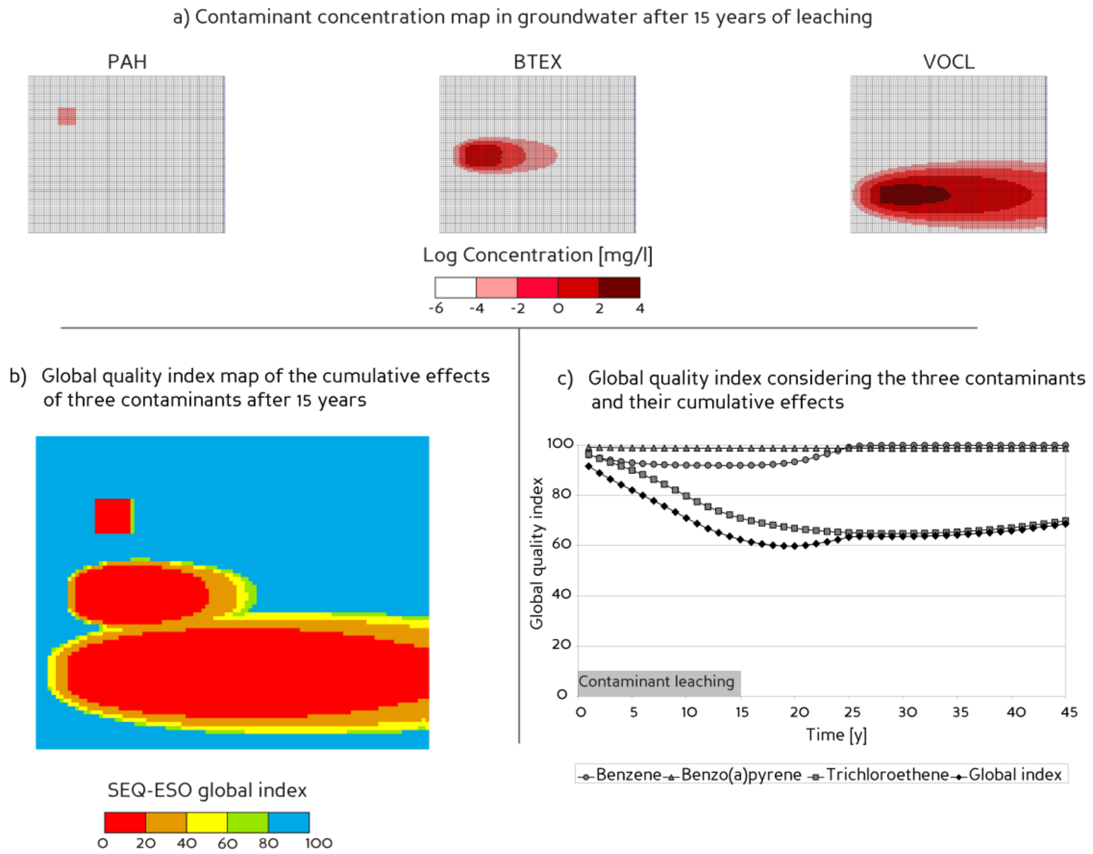


Figure 5: (a) Contaminants concentration after 15 year, (b) map of groundwater quality index after 15 years and (c) evolution with time of the global quality index using SEQ-ESO after spatial integration.

2.7.2 Landuse-recharge mapping at Vilvoorde-Zenne site.

In the first phase of the project a modeling approach was developed and tested on the Vilvoorde site. In order to produce detailed land-cover information for the study site, a land-cover map has been produced from very high-resolution (VHR) imagery (Ikonos, 05/08/2003 and 04/09/2003), with a 1-meter resolution for the panchromatic band and 4-meter resolution for the multispectral bands (blue, green, red and near infrared). Because high-resolution sensors have a limited spectral resolution, they do not allow distinguishing well between some types of land cover that are important in the context of hydrological modeling, based on spectral information only. To obtain accurate information on land cover an object-oriented classification approach has therefore been adopted. This approach allows using next to spectral information, geometric, textural and contextual information, defined at the level of homogeneous image objects. To assign the image objects to one of the selected land-cover classes a multiple layer perceptron (MLP) approach was adopted, using the NeuralWorks Predict software. MLP classification is a preferred technique for non-parametric classification, because next to spectral data, other kinds of data with non-normal class frequency distributions, like for example textural data, can be used as inout for the classification process. For training the MLP model, 100 samples were randomly selected for each land-cover class based on visual interpretation of the imagery. In order to evaluate the accuracy of the classification model it was validated on an independent validation set containing about 50 randomly samples image objects per land-cover class. After the application of some context-based post-classification rules we could enhance the accuracy of the classification and obtain a percentage of correctly classified image objects of 88% and a kappa index-of-agreement of 0.86. Figure 6 shows the final land-cover map (*Deliverable 2.1*), which was used as input for runoff and groundwater recharge modelling.

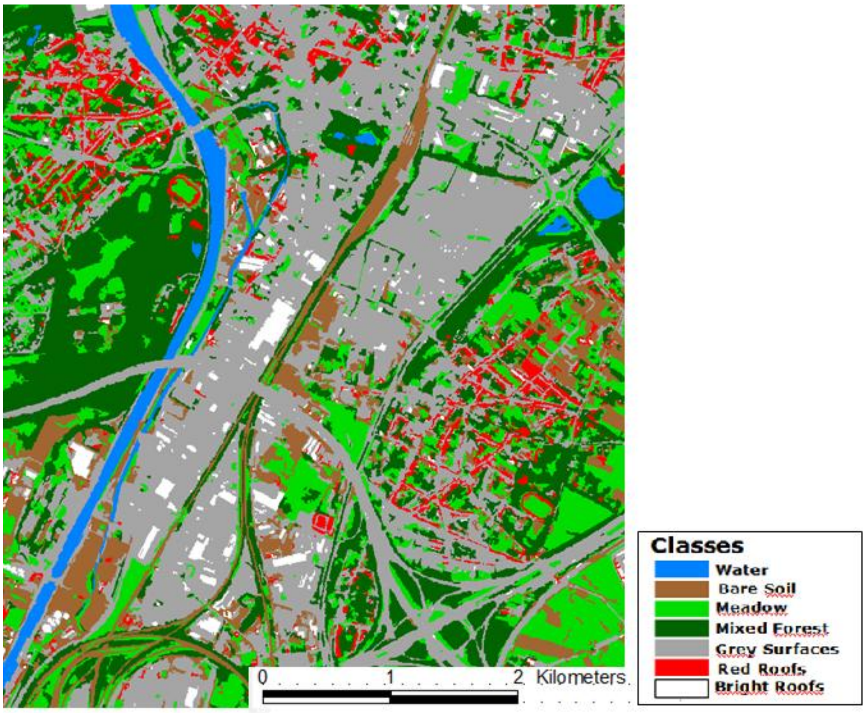


Figure 6: Land-cover map obtained for the Vilvoorde study site

The surface water budgeting was simulated with the spatially distributed WetSpass model. WetSpass estimates spatially distributed run-off, evapotranspiration and recharge in function of land-cover, soil texture and topography on seasonal basis. The model is fully integrated with GIS Arcview. Parameters such as land cover and related soil texture are connected to the model as attribute tables of the land use and soil raster maps. The resulting recharge is presented in Figure 7.

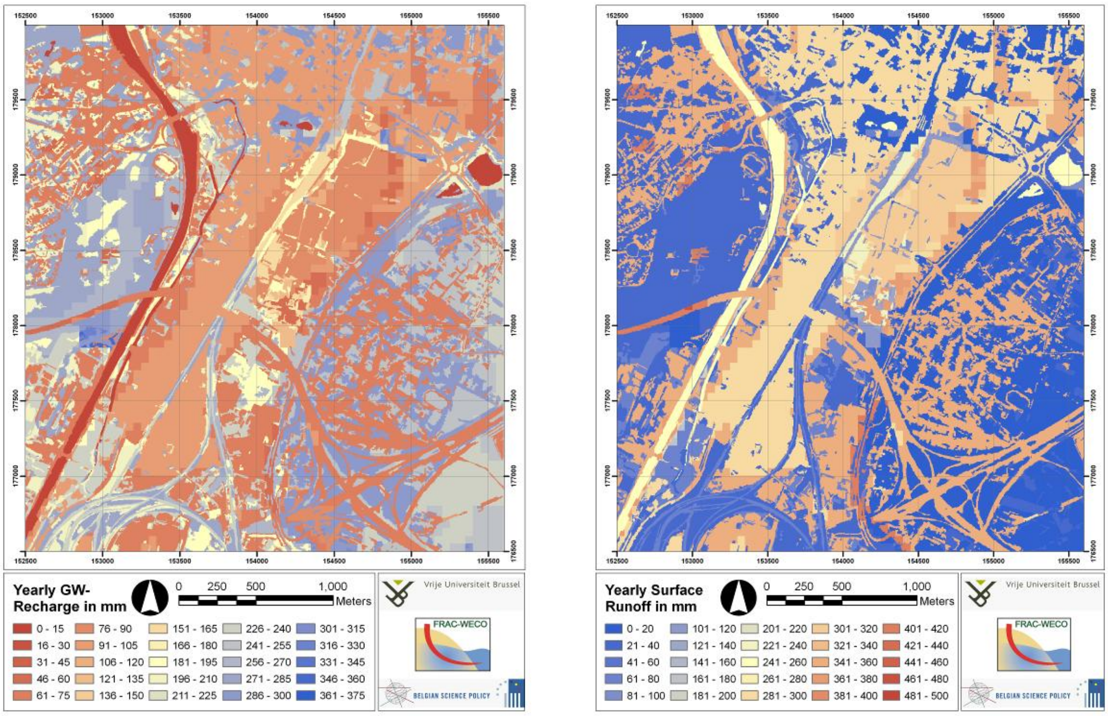


Figure 7: WetSpass simulated yearly long-term average recharge (left) and run-off (right) for the Vilvoorde study area.

The estimated yearly recharge ranges from about 0 to 350 mm/y, with an average value of 159±91 mm/y. The yearly surface run-off simulated with WetSpass ranges from about 2 to 467 mm/y, with an average value of 177±148 mm/y. From Figure 7 it is clear that the groundwater recharge and the surface run-off depend strongly on land-cover. The highest recharge values occur in forest areas, followed by bare soil and grass. On the other hand the figure shows that the highest run-off values occur in built-up areas. It is clear that the different outputs of WetSpass simulation show a strong spatial distribution over the area and they clearly demonstrate the importance of having access to detailed information on land-cover types, especially on the spatial distribution of impervious surfaces, in order to obtain reliable estimates of groundwater recharge and surface run-off.

For the Vilvoorde study area an existing groundwater flow model of VITO has been adapted and calibrated (finite difference steady state Modflow model). For adapting the model it showed to be necessary to (re-)evaluate the local geology. Hence, the information from 68 drillings and 179 cone penetration tests were studied to refine the hydrogeology. Boundary conditions were improved and recharge was simulated with the WetSpass model using the land-cover map obtained in WP2. After calibration the model was used to simulate the groundwater fluxes to the Zenne River. Measurements of the temperatures in the river bottom sediments provided via heat flow simulation an independent estimate of the groundwater fluxes. These results show an outflow of 50-88 m^3/day per river cell of 50 by 50 m. Water balances have been established for the model area and particle tracking analyses have provided insight in the potential transport of pollutants from four different sources towards the Zenne.

More detailed information about the land-cover mapping, recharge estimation and groundwater modelling of the Vilvoorde site is available in Dujardin et al. (2011).

2.7.3 Study in batch and column set-ups of the degradation of Chlorinated Aliphatic Hydrocarbons in the aquifer at Vilvoorde-Zenne site.

At the Zenne site in Machelen-Vilvoorde, Belgium, a groundwater plume of approximately 1.2 km, contaminated with chlorinated aliphatic hydrocarbons, (CAHs), is flowing from the nearby aquifer into the River Zenne. The area is contaminated as a result of industrial activities. For chlorinated aliphatic hydrocarbons (CAHs) reductive dechlorination, performed by certain anaerobic bacteria in the aquifer compartment, is one of the most important removal mechanisms. Since CAH degradation can only occur in the presence of an electron donor creation of an *in situ* biological reactive zone, through the injection of a carbon source into the aquifer near the river, was chosen as a remediation approach for aquifer compartment remediation. To this end, during the FRAC-WECO project, different batch conditions were run to determine at lab-scale the most appropriate carbon source to stimulate the CAH degradation in aquifer material obtained from three locations at the Zenne site.
Experiments for selection of electron donors to stimulate CAH degradation in aquifer in batch set-ups were perfomed at lab scale. The specific purposes are to:

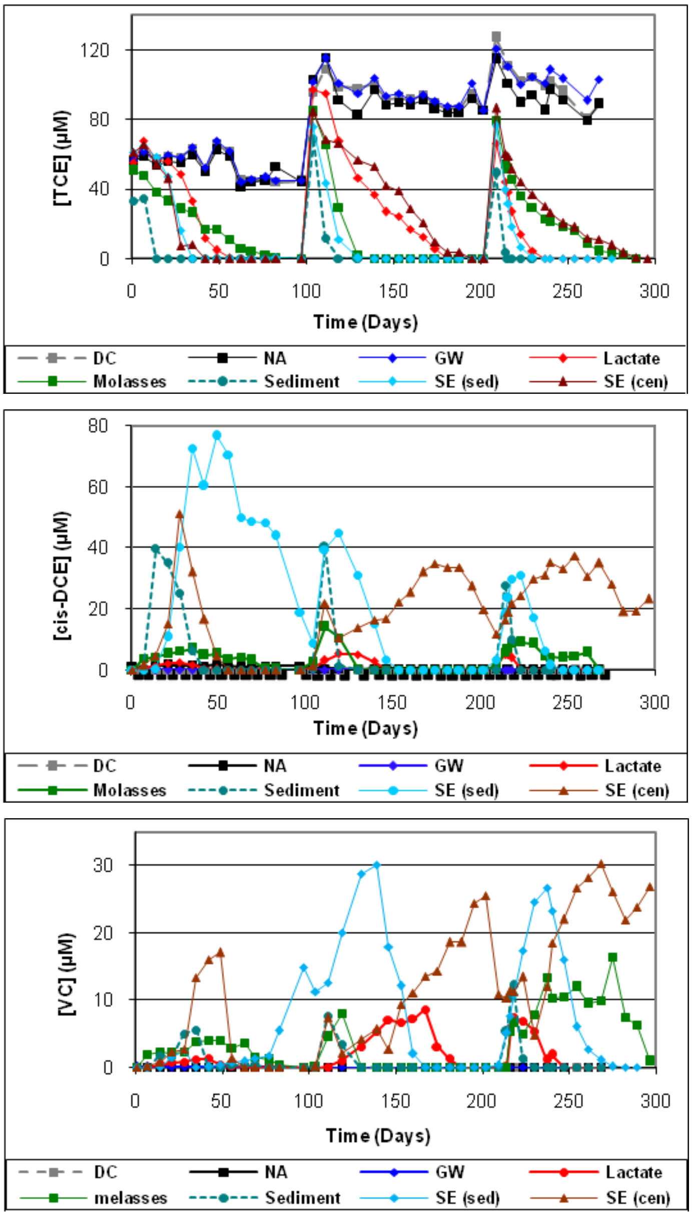
1- Determine kinetic parameters of the intrinsic and stimulated vinyl chloride (VC), cis-*cis*-1,2-dichloroethene (*c*-DCE), and thrichloroethene (TCE) reductive dechlorination activity in aquifer material by means of microcosm batch tests.

2- Determine the number of CAH-degrading bacteria and relevant catabolic genes that are present in the aquifer before and after the stimulation of CAH degradation.

The obtained parameters can then be used in the reactive transport model. This model can then predict how much CAHs will come into the receptor (the river Zenne at the studied site) under different stimulated and not-stimulated site conditions.

Batch microcosms were performed in order to investigate the VC, *c*-DCE and TCE biodegradation capacity of aquifer material and groundwater collected from three different locations in the test area (PB-26, SB-3 and SB-2) (Figure 8). Four different experimental conditions were set up in duplicate for each of the three locations: natural attenuation (corresponding to the in situ conditions), abiotic control (with killed microbial population to investigate sorption of the pollutant), sediment and (300 mg/L) lactate amendment. Four additional conditions were also tested for the PB-26 location: groundwater control and stimulation with (300 mg/L) molasses, sedimented extract of river sediment (to add DOC but also bacteria extracted from the sediment) and centrifuged sediment extract (to add only DOC from the sediment compartment). All bottles were incubated at 12 °C corresponding to the *in situ* groundwater temperature. Although tests were performed for the pollutants TCE, c-DCE and VC, results will only be presented for the TCE compound.

The results of TCE degradation and its daughter products c-DCE and VC to ethene at the location Pb26 at the Zenne site are shown in Figure 8. As it can be seen from Figure 8, no degradation was observed in un-amended cultures (see natural attenuation condition) and degradation happened only when external electron donors (lactate or molasses, Zenne River sediment, or an extract of the river sediment) were added to the microcosms. This shows that in order to tackle the CAH plume in the aquifer compartment, addition of external carbon sources is inevitable. Addition of river sediment showed the highest stimulation and resulted in a complete reductive dechlorination of initial 11 $mg \cdot L^{-1}$ TCE to non-toxic ethene in all three spikes and the three studied locations. In these sediment amended microcosms, TCE degradation started without a lag phase. This can be due to the presence of a high number of dechlorinating bacteria in river sediments which degraded TCE in shortest time. Methane production was also more pronounced in sediment microcosms which shows presence of active methanogenic community in river sediment which can create strong reducing conditions and also compete with dehalogenating bacteria for electron donor (data not shown). Also ethane formation was observed only in microcosms amended with river sediment (data not shown). This shows that the river sediment is rich in a microbial community capable of complete degradation of TCE to ethane which is missing in the aquifer compartment. After river sediment, the results of lactate amended microcosms was noticeable. However, in these microcosms, TCE degradation was accrued after an initial lag phase of 21 days. In microcosms supplemented with lactate, molasses or a sediment extract, compared to the initial TCE spike, TCE was degraded much faster after the second and third TCE spike. This is due to growth of halorespiring bacteria after the first TCE spike and/or establishment of efficient syntrophic and competitive reactions between those bacteria and electron donor providing organisms or organisms competing for available resources, respectively. In these microcosms, TCE degradation was stalled at ethene level and no ethane was produced (data not shown). Overall, the microcosm studies clearly demonstrated the presence of a larger TCE dechlorination potential in the eutrophic Zenne sediment compared to the aquifer adjacent to the river. None of the amended microcosms were as efficient in TCE degradation as those to which sediment was added. This shows that aquifer compartment not only lacks electron donor, but also efficient electron donor providing organisms and/or dehalogenating community.



a

b

c

Figure 8: (Stimulated) TCE degradation in microcosms with aquifer and groundwater from location PB26 (a) to *cis*-DCE (b), VC (c) and non-toxic end product ethene (d) in batch cultures. NA: natural attenuation, DC: dead control, GW: ground water control, SE (sed): sediment extract obtained after sedimentation, SE (cen): sediment extract obtained after centrifugation.

From these graphs, first order kinetic parameters were determined (TABLE V).

TABLE V: Degradation constant and half life time of TCE from TCE batch cultures (the values of first and third TCE spikes.are shown).

Batch cultures	SB2		Pb26		SB3	
	K (day^{-1})[a]	$T_{1/2}$ (days)[b]	K (day^{-1})	$T_{1/2}$ (days)	K (day^{-1})	$T_{1/2}$ (days)
NA[c]	ND[d]	ND	ND	ND	ND	ND
DC[e]	ND	ND	ND	ND	ND	ND
Lactate	0.010-0.669	68.63-1.04	0.171-0.220	4.05-3.15	0.062-0.358	11.23-1.93
Molasses			0.156-0.358	4.45-1.93		
Sediment	0.771-1.143	0.90-0.61	0.904-0.812	0.77-0.85	0.791-0.782	0.88-0.89
SE (cen)[f]			0.413-0.373	1.68-1.86		
SE (sed)[g]			0.344-0.091	2.02-7.63		

[a] K: degradation constant; [b] $T_{½}$: half life time of contaminant; [c] NA: natural attenuation; [d] ND: no degradation; [e] DC: dead control; [f] SE(cen): sediment extract obtained after centrifugation; [g] SE(sed): sediment extract obtained after sedimentation.

To determine the numbers of bacteria involved in the degradation of CAHs, Quantitative PCR was used to quantify different species and catabolic genes involved in the degradation of CAHs. More specifically, *Dehalococcoides* species were quantified since they are often detected in sites polluted with CAHs. *Dehalococcoides* is involved in the conversion of TCE to VC and subsequently to ethane. The catabolic genes *tce*A, *vcr*A and *bvc*A convert respectively TCE to VC, DCE to ethene and VC to ethene. Especially the detection of *vcr*A and *bvc*A is very important at the site since they convert the toxic VC to the harmless ethene.

Q-PCR analysis indicated that the numbers of *Dehaloccoides* were below the detection limit in the natural attenuation and dead control condition (Figure 9). However, in the conditions stimulated by lactate, molasses and sediment, the numbers of the CAH degrading bacteria increased from under detection limit to 10^5 cells/g soil after the addition of three TCE spikes to the microcosm. Also the catabolic genes *tce*A, *vcr*A and *bv*cA were only detected at the location Pb26 when a carbon source was added to the aquifer. From these results it was concluded that the right bacteria are present in the aquifer compartment at the Zenne site but the numbers were only high enough after the addition of extra carbon sources. This was in agreement with the CAH degradation since degradation only occurred in the presence of an added carbon source. Based on the numbers of *Dehalococcoides* detected in the aquifer, 10^5 cells/g soil can be used as the biomass factor in the reactive groundwater transport model.

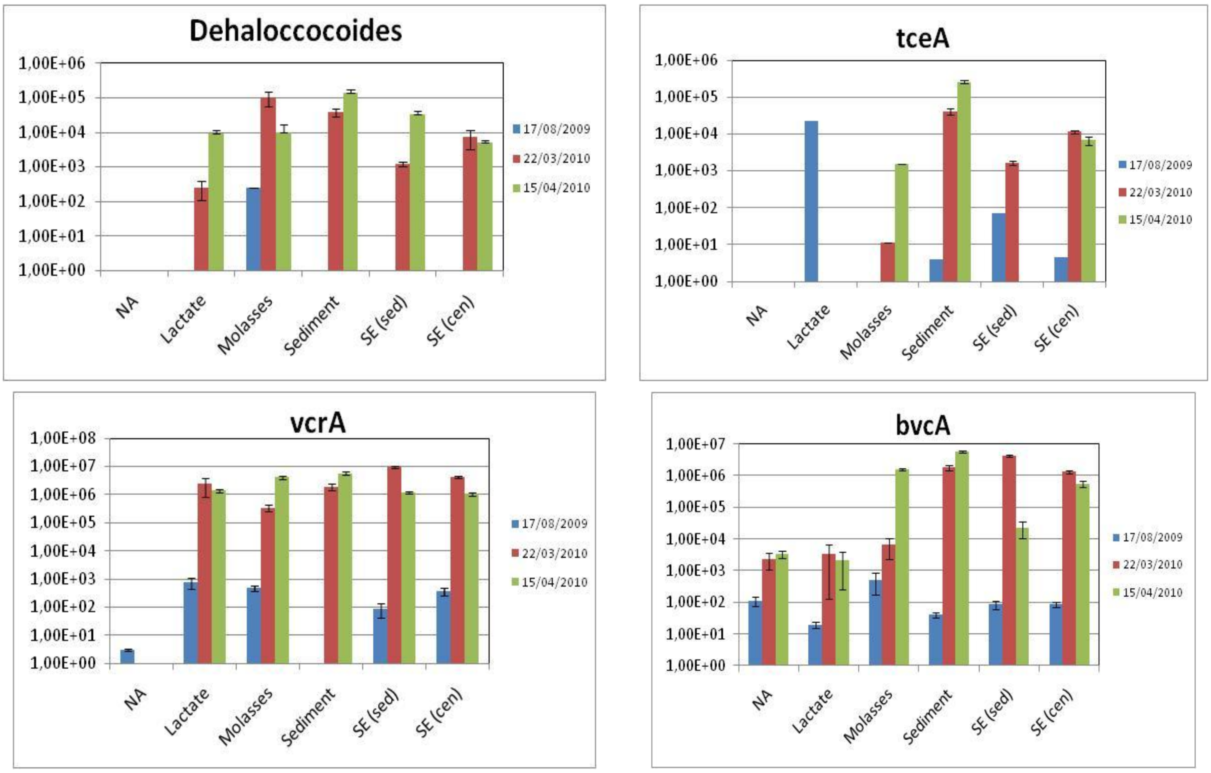


Figure 9: Quantitative PCR for the dechlorinating population Dehalococcoides and the reductive dehalogenase genes (tceA, vcrA and bvcA) in batches of PB26 stimulated with different electron donors.

The analyses of carbon sources suggested that the river sediment was the best carbon source for all three tested locations and has the highest effect in increasing the number of CAH degrading bacteria and the degradation rate of VC, c-DCE and TCE. Lactate had less effect on biostimulation at the first spike of the CAH compounds, but its effect on CAH degradation is similar to the river sediment at the third spike. The results also showed that sediment extracts have less efficiency in stimulation of CAH degradation and in increasing microbial numbers leaving out the possibility of using sediment extracts for field application. Besides, the difficulties in their preparation and handling make them less interesting choice compared to easily accessible and more efficient soluble carbon source such as lactate and molasses. We did not find effect of sample locations on CAH degradation rates from these batch experiments.

The here obtained VC, c-DCE and TCE degradation constants and biomass numbers can be used in the reactive transport model. They can be used to determine the decrease in the concentration of CAHs when these pollutants are transported through the groundwater from the source of pollution to the receptor. We have however to admit that these constants are very site dependent. However, by taking a certain range (e.g of 5%) around these constants, they can be used to predict which amount of pollutant will reach the river under the tested conditions.

2.7.4 Ecotoxicological Risk Assessment at Chimeuse East site.

Groundwater Quality TRIAD-like approach was applied on Chimeuse Est site located in the gravels and alluvia of the Meuse River, close to the city of Liège (more details in Deliverable D.3.2. (Crévecoeur & Thomé, 2010b) and validation of the methodology on Morlanwelz site (paper in submission). Contamination at this site is especially characterized by hydrocarbons, cyanides and heavy metals.

Boreholes were selected on this site (P23, P24, P25, P26, P28 and P29) and sampled between August 2009 and June 2010 (5 campaigns). The “Groundwater Quality TRIAD-like” measurements applied at Chimeuse Est site is summarized in Figure 10.

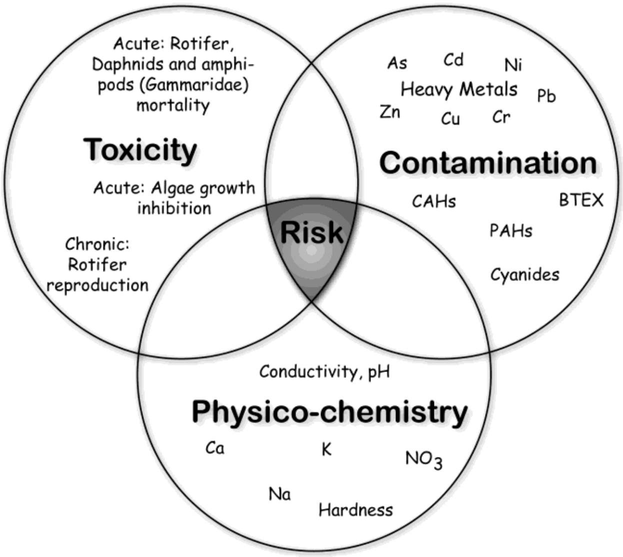


Figure 10: Schematic representation of the components of the TRIAD-like approach.

The groundwater quality of the studied site is assessed using a TRIAD-like approach based on 0 to 1 indices, the lower the score the better the quality. The results of the methodological approach are shown in TABLE VI.

TABLE VI: Risk indices values of the TRIAD-like components for groundwater at Chimeuse Est site (each borehole and globally; “0” for “no risk” and “1” for “high risk”).

	Groundwater samples						
TRIAD components	P23	P24	P25	P26	P28	P29	Chimeuse Est site
Chemistry							
ChemRI	0.9943	0.7782	0.9854	0.9984	0.7367	1.0000	0.9155
ChemRI Heavy metals	0.9820	0.7758	0.9792	0.9779	0.5170	1.0000	0.8720
ChemRI BTEX	0.4352	0.0000	0.0000	0.0959	0.0000	0.7211	0.2087
ChemRI Cyanides	0.4790	0.0000	0.1978	0.1945	0.4721	0.2084	0.2586
ChemRI CAHs	0.0103	0.0083	0.0064	0.8936	0.0074	0.0028	0.1548
Physico-chemistry							
PhysRI	0.6596	0.4557	0.5554	0.5483	0.4462	0.7872	0.5754
Ecotoxicology							
EtoxRI	0.8308	0.5603	0.6053	0.7485	0.3785	0.9494	0.6788
EtoxRI Acute	0.8087	0.5060	0.6100	0.6267	0.2906	0.9167	0.6264
EtoxRI chronic	0.8529	0.6146	0.6007	0.8702	0.4664	0.9821	0.7312
EnvRI	0.8282	0.5981	0.7124	0.7651	0.5205	0.9122	0.7232

The global ChemRI is similar to the ChemRI estimated for heavy metals, except P28. Benzene is also problematic at P23 and P29. Cyanides are present everywhere, except at P24. On the contrary, a risk caused by chlorinated compounds is only assessed at P26. The TRIAD-like approach application confirms the widespread measured metal contamination (As, Cd, Cu, Ni, Pb, Zn) at this site. PhysRI is lower compared to ChemRI. Groundwater sampled at P29 is acid and is responsible for high PhysRI. The ecotoxicological parameter is characterised by spatial heterogeneity, from moderate to relatively high risk.

Results show statistically significant differences (ANOVA followed by Tukey test, $p < 0.05$) among the three risk indices for P23, P24 and P26 and between ChemRI and the two other risk indices for P25 and P28. On the contrary, groundwater sampled at P29 show statistically significant differences only between ChemRI and PhysRI (Figure 11).

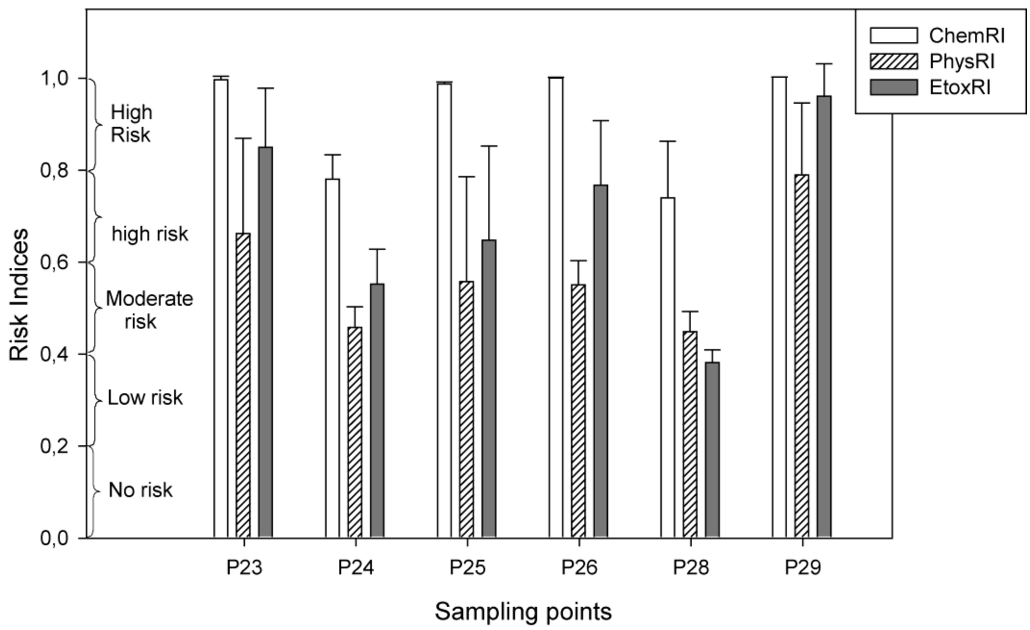


Figure 11: Risk indices (ChemRI, PhysRI and EtoxRI) representation for the different sampling points at Chimeuse Est site.

In addition of quantitative results, risk indices can be compared using graphical presentation detailed in Deliverable D.3.2 (Crévecoeur & Thomé, 2010b).

In conclusion, the GwQT provides evidence of the poor health status of groundwater at Chimeuse Est site (EnvRI = 0.7232) as a result of heavy metals pollution (CAHs at P26) with spatial heterogeneity among sampling points. None of the three TRIAD components (chemistry, physico-chemistry and ecotoxicity) could reliably predict the other one. Integration of new variables such as biomarkers, other bioassays, other physico-chemical measurements and studies of bioavailability should contribute in the future to the improvement of this integrated approach for the characterization of groundwater quality. Moreover, higher numbers of sampling points per site and samples are crucial to reduce uncertainties.

A more than additive effect (synergism) was indicated for all bioassays (*e.g. Brachionus calyciflorus* chronic bioassay, Figure 12), except with *Daphnia magna* (antagonism, “less than additive”). Therefore, mixture toxicity is higher (lower for *Daphnia magna* bioassay) to the toxicity that would be expected if we sum each pollutant toxicity.

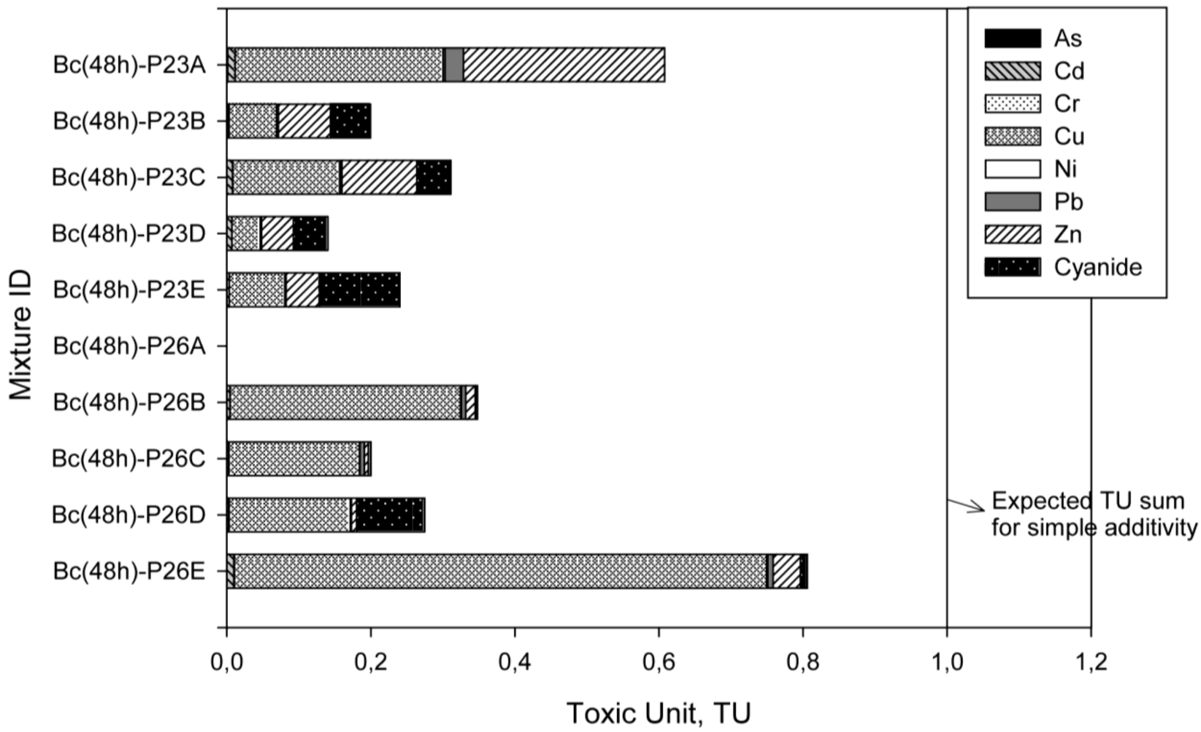


Figure 12: Toxic Units sum in 9 mixtures of groundwater sampled at Chimeuse Est site (P23 and P26), *Brachionus calyciflorus* (Bc) chronic bioassay.

Among the metals, copper (followed by zinc and cyanides) posed highest risk to aquatic species. Furthermore, results demonstrate that no single test is certain to give conclusive results, wherefore it is preferable to use a battery of bioassays.
A small amount of a very toxic chemical in a mixture can have a much larger effect on the biological response than a large amount of a slightly toxic chemical. Therefore, the Toxic Unit approach is a useful tool to determine the pollutant causing the toxicity.

Chemicals at concentrations as low as 10% of their respective Water Quality Standards have been found to be toxic when acting together as a mixture such as Cu, Cd. These results indicate that groundwater quality guidelines for individual chemicals could underestimate the overall exposure effect, and therefore would not be protective. The implication here is that regulatory limits for individual metals may not be sufficiently protective, particularly when the element is occurring in a mixture (Cooper & al, 2009).

Two examples are proposed to illustrate the methodology and to show its usefulness for groundwater management in urbanised and industrialised areas: a synthetic example and the real scale application of a deteriorated groundwater body in Belgium.

2.7.5 Regional scale application on the Groundwater Body RWM073

Context

The Meuse River flows in the Walloon Region of Belgium along 128 km from the French to the Dutch borders. The alluvial aquifer located in the deposits of the river contains an important groundwater resource which is exploited for water distribution and industry thanks to many water catchments located in the alluvial plain (Haddouchi, 1987; Brouyère et al., 2006). In the region of Liège, heavy industries related to coal extraction, metallurgy and chemistry have been developed for more than two centuries. These industries were preferentially settled in the alluvial plain, near the river, for facilitating transportation by boat of primary and final products. This, together with a growth in the urbanization, has resulted nowadays in the existence of numerous potentially contaminated sites and a generalized contamination of groundwater in the alluvial aquifer.

The portion of alluvial aquifer corresponding to the industrial area, located North-East of the Meuse Belgian stream (Figure 15a), has been defined as a distinct groundwater body (named RWM073) from the rest of the alluvial aquifer to allow defining specific measures related to the issue of deterioration in groundwater quality.

The problem of regional groundwater contamination is known and documented through water analyses at different locations, but important questions remain. There is no real global estimation of the actual level of water quality deterioration at the scale of the groundwater body, there is no estimation of the actual contribution of industrial activities to this deterioration as compared to other possible contamination sources, such as bedrock groundwater rebound after coal mine closure or diffuse atmospheric pollution, and there is no integrated referential for land cleanup prioritization and cost – efficiency assessment. To contribute to answer these questions, an application of the regional-scale risk assessment approach has been initiated on this groundwater body. The first steps and results are described here below as a real case illustration of the methodology.

The Meuse river alluvial gravel groundwater body RWM073 stretches on 40 kilometres. The alluvial plain has a mean width of 3 km. The usual top-bottom geology consist of 2 to 4 meters of backfill deposits, 1 to 4 meters silt sand clay deposits and approximately 8 meters of alluvial gravels lying on a shaly bedrock that here is considered as the impervious lower boundary of the aquifer. A data mining on the hydraulic properties of the aquifer has revealed hydraulic conductivity values ranging from 10^{-3} to 10^{-6} m/s with a mean value of 8×10^{-5} m/s. Groundwater flows mainly towards the Meuse River with a mean hydraulic gradient of 0.003 m/m.

Estimation of groundwater recharge

Groundwater recharge is among the most important factor acting on groundwater flow and pollutant vertical leaching through the unsaturated zone from source to the aquifer. The surface of the RWM073 groundwater body is covered at 35% of industrial area and at 55% of urbanized area (SPW, 2007). This has a strong effect on groundwater recharge and particularly on its spatial distribution Since we decided to upscale the approach to the level of the groundwater body in the second phase, the land-cover classification methodology developed during the first phase had to be adapted. Because of the large area covered by the new test site (RWM073), we had to make use of lower resolution data. A Landsat ETM+ image of July 2001 (a 15-meter resolution for the panchromatic band and 30-meter resolution for the multispectral bands) was used. Given that this spatial resolution is too low to obtain detailed information about the land-cover in the urbanized areas we opted for a stratified classification approach, distinguishing built-up areas (urban and industrial land use) from natural/agricultural areas. In the first step of the stratified approach the satellite image is classified on pixel level, resulting in 6 land-cover classes (water, grass, forest, bare soil, grey urban and bright urban). In the next step of the stratified approach all the urban pixels are characterized at sub-pixel scale to determine the fraction of impervious area inside every urban pixel. To obtain more detailed information about the natural land cover classes we opted for a multi-temporal approach to be able to characterize the different types of crops, trees, etc. Because of the clouds we could not get satellite images of different seasons or growing stages what made the temporal analysis impossible. Since it was needed to obtain more detailed information about the different natural land cover classes of the RWM073 study site a combination of our obtained land-cover map at pixel size and a land-cover map of the Walloon Region (COSW) was performed. The COSW of the WR could provide us information about the different types of crops present in the study area. By crossing both images, we were able to characterize the different types of vegetation inside the natural areas of the RWM073 site, what resulted in 12 hydrological more relevant land-cover classes (urban, bare soil, agriculture, meadow, orchards, shrubs, grass, spruce, water, deciduous forest, coniferous forest and mixed forest). Figure 13 shows the obtained pixel and supixel classification of the RWM073 site.

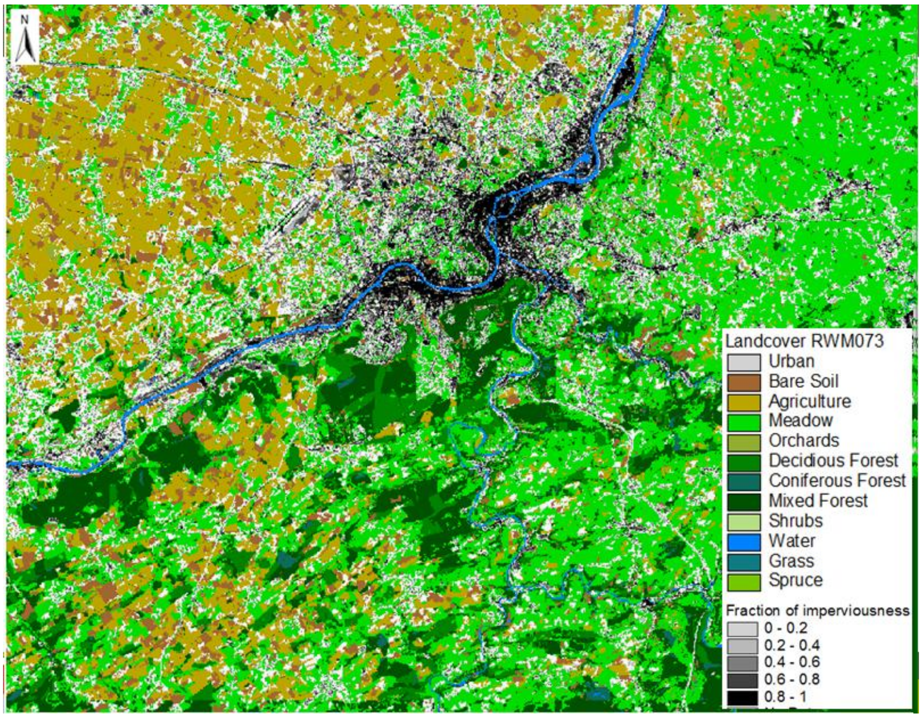


Figure 13: Land-cover map obtained for the Vilvoorde study site

The obtained land-cover information is then used as an input for the high-resolution groundwater recharge modelling using WetSpass. In WetSpass, every land-cover class is characterized by a given fraction of imperviousness, and all these values are grouped in the landuse parameter tables. But with the subpixel classification approach, every urban pixel has his own fraction of imperviousness. So the WetSpass model has to be adapted to take into account the fraction of imperviousness inside every urban pixel. Instead of using the landuse parameter tables as an input in WetSpass to obtain information about the fixed fractions of imperviousness inside the urban pixels, WetSpass gets this information immediately from input grids, containing the different types of fraction for every pixel. Therefore 8 grids were created containing the fractions of imperviousness, vegetation, open water and bare soil for every pixel, and this for summer and winter season. Figure 14 illustrates the yearly average recharge estimations for RWM073.

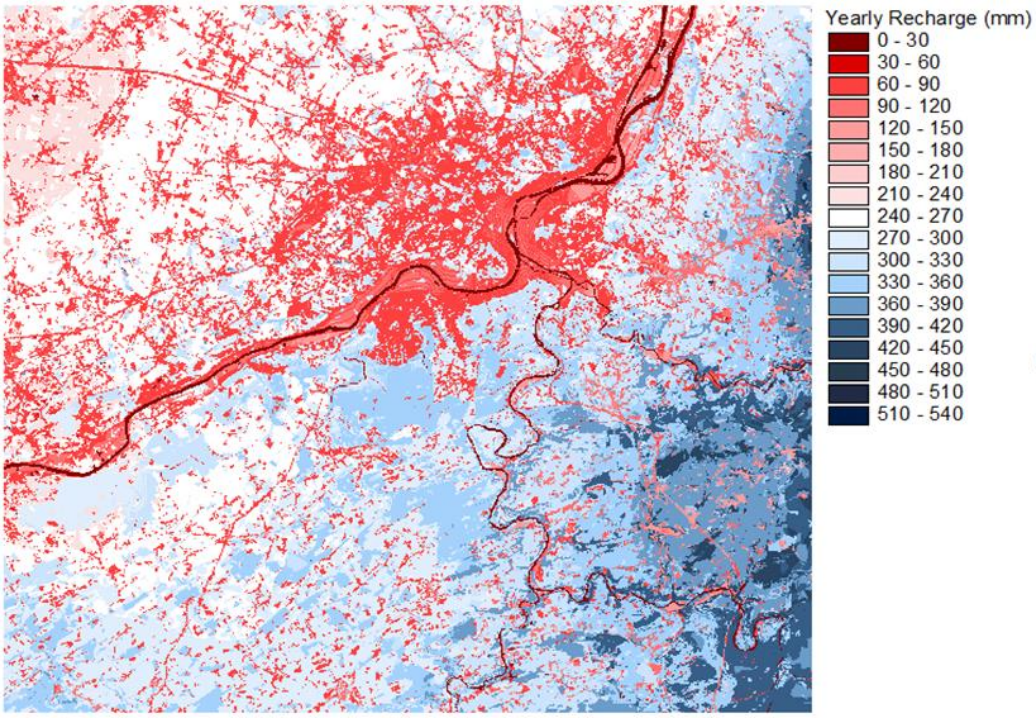


Figure 14: WetSpass simulated yearly long-term average recharge

A mean yearly recharge of 249±104 mm/y is obtained for the RWM073 site, with a mean summer recharge of 9±26 mm/6 months and a mean winter recharge of 240±97 mm/6 months. The highest recharge values occur in the forest areas, especially coniferous forest zones. It is again clear that the land-cover has a strong influence on the spatial distribution of the groundwater recharge in the study area.
The result of the groundwater recharge is then stored into the geospatial database to be easily integrated into the groundwater flow and transport model.
Data exchange between the high resolution spatially distributed recharge results and the groundwater flow model is easily made with ASCII, text or raster file formats.

Groundwater flow and transport modelling

Groundwater flow and transport simulations are performed using MODFLOW (Harbaugh et al., 2000) and MT3D (Zheng and Wang, 1999). The model is developed using the finite difference method with a constant cell size of 20 by 20 meters. The top of the model is given by a DTM and the cell thickness is assumed constant (15 m). The external boundary conditions are represented by specified fluxes around the alluvial plain to consider groundwater flowing from the hill slope of the valley, by the infiltration recharge and by the River Meuse acting as regional drain of the aquifer. The groundwater flow model is calibrated in steady state mode using piezometric head measurements available in the area (Figure 15b). Transport parameters are defined based on previous experiments and scientific works in the Meuse alluvial aquifer (Derouane and Dassargues, 1994; Brouyère, 2001; Batlle-

Aguilar et al., 2008; 2009). Transport simulations are run over a 20 years period. Flow and transport simulations were run using three different groundwater recharges. The first is a mean infiltration estimated on a water budget performed on the groundwater body, is uniformly distributed over the groundwater body and values 120 mm/y. The second and third groundwater recharges are the results of Wetspass modelling respectively at pixel (249 mm/y) and sub-pixel level (285 mm/y) and are thus spatially distributed. Eventually, the high resolution sub-pixel spatially distributed recharge was kept for the modelling.

Simulations and results

The GIS-based regional risk assessment application has been used to extract the most common industrial activities recovered on the groundwater body, *i.e.* industrial plants included in the categories “mining”, “gas station” and “metallurgy”. Only important plants with a spatial extent larger than 5000 m² have been considered. In this example, benzene is the only contaminant taken into consideration, with a worst case scenario assuming that all potential sources are active. The sources of contaminant are assumed to leach continuously in the recharge to the aquifer at maximum solubility and contaminant spreading in groundwater is modelled considering advection, dispersion, sorption and degradation processes.

The influence of Wetspass calculated recharges are naturally a rise of the groundwater table elevation on the whole groundwater body since they are higher than the recharge estimated by the water budget. But the most important contribution of high resolution recharge is for the transport of contaminant to and through the aquifer. The pollutant input in the model is set as a specified concentration in the recharge. So if the recharge increases, the mass flux of pollutant reaching the groundwater table increases. For the water budget mean recharge, the yearly benzene mass flux reaching the groundwater is 642 tons for the whole groundwater body. For pixel and sub-pixel high resolution recharge the benzene flux is 573 and 763 t/y respectively. The shape of the pollutant plumes appears to be narrower with the two high resolution recharge methods.

Groundwater quality indicators

Once the simulation is performed in GMS, results files are exported to the GIS application to calculate maps of the SEQ-ESO indicators and the global quality index at each time step. Figure 15b shows the map of benzene concentrations in the alluvial aquifer and Figure 15c, a map of the global quality index, both after 10 years. This map highlights highly contaminated zones. The last graph (Figure 15d) shows the evolution with time of the SEQ-ESO global quality index for the whole groundwater body. Contaminant plumes reach steady state and equilibrium between source leaching and contaminant advection, dispersion, degradation and drainage by surface water after approximately 5 years. It shows that, a single polluted site does not have a significant effect on the overall quality of that whole groundwater body but that, with reasonable assumption and despite the turnover of groundwater and the degradation of pollutants, multiplication of industrial contaminated sites can have a strong effect on the groundwater quality at the scale of the groundwater body.

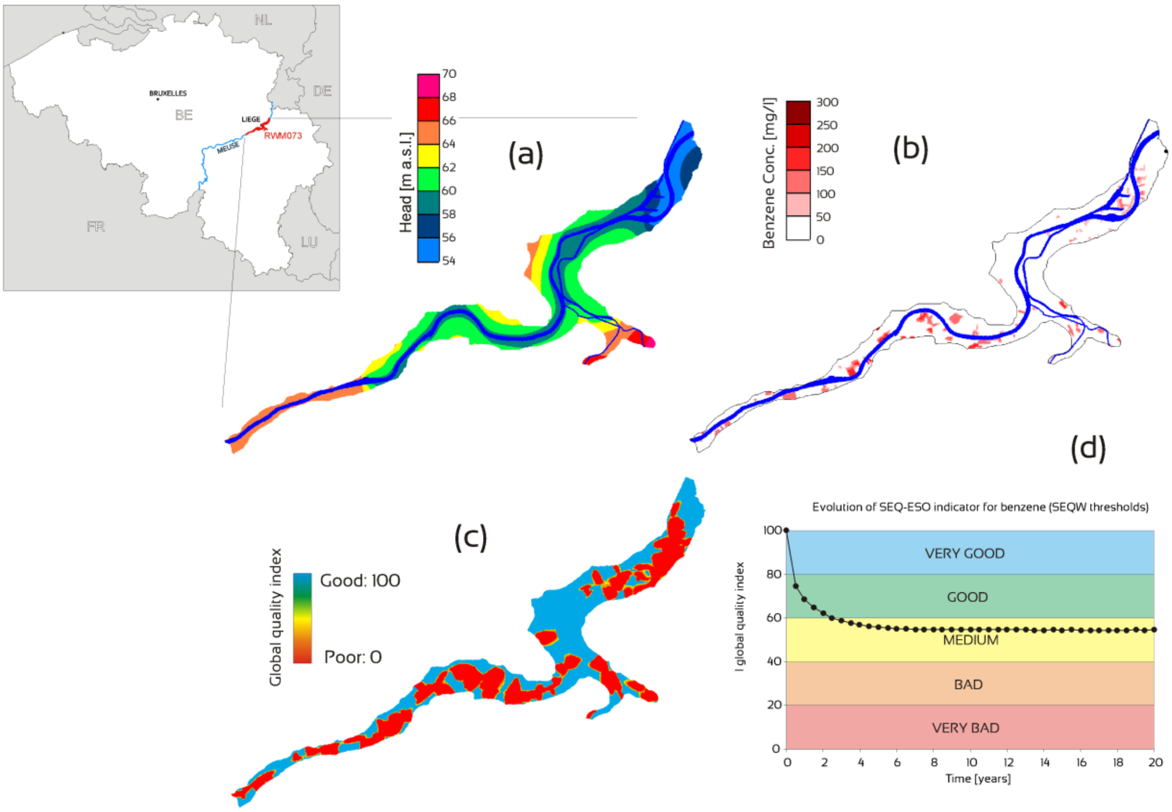


Figure 15: Preliminary results of regional risk assessment on groundwater body RWM073 considering industrial activities releasing benzene into the groundwater. The resulting SEQ-ESO indicator suggests a "medium" quality status of the groundwater body.

Similar map can also be performed using ecotoxicological risk indicator for benzene. If concentration of benzene (C_i) is lower than PNEC value (80µg/L), no hazard is expected for the environment. Otherwise, the ecotoxicological risk index is assessed using colours corresponding to different effect concentrations (TABLE VII).

TABLE VII: Ecotoxicological risk and indicator of benzene assessed for *Daphnia magna* bioassay (NOEC, No Observed Effect Concentration; LOEC, Low Observed Effect Concentration; $L(E)C_{50}$ ($L(E)C_{100}$), 50% (100%) of effect)

Ecotoxicological risk	Risk Indicator
Ci ≤ 3mg/L (NOECi)	No risk (- -)
3mg/L < Ci ≤ 4.76mg/L (LOECi)	Low risk (-)
4.76mg/L < Ci ≤ 10mg/L (LC50,i)	Moderate risk (±)
10mg/L < Ci < 1400mg/L	Relatively high risk (+)
Ci = 1400mg/L (L(E)C100,i)	High risk (+ +)

These values coming from ecotox databases (EPA and INERIS websites) are determined for benzene tested on *Daphnia magna*, a planktonic species. However, those data differ according to the pollutant considered, the species (*e.g.* NOEC of benzene for algae specie, Pseudokirchneriella subcapitata is 5.81mg/L) and endpoint tested. Moreover, interactions between pollutants could not be considered in this ecotoxicological risk index.

The influence of high resolution spatially distributed recharge has also been investigated. Maps of SEQ-ESO indicators show that the areas of bad quality index for the groundwater are more restricted in space with the high resolution recharge than with the water budgeted recharge. The spatially distributed recharge gives smaller but more concentrated contaminant plumes in the groundwater. The global quality index for the whole groundwater body is lower (worst) for the high resolution recharge calculation method.

Discussion of results

Simulations were run with the strong assumption of a worst case scenario. It considers benzene as the only contaminant, all the industrial sites of more than 5000 m² as actually contaminated on their entire extend area and concentration in benzene in the recharge equal to its maximum solubility in water. These assumptions mean a large amount of contaminant injected into the groundwater body. A real case is for sure much more complex. Many different contaminants are usually founded into groundwater with different physico-chemical properties (sorption, degradation). Sources strength is usually smaller since not all the surface of an industrial site is contaminated. But leaching can be greater in case of non aqueous phases. This attests that no definitive conclusions can be drawn at this level and that data on contaminated sites characterization remain the most important lack to fill for a regional risk assessment.

2.8 Conclusions and perspectives

2.8.1 Contribution of the FRAC-WECO project to regional contamination issues

A regional scale risk assessment methodology for groundwater bodies has been developed. This RRA is a flexible approach for evaluating the pressure exerted by various sources of contamination on groundwater resources. The methodology is based on the aggregation of various cumulative sources of contaminants of different chemical natures, properties and toxicities into a set of “easy to use” spatially distributed or aggregated indicators.

The spatially-distributed indicators (maps of indicators) can be used to identify most problematic sectors (hot contamination spots) with larger volumes of heavily contaminated groundwater and the evolution with time of groundwater quality at different locations in the catchment. The spatially-aggregated indicator can be used to report on the global status of groundwater quality in the groundwater body, for groundwater quality trend assessment and as a referential for site prioritization and for the evaluation of programs of measures aiming at restoring groundwater quality in the groundwater body, using cost-efficiency approaches.

Thanks to these capabilities, the regional risk assessment methodology is compliant with the ongoing legislation in the Walloon region, based on the SEQ-ESO and it fits very well with the guidelines of the EU Water Directive which promotes the use of aggregated indicators able to reflect status and trends in groundwater quality and to evaluate in a rigorous way the risk of not reaching a good status by specific milestones such as 2015.

A synthetic example and a first real-case application for the alluvial aquifer of the Meuse River in the region of Liège (Belgium) are developed. These examples confirm that the approach provides a useful referential for decision-making in relation with regional contamination of groundwater. The synthetic test case demonstrate that the approach is able to turn a complex multiple contamination into a simple global quality index, evolving with time and easily incorporated or compared to a risk index.

The application on the RWM073 allows also defining the weakest points and drawbacks of such a regional approach and the priorities for further developments.

2.8.2 Perspectives

First, the regional risk assessment approach proposed here is data demanding. However, this is the case for all these regional scale approaches that require relatively important preparatory works for data acquisition, organisation and processing. This drawback is partly overcome by the use of a geospatial database and specific interfacing tools between the GIS system, the groundwater flow and transport modelling application and the regional SEQ-ESO calculation module. In this context, further work is ongoing on developing customised user-interfaces for a better integration of the whole procedure.

Secondly, there are many sources of uncertainties in the different data feeding the approach. In particular, the pollution sources are not always perfectly identified and characterized and hydrogeological parameters and contaminant properties remain difficult to identify and quantify at the regional scale. Further investigations are required on the reconstruction of the history of contamination of the groundwater body and on a better evaluation of source strengths. A key next step will be to implement a statistical approach for handling all the uncertainties that remain at regional scale. More particularly, it is expected to obtain, in a near future, statistics on contaminant leaching in relation with different industrial and environmental factors such as characterization of contaminated site (industrial activity, area, land use) and properties of the soil (groundwater flow, lithology). To reach that objective, information on contaminated sites available in the French database ADES developed and managed by BRGM agencies, will be used. This database contains analyses for contaminants in groundwater for most industrial plants in France as requested by the regional environmental agencies.

Finally, the regional flux-based risk assessment approach presented here is now used as a referential for a cost - benefit assessment of total or partial remediation of the contaminated groundwater body, according to different management scenarios. This analysis starts from the actual groundwater body quality state for which groundwater restoration scenarios (based on natural attenuation or active remediation) are suggested. The regional risk assessment method is then applied on these scenarios to evaluate the resulting improvement of the groundwater quality index when management plans are applied. This allows evaluating, in monetary terms, the costs for given improvements in the groundwater body quality status.

3. POLICY SUPPORT

The regional scale risk assessment method for groundwater bodies is proposed here as a flexible approach for evaluating the pressure exerted by various sources of contamination on groundwater resources by a “easy to use” spatially distributed or aggregated indicator.
The spatially-distributed indicators (maps of indicators) can be used to identify most problematic sectors (hot contamination spots) with larger volumes of heavily contaminated groundwater and the evolution with time of groundwater quality at different locations in the catchment. The spatially-aggregated indicator can be used to report on the global status of groundwater quality in the groundwater body and for groundwater quality trend assessment,
Thanks to these capabilities, the regional risk assessment methodology is compliant with the ongoing legislation in the Walloon region, based on the SEQ-ESO and it fits very well with the guidelines of the EU Water Directive which promotes the use of aggregated indicators able to reflect trends in groundwater quality.

Combination of TRIAD-like and Toxic Unit approaches will allow classification of sites into categories according to the degree of contaminant-induced deterioration. Furthermore, it will provide a helpful decision support in order to determine the micro-pollutant concentrations which can not be exceeded in the environment to prevent ecological damages to the ecosystem.

The socio economic study shows that beside direct and optional benefits that may be expected from a good groundwater quality (2M€/year from 2030), the indirect value is almost as important (7,5 M€/year). It emphasizes the fact that with an effective individual information and sensitization, citizen can feel concerned for environmental issues and become ready to participate to the improvement effort.

4. DISSEMINATION AND VALORISATION

The Regional Risk Assessment methodology developed in the FRAC-WECO project is already used in another research project: Pollusol 2. This project is funded by the Walloon agency for environment quality (SPAQuE). It aims to determine soil and groundwater quality status in the vicinity of large industrial and urban areas and also to develop a GIS-based tool to evaluate groundwater quality at megasite and regional scale. The RRA has already been integrated to this tool that allows from the GIS interface to run groundwater flow and transport model, to apply contamination scenarios and to calculate a quality index for an entire aquifer or for the megasite of interest.

5. PUBLICATIONS

5.1 Submitted

Crévecoeur S. 2011?: Groundwater quality assessment using a TRIAD-like approach", explaining GwQT methodology with an application on Morlanwlez (and Chimeuse) sites. Submitted to *Environmental Pollution* journal.

Hamonts,K., Kuhn T., Vos J., Maesen M., Kalka H., Springael D., Meckenstock R.U. and Dejonghe W., 2011?: Temporal Variations in Natural Attenuation of Chlorinated Aliphatic Hydrocarbons in Eutrophic River Sediments Impacted by a Contaminated Groundwater Plume. Submitted to Environmental Sciences & Technology.

Dujardin J., Jamin P., Bashir I., Batelaan O., Canters F., Brouyère S. and Dassargues A. , 2012?: Regional flux-based risk assessment of contaminants in a groundwater body using remotes sensing imagery integrated in a modeling procedure.

Dujardin J., Anibas C., Bronders J., Jamin P., Dejonghe W.$_{c}$ Brouyère S. and Batelaan O., 2012?: Improving groundwater risk management by combining different flux estimation techniques. Submitted to Groundwater monitoring and remediation.

5.2 Peer review

(See annexe 1 for complete publications)

Batlle-Aguilar J., Brouyère S., Dassargues A., Morasch B., Hunkeler D., Höhener P, Diels L., Vanbroekhoven K., Seuntjens P. and Halen H. Benzene dispersion and natural attenuation in an alluvial aquifer with strong interactions with surface water. Journal of Hydrology, Volume 369, Issues 3-4, 15 May 2009, Pages 305-317.

Brouyère S., Jamin P., Orban Ph., Dassarges A., Popescu I.-C., Hérivaux C., 2010: A regional flux-based risk assessment approach of contaminated sites on groundwater bodies. Groundwater Quality Management in a Rapidly Changing World, Red Book of the 7th International IAHS Groundwater Quality Conference 2010.

Dujardin J., Batelaan O., Canters F., Boel S., Anibas C., and Bronders J., 2010: Improving surface-subsurface water budgeting using high resolution satellite imagery

applied on a brownfield, Science of the Total Environment 409 (2011) pp.799-808, DOI :10.1016/j.scitotenv.2010.10.055

Jamin P., Dolle F., Chisala B., Orban Ph., Popescu I.-C., Hérivaux C., Dassarges A. and Brouyère S., 2012: A regional flux-based risk assessment approach for multiple contaminated sites on groundwater bodies. Journal of Contaminant Hydrology, vol. 127 (1-4), pp. 65-75.

5.3 Oral communications and posters

W. Dejonghe et al.:Sediment biobarriers for chlorinated aliphatic hydrocarbons in groundwater reaching surface water. Fifth international conference on remediation of contaminated sediments, 2-5 february 2009, Jacksonville, Florida.

W. Dejonghe et al.: Sediment biobarriers for chlorinated aliphatic hydrocarbons in groundwater reaching surface water. The tenth international in situ and on-site bioremediation symposium, 5-8 May 2009, Baltimore, Maryland.

Dejonghe W and K. Hamonts: Sediment biobarriers for chlorinated aliphatic hydrocarbons in groundwater reaching surface water. Riskpoint International Workshop, 5-6 October 2009, Denmark.

Brouyère S. et al.: A regional flux-based risk assessment approach of contaminated sites on surface water and groundwater bodies. Contaminated site management in Europe, 27-29 October 2009, Ghent, Belgium.

Dujardin S. et al.: Improving surface-subsurface water budgeting for brownfield study sites using high resolution satellite imagery. Contaminated site management in Europe, 27-29 October 2009, Ghent, Belgium.

Brouyère S and Battle-Aguilar J: Modeling ground water and benzene discharge to a river from an alluvial aquifer subject to strong interactions with surface water. AGU, 14-18 December 2009, San Francisco, California.

Brouyère S., Jamin P., Dolle F., Orban P., Hérivaux C., Popescu I.C., Dassargues A.: A regional flux-based risk assessment approach of contaminated sites on groundwater bodies. 3rd International Meuse Symposium, Poster, April 22-23 2010, Liège.

Brouyère S., Jamin P., Dolle F., Chisala B., Orban P., Popescu I.C., Hérivaux C., Dassargues A.: A regional flux-based risk assessment approach of contaminated sites on groundwater bodies. Groundwater Quality 2010, Oral presentation, June 13-18 2010, Zurich.

Dujardin J., Jamin P., Batelaan O., Canters F., Dassargues A. and Brouyère S.:Integrated remote sensing imagery and subsurface flow and transport modeling for regional scale risk assessment of contaminants on groundwater bodies. European Geoscience Union General Assembly, 03-08 April, 2011, Vienna

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8. ANNEXES

ANNEXE 1: COPY OF THE PUBLICATIONS

ANNEXE 2: MINUTES OF THE FOLLOW-UP COMMITTEE MEETINGS

ANNEXE 3: DETERMINATION OF DEGRADATION CONSTANTS AND BIOMASS NUMBERS FOR A SITE CONTAMINATED WITH CHLORINATED ALIPHATIC COMPOUNDS – ZENNE SITE AS A CASE STUDY

ANNEXE 4: ECOTOXICOLOGY

ANNEXE 5: SOCIO-ECONOMIC COST-BENEFITS ANALYSIS

THE ANNEXES ARE AVAILABLE ON THE WEBSITE

http://www.belspo.be/belspo/SSD/science/pr_terrestrial_fr.stm