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Integrated methodology for assessing the HCH groundwater pollution at the multi-source contaminated mega-site Bitterfeld/Wolfen

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Abstract A large-scale groundwater contamination characterises the Pleistocene groundwater system of the former industrial and abandoned mining region Bitterfeld/Wolfen, Eastern Germany. For more than a century, local chemical production and extensive lignite mining caused a complex contaminant release from local production areas and related dump sites. Today, organic pollutants (mainly organochlorines) are present in all compartments of the environment at high concentration levels. An integrated methodology for characterising the current situation of pollution as well as the future fate development of hazardous substances is highly required to decide on further management and remediation strategies. Data analyses have been performed on regional groundwater monitoring data from about 10 years, containing approximately 3,500 samples, and up to 180 individual organic parameters from almost 250 observation wells. Run-off measurements as well as water samples were taken biweekly from local creeks during a period of 18 months. A kriging interpolation procedure was applied on groundwater analytics to generate continuous distribution patterns of the nodal contaminant samples. High-resolution geological 3-D modelling serves as a database for a regional 3-D groundwater flow model. Simulation results support the future fate assessment of contaminants. A first conceptual model of the contamination has been developed to characterise the contamination in regional surface waters and groundwater. A reliable explanation of the variant hexachlorocyclohexane (HCH) occurrence within the two local aquifer systems has been derived from the regionalised distribution patterns. Simulation results from groundwater flow modelling provide a better understanding of the future pollutant migration paths and support the overall site characterisation. The presented case study indicates that an integrated assessment of large-scale groundwater contaminations often needs more data than only from local groundwater monitoring. The developed methodology is appropriate to assess POP-contaminated mega-sites including, e.g. HCH deposits. Although HCH isomers are relevant groundwater pollutants at this site, further organochlorine pollutants are present at considerably higher levels. The study demonstrates that an effective evaluation of the current situation of contamination as well as of the related future fate development requires detailed information of the entire observed system.

Keywords HCH · Hexachlorocyclohexane · Groundwater pollution · Mega-site · Bitterfeld · Geological 3-D model · Hydrogeological model · Regionalisation · Stockholm Convention

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Introduction

Three hexachlorocyclohexane (HCH) isomers—α-HCH, β-HCH and lindane (industrial γ-HCH)[1]—were listed as "new" persistent organic pollutants (POPs) in the Stockholm Convention. Waste isomers from lindane production were mostly dumped near the production sites. Thus, these deposits need to be identified and investigated (Vijgen et al. 2010; Vijgen 2006). Located in Eastern Germany (former German Democratic Republic), the local chemical industry at the Bitterfeld/Wolfen location produced a relevant lindane and dichlorodiphenyltrichloroethane (DDT) contingent from 1951 to 1982 (Sommerwerk 2003; Chemie AG Bitterfeld-Wolfen 1993). Increased HCH concentration levels in fish have been detected in the adjacent rivers Mulde and Elbe after the flooding event in August 2002[2] (German Environmental Agency 2005; supporting information, Figs. 1 and 2) being an example where increasing flooding events remobilize POPs from deposits (Wölz et al. 2008).

The regional groundwater contamination of Bitterfeld/Wolfen has been strongly formed by several environmental impacts initiated from a range of chemical production, multiple resulting waste dumps and extensive open-pit lignite mining for more than a century (Sommerwerk 2003). Due to a long-term mining history, the region has arisen to one of the earliest chemical production sites in the world. Industrial manufacture of chlorinated chemicals, which make up the main contaminants, started at the end of the nineteenth century with the operation of one of the world first chloro-alkali electrolysis (Sommerwerk 2003). Today, the region is one of the best investigated contamination sites worldwide.

The hydrochemical situation is characterised by a complex mixture of organic compounds (chlorinated aliphatic and aromatic hydrocarbons) which comprises a high diversity of individual organic substances as well as a high regional variability of these contaminants within the Quaternary and Tertiary aquifers (Heidrich et al. 2004; Rückert and Weiss 2004; Wycisk et al. 2003; Weiss et al. 2001).

A comprehensive understanding of related transport and natural attenuation processes needs a detailed knowledge of the complex hydro-stratigraphical settings of the subsurface, which is directly linked to the groundwater flow and transport dynamics as well as the resulting impacts on environmental receptors. North-western of the Bitterfeld area, several former open-cast mining fields had been used directly for dumping residuals from chemical production. Hydrogeologically, these waste sites are located in areas that were irreversible restructured by mining activities with respect to their natural bedding conditions. Since the industrial dumpsites are not completely sealed, the groundwater was/is significantly affected by the chemical inventory of chlorinated aliphatic and aromatic hydrocarbons of the deposits and former releases from production processes. The regional long-term groundwater monitoring considers approximately 180 individual organic substances with about 60 priority contaminants (supporting information, Table 1). One of the largest deposits is the abandoned chemical deposit "Antonie" containing about 5,000,000 m^3 of various industrial residues (Fig. 1). The chemical inventory includes more than 60,000 tons of largely HCH waste isomers from the local lindane production (Heinisch 1992; Walkow et al. 2000), 70,000 tons sulphuric acid residuals, 107,000 tons neutralisation sludge, 48,000 tons of DDT sludge and 32,000 tons of other substances. Traces of the HCH isomers are present in all environmental compartments, in the surface water (Paschke et al. 2006a; Vrana et al. 2006), in groundwater (Vrana et al. 2005) and in the soil layer (Schwartz et al. 2006), respectively, in the soil solution (Kalbitz and Popp 1999) and in the atmosphere (Popp et al. 2000; Paschke and Popp 2005; Paschke et al. 2006b).

Due to the extensive lignite mining, the hydraulic system behove long-term changes in their dominating groundwater flow directions and gradients. A detailed understanding of these impacts is essential for the reproduction of the contaminant release history in these regionally contaminated areas (Wycisk et al. 2004). Thus, a numerical modelling approach has been chosen to identify these mining-induced changes to the hydraulic system. At the same time, accurate subsurface information about the aquifer heterogeneity and their related hydraulic properties is inevitable to minimize the uncertainties of estimated pollutant transport pathways as well as the contaminant interaction with surface water and their aquatic biocenosis.

Finally, a regional geo information system (GIS)-based 3-D model was developed which includes any relevant information and aspects for the site characterisation. The GIS model involves the following data: (a) conceptual model of the regional contamination, (b) regionalisation of groundwater pollutants, (c) high-resolution 3-D subsurface models of the regional aquifer stratigraphy, (d) a numerically reproduced groundwater flow dynamics and (e) a land use sensitivity classification. Moreover, this paper provides recommendations for further actions and highlights the importance of an interdisciplinary groundwater assessment/modelling approach for organochlorine production sites being, e.g. characterised by impacts of HCH pollution. The case study was selected as part of the POPs-contaminated site series (Weber et al. 2008a) since it represents a best practise approach of assessing the groundwater situation of a POPs-contaminated mega-site (Weber et al. 2008b). Furthermore, such a detailed site investigation supports the estimation of needs and costs of

[1] The term "lindane" should be distinguished from γ-HCH; γ-HCH is one of several HCH isomers, and is a component of technical HCH as well as of lindane. Lindane contains 99 % of pure γ-HCH.

[2] After year 2002, the regional groundwater flow situation changed in consequence of the flooded open pit mine Goitzsche and caused therewith an adjustment of the local site management.

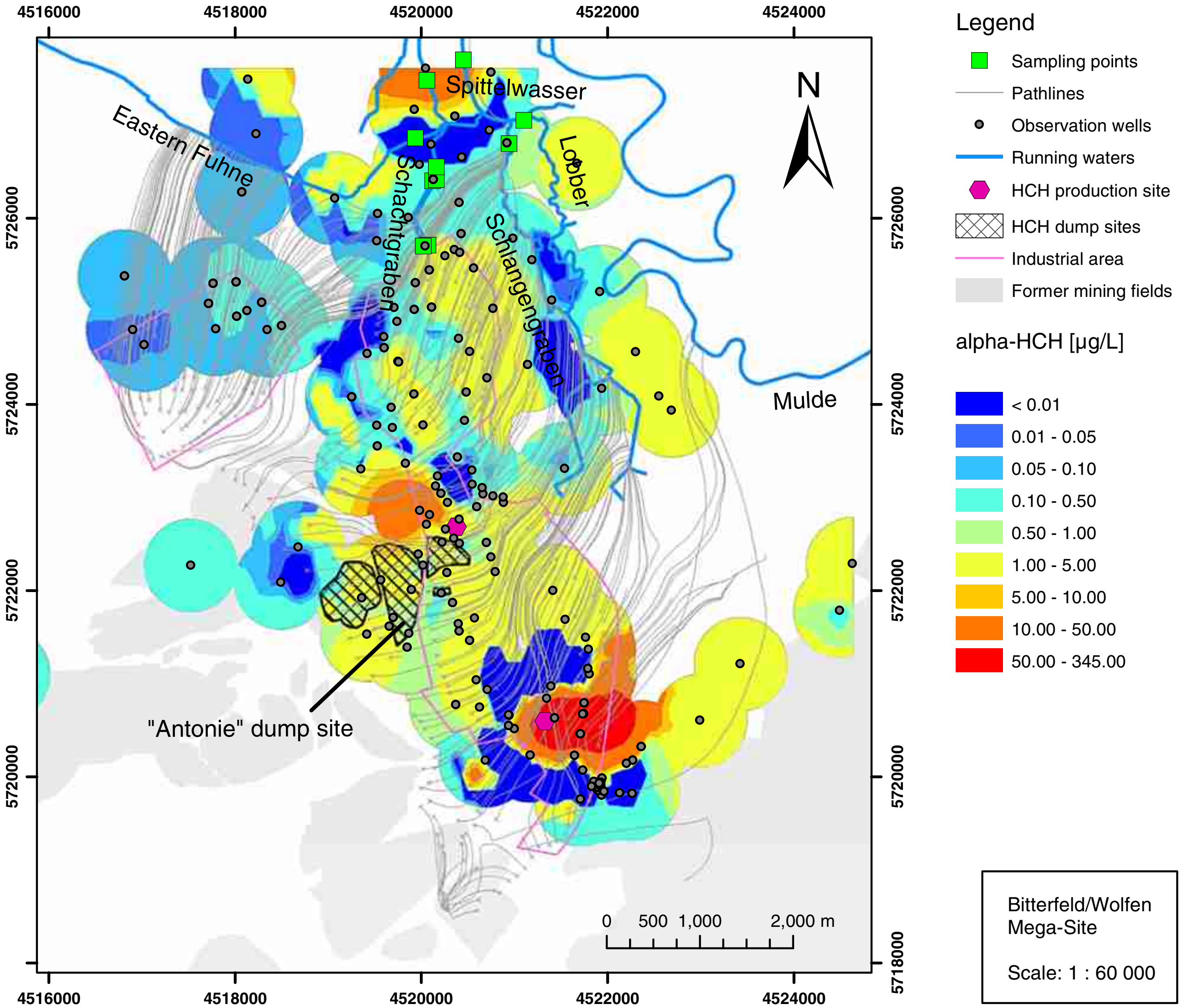

Fig. 1 Regionalised α-HCH distribution pattern within the upper Quaternary aquifer of the Bitterfeld/Wolfen region. Pathlines represent the regional groundwater flow situation 2005 and indicate a dominating northwards aligned groundwater flow direction. They do not consider the impacts of the installed hydraulic measures. The contaminant distribution was processed by using the ordinary kriging algorithm

appropriate remediation strategies which are appraised for the "Antonie" landfill and its impacts to about 700–2,000 million euros (Großmann and Kerndorff 2006).

In the following, this case study reflects exemplarily an integrated methodology for assessing the current situation of the large-scale groundwater contamination of the Bitterfeld/Wolfen mega-site. Besides multiple field sampling campaigns (1), this approach involves geo-statistical techniques for the regionalisation of nodal analytical data to create a conceptual model of the present contaminant distribution patterns (2). The model conception of contaminant distributions is consolidated and verified by 3-D aquifer geometries received from high-resolution geological 3-D modelling (3). As the spatial contaminant patterns had been affected significantly by long-term mining-related groundwater abstraction, 3-D numerical groundwater flow modelling was used to reproduce the regional dynamics of the prior groundwater flow/solute transport (4). Finally, intermediate results were integrated to analyse the current situation of pollution and to indicate model-based future fate scenarios of groundwater contaminants.

Material and methods

Regional groundwater monitoring data of about 10 years were analysed, containing approximately 3,500 samples and up to 180 individual organic parameters from almost 250

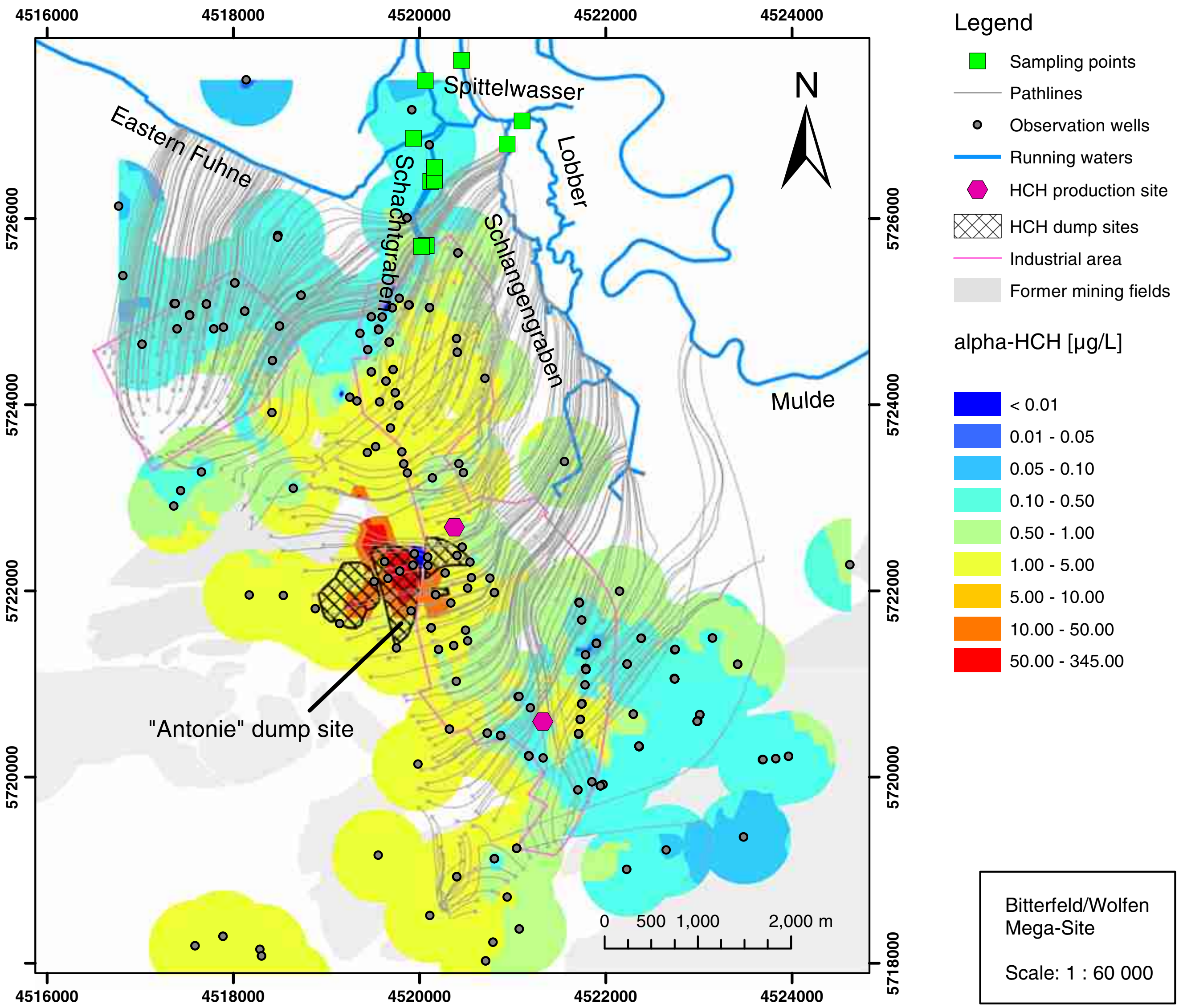

Fig. 2 Regionalised α-HCH distribution pattern within the lower Tertiary aquifer of the Bitterfeld/Wolfen region. Pathlines represent the regional groundwater flow situation 2005 and indicate a dominating northwards aligned groundwater flow direction. They do not consider the impacts of the installed hydraulic measures

observation wells from the nation SAFIRA project (Weiss et al. 2004). Furthermore, run-off measurements as well as water samples were taken biweekly from small creeks during a period of 18 months.

Since groundwater analytics only provide nodal information about the contaminant distribution within the hydrogeological system, interpolation procedures were used to create continuous distribution patterns for selected pollutants. Using GIS integrated interpolation procedures, contaminant distribution patterns were created for all HCH isomers as well as DDT/D/E isomers, separately for the upper and lower aquifer system. This regionalisation procedure is based on established regionalisation algorithms such as inversed distance weighting and different types of the geostatistical kriging technique. Best results were obtained from the ordinary kriging approach. After performing the kriging interpolation, a 500-m buffer was placed around each individual sampling point, of more than 200 groundwater observation wells, for avoiding extrapolation artefacts. Subsequently, the interpolated pollutant distribution patterns were analysed concerning their spatial dimensions which is enabling an estimation of the affected area of the Bitterfeld/Wolfen region.

Moreover, the software system GSI3D© (former GeoObject) has been used to generate a 45-km^2-wide, geological high-resolution 3-D model of the entire industrial region Bitterfeld/Wolfen and their surrounding downstream area (Steinmetz 2007; Wycisk et al. 2009). This geometric model has been incorporated subsequently into a numerical groundwater model. Using the finite

Table 1 Descriptive statistics of surface and groundwater samples

	HCH (alpha)	HCH (beta)	HCH (gamma)	HCH (delta)	DDT (op)	DDD (op)	DDE (op)
Surface water: Schlangengraben							
Samples	27	11	9	8	2	4	1
Max	0.035	0.389	0.034	0.061	0.004	0.002	0.000
Median	0.010	0.074	0.009	0.021	0.002	0.001	0.000
Standard deviation	0.008	0.124	0.011	0.020	0.002	0.001	0.000
Surface water: Lobber							
Samples	26	10	8	7	0	4	1
Max	0.087	0.130	0.129	0.085	n.d.	0.002	0.000
Median	0.013	0.049	0.012	0.013	n.d.	0.001	0.000
Standard deviation	0.019	0.039	0.054	0.028	n.d.	0.001	n.d.
Quaternary aquifer							
Count	171	171	171	171	123	127	127
Max	345.750	124.000	414.390	348.330	13.35	0.590	0.470
Median	0.265	0.130	0.175	0.240	0.025	0.021	0.017
Standard deviation	35.126	13.000	42.439	34.120	1.608	0.064	0.051
Tertiary aquifer							
Count	165	165	165	165	134	152	152
Max	525.000	91.500	125.000	545.000	528.000	71.400	4.052
Median	0.320	0.055	0.174	0.430	0.025	0.025	0.025
Standard deviation	40.824	7.125	9.752	42.471	45.607	5.800	0.330

Mean values were calculated for the aquifer characteristics when multiple measurements have been available at single well locations (contaminant concentration is given in micrograms per litre)

n.d. not detected

element model FEFLOW© for simulating groundwater flow and solute transport processes, the entire model domain of the regional flow/transport model amounts 330 km^2 and is structured into 13 individual hydrogeological subformations. Mainly, the model structure includes the upper Quaternary aquifer horizons, an underlying regional aquitard that is consisting of lignite seams and accompanying less conductive sediments such as clay and silt, and the lower Tertiary aquifer units (supporting information, Fig. 3). The Rupelian Silt, a massy aquitard of supra-regional scale, closes the vertical model structure downwards. Numerical flow model boundary conditions were derived from the surrounding surface water system as well as from standing water bodies within the model domain. Those data, preprocessed and stored in a GIS system, were implemented in the 3-D numerical model to represent the regional flow and solute transport dynamics before and after the flood event in August 2002. The successful model calibration process ensured a representative simulation of local piezometer dynamics (Gossel et al. 2009). Based on numerical modelling, groundwater flow and conservative solute transport simulations were carried out to analyse the regional solute transport dynamics and to give pollutant future fate indications.

Results

3-D site assessment study

Groundwater contamination

In order to characterise the spatial dimension of the regional groundwater pollution as well as to suggest selected substances which should be investigated in more detail, organic compounds were ranked regarding their risk potential (supporting information, Table 1). The database of the Bitterfeld/Wolfen SAFIRA Project provides regional groundwater monitoring data of more than 10 years. Approximately 3,500 samples and up to 180 individual organic parameters of about 250 observation wells were available for analysis (Wycisk et al. 2005; Weiss et al. 2002).

According to this study, groundwater monitoring data of HCH and DDT/D/E pesticide isomers were taken into account for statistical analyses (Table 1). The occurrence of pesticide isomers in the environment is related to prior local production and long-term discharge from multiple dump sites.

In 1998, an independent, land use-related classification has been elaborated to identify the spatial distribution patterns of the observed HCH and DDT/D/E isomers. Mean values of the sampling campaign 1998 were compiled and

Table 2 Contents (in micrograms per kilogram) of HCH and DDT isomers from a vertical soil profile at the Spittelwasser location (Schwartz et al. 2006)

Depth (m)	α-HCH	β-HCH	γ-HCH	δ-HCH	DDE (op)	DDE (pp)	DDD (op)	DDT (op)+DDD (pp)	DDT (pp)
0.0–0.10	440.0	702.5	23.3	10.3	66.6	200.5	19.1	714.9	698.0
0.10–0.20	535.6	574.3	9.5	b.d.l.	1.6	11.7	24.8	187.0	43.8
0.20–0.30	109.0	60.1	0.4	b.d.l.	0.1	0.3	0.1	1.5	0.4
0.30–0.60	0.5	6.4	0.3	b.d.l.	b.d.l.	b.d.l.	b.d.l.	b.d.l.	b.d.l.
0.60–0.80	0.4	2.4	0.1	b.d.l.	b.d.l.	0.1	0.2	3.9	b.d.l.
0.80–1.00	0.2	7.0	b.d.l.	b.d.l.	b.d.l.	b.d.l.	b.d.l.	b.d.l.	b.d.l.

These isomers are originated from the industrial process and are typically enriched in the upper sediment/soil profiles

b.d.l. below detection limit

included 222 readings for HCH and 166 for DDT/D/E isomers (Wycisk et al. 2002, 2003; Thieken 2002).

(a) Industrial and mining areas (HCH 2.6 μg/L; DDT/D/E 0.3 μg/L),
(b) Urban areas (HCH 2.0 μg/L; DDT/D/E 0.2 μg/L),
(c) Agricultural areas (HCH not detected, DDT/D/E not detected) and
(d) Alluvial plain of the Mulde River (HCH 1.8 μg/L; DDT/D/E 0.2 μg/L).

In addition, descriptive statistical data of α-, β-, γ-, δ-HCH and DDT/D/E isomers is given in Table 1, which was recorded from the very beginning of the regional groundwater monitoring in 1991 until 2001.

To some extent the HCH mean values from Table 1 show higher concentration levels than from the recorded data of the survey in 1998 above. In contrast, the range of the DDT/D/E mean values from the 1998 survey campaign and the groundwater monitoring (1991–2001) is quite similar. Thus, data from a long-term monitoring provide much more information about the contamination characteristics than a single survey and suppress the over- or underestimation of the polluted system.

Surface water contamination

Detailed investigations have been carried out on the interaction of contaminants in groundwater and surface water stream beds (Kalbus et al. 2008). The local surface waters Schlangengraben and Lobber are hydraulically well coupled to the groundwater system and indicate effluent conditions. Run-off measurements as well as water samples were taken biweekly from these streams (location see Fig. 1) which characterise tributaries towards the Spittelwasser and the Mulde River.

The surrounding 60-km^2 wide lowland, which is dewatered by the Spittelwasser and its adjacent creeks, contains approximately 20,000 tons of sediment which is heavily polluted by toxic compounds (Schwartz et al. 2006). Major problems here are the remobilisation of contaminated river sediments. Exemplarily, HCH concentration values of a vertical sediment profile of the Spittelwasser area are given in the following Table 2.

Concentration levels of the surface waters Lobber and Schlangengraben are listed in Table 1. The maximum values of HCH isomers of the Schlangengraben range from 0.03 to 0.39 μg/L but are based on a single short-term survey of a few months. A more detailed monitoring programme is in progress.

The sampling locations Schlangengraben and Lobber are located in the downstream of the industrial area Bitterfeld-Wolfen, in a lateral distance to the major contamination area (deposit Antonie and former production locations; see Fig. 1) of approximately 3–4 km. Therewith, a regional groundwater flow and transport modelling focuses on the annually load dynamics from the effluent groundwater into the local surface waters.

Additionally, statistical cross-correlations had been performed on surface water analytics and precipitation data. A direct correlation has been found for the Schachtgraben stream (see Fig. 1) between increasing HCH concentrations levels and precipitation; maximum concentration of alpha-HCH=27 μg/L, beta-HCH=13 μg/L. Increased concentration rates occurred 36 to 48 h after a rainfall event as a result of leaching and erosion processes of contaminated soil material (Neumann 2006). During a rain event, fine-grained soil sediment that contains HCH particles was eroded and transferred into the surface water system. In consequence, being partially dissolved in water, HCH isomers accumulate in the stream bed sediment. As a temporal securing measure, the contaminated soil sediments near surface waters have been covered with non-contaminated soil material.

Regionalisation of contaminants

The long-term HCH contaminant discharge originated from disposal(s) and contaminated soils into the upper (Quaternary) and lower (Tertiary) aquifer and was spread due to the regional dominating groundwater flow dynamics. Local hydraulic measures have been established to suppress a further spacious contaminant discharge into the local aquifer system.

To characterise pollutant sources, the release history and the future fate, in a first step, an aquifer-related regionalisation of the analytical data from regional groundwater monitoring is necessary for any further assessment of contamination characteristics or chronological development. Here, a reliable regionalisation of groundwater monitoring data constitutes a specific component of the integrated assessment study.

The long-term contaminant discharge from multiple contaminant sources was significantly influenced by mining-induced changes of the regional groundwater system. Regional groundwater drawdown in former open-pit mining fields, for ensuring a safe mining operation, leads to extensive changes of pre-existing groundwater levels, groundwater flow directions and gradients. Thus, long-term contaminant release from deposits and local production areas was influenced by various mining-related changes of the hydraulic system and formed the presently observed contaminant distribution patterns with all their complexity. Multiple HCH sources lead to high complex distribution patterns at a regional scale that are not comparable to single-source plume geometries. Therefore, a clear source localisation is complicated and comprises various probable input scenarios.

Three main areas of high HCH concentration levels (hot spots) could be outlined by the regionalisation procedure: The main HCH deposit Antonie, former HCH production areas and the northern located Spittelwasser tributary with its adjacent creeks (Fig. 1).

Different contaminant distribution patterns were found for the Quaternary (upper unit, Fig. 1) and the Tertiary (lower unit, Fig. 2) aquifer, which are caused by their geological bedding conditions and hydrogeological characteristics. Here, the (hydro)geological setting of the upper aquifer is more heterogeneous than of the lower aquifer system (supporting information, Fig. 3) which is also indicated by the diversity of related contaminant distribution patterns (Figs. 1 and 2). These contaminant distribution patterns imply some correlation between high α-HCH concentration levels and the vicinity of disposals and production sites. Moreover, those patterns illustrate the regional scale of the groundwater contamination. Only to some extent, the patterns show a systematic concentration decrease in downstream direction.

A GIS-based classification of the α-HCH regionalisation enables a quantitative estimation of the overall affected area of the region. Here, analysed for the Quaternary aquifer, a total area of about 38 km^2 is affected. A more detailed classification related to specific background concentration values is given in Table 3.

Geological and hydrogeological 3-D modelling

Beside the visualisation purpose, a geometric geological 3-D model enables volumetric calculations of individual or partial sediment bodies such as the remaining lignite volume or whole aquifer volumes. These estimations are highly relevant for assessing the natural attenuation potential as well as retardation processes of the subsurface layers.

Furthermore, the 3-D geometrics provide geometric data for the structural setup of a numerical model. Subsequently, a numerical finite element model has been established (Gossel et al. 2009) which involves the following objectives:

1. A transient pathline generation to identify potential contaminant pathways originated from the production areas and the identification of affected observation wells or surface water bodies after the end of active lignite mining (see Figs. 1 and 2).
2. Predictive calculations of the meanwhile changed hydraulic situation during and after the flooding event of the Goitzsche Lake in August 2002 (Wycisk et al. 2004).

Moreover, the numerical groundwater model handles the mining-induced changes of the former time period 1894 to 2006. The model reflects the long-term mining activities and their related groundwater withdrawal/groundwater rise and considers additionally the Mulde flood event in 2002. The numerical model does not reflect the effects of the recently installed hydraulic measures/emission systems.

Table 3 Amounted areas (in square kilometres) were evaluated from a GIS-based/geostatistical regionalisation procedure for each HCH isomer

Concentration (μg/L)	α-HCH (km^2)	β-HCH (km^2)	γ-HCH (km^2)	δ-HCH (km^2)
Class 1 (≤0.1)	8.51	4.58	11.99	3.58
Class 2 (≤0.5)	7.70	9.26	20.77	18.03
Class 3 (≤1.0)	4.39	7.68	2.78	4.51
Class 4 (≤10.0)	1.79	10.94	0.91	6.79
Class 5 (≤max. value)	1.94	1.79	0.92	1.35
	24.33	27.34	37.37	34.26

Within the Quaternary aquifer, an area of more than 37 km^2 is affected by the HCH (gamma) isomer. The regionalisation included 391 samples from the upper aquifer system

Only diffusion and dispersion parameters have been implemented first to study the local hydraulic and dominating transport characteristics. As the solute transport modelling in this stage is only considered conservatively, it is intended to implement also reactive solute transport modelling which will lead to more comprehensible and reliable predictions about the future fate development of contaminants. Therefore, further investigations are required to characterise the dominating reaction kinetics representatively, since the interaction of more than 1,000 individual chemical substances and their influence on specific reaction kinetics is not revealed in detail yet.

For the numerical model setup, the GIS-based data structure was of great advantage in many aspects of data pre- and post-processing: (1) to prepare the geometric model structure, (2) the setup of hydrogeological parameters/boundary condition databases, (3) the calibration process by comparing simulated results with measured data, (4) to verify the model setup and (5) to visualise model related data as well as simulation results. Thus, a full integration process of geological field data, high-resolution 3-D geology, flow and transport modelling and future fate contaminant scenarios has been done successfully. According to the study site, the simulation of groundwater flow and transport is of great importance for any source–receptor-related environmental assessment study to understand the groundwater-driven transport processes in history, before and after the flooding event in 2002 as well as in the future (Wycisk et al. 2004).

Currently the development and integration of forensic numerical modelling techniques are running, which focuses the estimation of pollution impacts from presently known origins or to reveal even unknown contamination sources. Moreover, this work also aims the implementation of reactive representation of solute transport processes for site assessment.

Discussion

As already mentioned, high HCH concentration levels have been revealed from the regionalisation procedure close to the Antonie deposit, the former production locations and in proximity of the Spittelwasser creek. In the vicinity of the southern production site, only the upper aquifer (Quaternary) is highly affected by all four isomers which implies that large amounts of HCH were transferred into the soil/subsurface during production or from storage. In general, the HCH contamination is covering an area of about 40 km^2 and is therefore of regional scale. Predominant contaminant discharge along high hydraulic conductivity zones (supporting information, Fig. 3) and the aeolian transport in combination with surface erosion processes are considered to be the dominating transport mechanisms. Groundwater flow velocity fields (gradients) were additionally accelerated by mining related groundwater withdrawal. In areas where subglacial channels notch the lower (Tertiary) aquifer system (Fig. 1, southern HCH hot spot), the aquitard is missing and enables a vertical exchange between the upper and lower aquifer unit. Limiting factors of solute transport are the high sorption potential of the lignite seam complex and the relative low water solubility of the HCH isomers (Table 4).

Table 4 Solubility (in milligrams per litre) of HCH isomers in water at 20°C (Eichler 1983)

Water solubility (mg/L)	α-HCH	β-HCH	γ-HCH	δ-HCH
	1.5	0,2	6.0	9.0

As chemical and biological transformation reactions need to be considered to assess the regional transport dynamics more in detail, solute transport was implemented in this study only conservatively since respective reaction kinetics and parameters of the study site are not understood clearly enough or available yet. Especially the presence of hundreds to thousands individual organic compounds and the unknown interaction among each other complicate a clear differentiation of individual substance-specific reaction kinetics.

In the surrounding of the Antonie disposal, high HCH concentration levels are observed in the Quaternary unit and the lower Tertiary horizon. While alpha- beta-, and delta-HCH isomers are observed at high levels in both aquifers, the gamma-HCH isomer is not present in the upper Quaternary layer but is observed in the Tertiary system below the dump site Antonie (supporting information, Fig. 4). This indicates that the major amount of the relatively high water-soluble gamma-HCH (6 mg/L) has already been extracted from the water accessible areas of the deposit over the last decades and was already transferred into the deeper Tertiary aquifer. Obviously, since that point of time, a measurable gamma-HCH discharge into the upper aquifer does not occur anymore.

The local HCH contamination around the Spittelwasser mainly is concentrated to the floodplain area and the surrounding area. Most probably, this contamination originated from the release of industrial waste water that was discharged via the Spittelwasser for the four decades of local HCH production. The most water-soluble HCH isomers (Table 4)—gamma-HCH (6 mg/L) and delta-HCH (9 mg/L)—have not been detected in the sediments which implies washout or degradation processes.

Again, the assessment of the Bitterfeld area has shown that zones of high HCH contamination mostly occur in close distance to their production sites. The groundwater pollution of the former industrial region has been analysed related to its input areas such as deposits, production location(s) and wastewater release.

Generally it has to be assumed that at most lindane production sites worldwide, HCH isomers from production locations, waste dumps, landfills or contaminated soils are spread via surface waters and/or groundwater for long time scales and large distances. This assumption is based on the facts that (1) the related HCH waste at such deposits is in the scale of thousands of tons and therefore represents a large contaminant reservoir (Vijgen 2006; Vijgen et al. 2010), (2) chemical production plants are often situated near rivers, (3) at production sites in (sub-)tropical countries[3] the major sediment transport occurs during the rainy season that is mobilizing diffuse contaminant sources, (4) meanwhile also in Europe an increasing number of flood events lead to an intensified POPs discharge into rivers which is expected to increase in future due to climate change (Wölz et al. 2008; Weber et al. 2011) and (5) large amounts of groundwater are often abstracted around those production sites for generating process water or by pump and treat systems for securing hazardous deposits. Vijgen (2006) and Vijgen et al. (2010) state with respect to the numerous existing HCH waste disposals worldwide, being classified by Weber et al. (2008a, 2011) as POPs waste deposits, that a successful implementation of the Stockholm Convention requires a detailed site investigation of the HCH/POPs waste deposits and their pollutant releases for an adequate management of related impacts on human health and ecosystem integrity.

For the Bitterfeld site, HCH and other organochlorine releases into groundwater, surface water and atmosphere have been documented. It has been concluded for this site that traditional site assessment methods based on “simple” groundwater and soil sampling oversimplify the system and underestimate the contaminant transport and related risk dynamics. The individual size and the complexity of the contaminant discharge at such mega-sites often lead to refusals or failures of remediation measures focused on source/groundwater treatment. Thus, for creating a solid decision-making basis, these multifaceted settings require a comprehensive approach for site investigation, characterisation, risk assessment and site management as well as for the quality assurance of all measures involved. A detailed knowledge about the subsurface architecture as well as of the tempo-spatial development of surface and subsurface flow, the transport dynamics and the final estimation of related pollutant release is essential for an appropriate and sustainable management of such contaminated mega-sites. Exemplarily developed for the Bitterfeld site, results from the numerical modelling and the analysis of dynamic flow fields provide relevant information for supporting the decision-making process on remediation strategies.

[3] e.g. Brazil (Torres et al. 2010), India (Abhilash and Singh 2009; Jit et al. 2011) or South Africa (Vijgen 2006)

Conclusion

Multiple approaches of, e.g. a high-resolution 3-D subsurface model, groundwater and surface water monitoring, regionalisation techniques, as well as a numerical groundwater flow and transport modelling have been applied and combined in a first stage to characterise the dimension of the HCH contamination and to understand contaminant history by an isomer-specific interpretation. Simulation results can be visualised and analysed in GIS or 3-D geological/hydrogeological model representations to receive detailed information about contaminated layers, potential pollutant traps of dense non-aqueous phase liquids, additional adsorption facilities and ongoing discharge of HCH and other pollutants. From these points of view, a detailed geological 3-D model carries more importance for environmental risk assessment studies than only providing the structural data input for numerical model creation. Prospectively, more detailed reactive transport modelling will lead to an appropriate reproduction of the presently observed distribution patterns. In consequence, this would verify the numerical model to give more specific future fate predictions of pollutants and enable the calculation of mass fluxes into environmental compartments.

Contaminated mega-sites that are characterised by complex aquifers settings and a groundwater contamination at a regional scale highly require such a three-dimensional model approach which is able to integrate all the tempo-spatial information and to query and combine them for reliable process-orientated environmental assessment.

The regional multi-species groundwater pollution of the former HCH/organochlorine production site Bitterfeld/Wolfen emphasise the necessity to incorporate the complete contaminant spectrum for an appropriate risk assessment as a sustainable basis for efficient remediation decisions! The integrated methodology applied here can be transferred to other HCH or otherwise polluted sites as a supporting instrument for the decision-making process on site-specific remediation concepts.

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