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Prediction of diffuse sulfate emissions from a former mining district and associated groundwater discharges to surface waters

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SUMMARY

Rivers draining mining districts are often affected by the diffuse input of polluted groundwaters. The severity and longevity of the impact depends on a wide range of factors such as the source terms, the hydraulic regime, the distance between pollutant sources and discharge points and the dilution by discharge from upstream river reaches. In this study a deterministic multi-mine life-cycle model was developed. It is used to characterize pollutant sources and to quantify the resulting current and future effects on both groundwater and river water quality. Thereby sulfate acts as proxy for mining-related impacts. The model application to the Lausitz mining district (Germany) shows that the most important factors controlling concentrations and discharge of sulfate are mixing/dilution with ambient groundwater and the rates of biological sulfate reduction during subsurface transport. In contrast, future impacts originating from the unsaturated zones of the mining dumps showed to be of little importance due to the high age of the mining dumps and the associated depletion in reactive iron-sulfides. The simulations indicate that currently the groundwater borne diffuse input of sulfate into the rivers Kleine Spree and Spree is ~2200 t/years. Our predictions suggest a future increase to ~11,000 t/years within the next 40 years. Depending on river discharge rates this represents an increase in sulfate concentration of 40–300 mg/L. A trend reversal for the surface water discharge is not expected before 2050.

1. Introduction

The Lausitz mining district is the largest open pit lignite mining area in Germany and one of the largest in Europe. It was intensely mined for more than a century whereby the highest annual production of lignite (~200 Mt) was reached in the 1980s. Following the reunification of Germany in 1990 most mines were closed and only 4 lignite mines are still operating. In these areas the oxidation of sulfide bearing minerals during and after mining causes the mining dumps to act as long-term pollutant source (Evangelou, 1995). Until the beginning of this Millennium the groundwater drawdown that was originally required for the operation of the open pit mines has largely prevented the discharge of groundwater from the mines and thus the generation of mine drainage. However, following the closure of the mines, groundwater tables in these areas are successively rising. Where natural groundwater flow conditions are re-established, which often requires more than a decade, polluted groundwaters migrate towards their respective surface water discharge points. The temporally and spatially varying contaminant concentrations in the groundwater as well as the evolution of the overall mass discharge to the surface waters depend on the local emissions from the mining dumps, the changing hydraulic conditions, the degree of dilution and the biogeochemical processes within the plume (Hoth et al., 2001; Kohfahl and Pekdeger, 2004; Moncur et al., 2005; Rolland et al., 2001; Salmon and Malmström, 2004; Wisotzky and Obermann, 2001; Younger, 2001). Over the past 20 years groundwater observation wells within the Lausitz district demonstrate increasing concentration of sulfate, iron, aluminum and heavy metals downstream of selected mining dumps (Bilek, 2006; Graupner et al., 2012; Kohfahl and Pekdeger, 2004, 2006; Rolland et al., 2001; Werner et al., 2001). Eventually the increasingly polluted groundwater can cause a wide range of public health, environmental, and facility safety issues.

In conjunction with the rising groundwater levels the rivers within the mining area successively regain their function as discharge features for the regional aquifers. Where mine drainage

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reaches the river banks it can deteriorate surface water quality substantially. Detrimental effects such as increasing acidity, high sulfate and iron concentrations and subsequent sudden fish mortality were already observed at one of the tributaries of the Spree River, the Kleine Spree River, which crosses a district adjacent to mining dumps. After passing the mining dumps sulfate concentrations were shown to increase by up to 220 mg/L, depending on discharge rates, while the corresponding pH values declined 1 unit. The first deteriorating effects resulting from diffuse inputs into the Spree River, the largest river crossing the district, have currently been observed. However, a notable impact resulting from mine drainage is expected for the coming years. While elevated sulfate concentrations in rivers are not known to be toxic, they may inhibit plant growth (Davies, 2007). More significant are the concerns regarding the impacts on the security of public drinking water supplies and the additional treatment costs given that a substantial fraction of the drinking water of Berlin (~60%) (Möller and Burgschweiger, 2008) is provided through bank filtration from the Spree River.

Legal frameworks such as the EU Water Framework Directive (WFD) (EU-WFD, 2000) enforce the implementation of a monitoring system for groundwater and for surface water bodies to provide water quality data in mining districts. However, initially these data provide only information on pollutant concentrations that cannot be used directly to infer future concentrations and mass fluxes to determine the future impacts on surface water bodies. Therefore, model simulations that assess the current situation and predict the future evolution of the groundwater quality and its impact on surface waters are an important and necessary step for the implementation of the EU WFD (Hojberg et al., 2007; Jorgensen et al., 2007). This is especially the case in catchments with complex and widespread pollutant source zones like mining districts.

Several previous studies investigated and reported current mine drainage impacts on river water quality (Broshears et al., 1996; Chen et al., 1999; Gozzard et al., 2011; Kim and Chon, 2001; Kimball et al., 2010; Mayes et al., 2010; Mighanetara et al., 2009; Nyamadzawo et al., 2007; Olías et al., 2004, 2006; Raymond and Oh, 2009; Sarmiento et al., 2011; Sracek et al., 2012). For example, the AMD impact on the Odiel river basin (Spain) and the consequences for the water quality were monitored over 5 years (Olías et al., 2006) and the data were used to estimate the current annual metal fluxes that are discharging into the ocean. For the same river basin water samples from a single year in combination with statistical methods were used to (i) identify and characterize the main sources of AMD and (ii) seasonal effects of AMD on river water quality (Olías et al., 2004). In conjunction with a sediment characterization the data were also used to describe how AMD might alter the physical, chemical and biological characteristics of a natural stream (Olıas et al., 2004; Sarmiento et al., 2011; Sracek et al., 2012). A similar study, complimented by experiments, was carried out for the Animas river (USA) (Kimball et al., 2010) and for the Imgok Creek (South Korea) (Kim and Chon, 2001), while for the river Tamar catchment contaminant fluxes from point and diffuse sources were estimated from surveys for the years 2005 and 2006 (Mighanetara et al., 2009). Furthermore, element fluxes of the last 100 years were estimated for 3 watersheds in Pennsylvania (USA) based on long time monitoring data of river discharges and its associated water quality. The results were linked with the mining activities (Raymond and Oh, 2009). For the Beaver Creek watershed (USA) a detailed study investigated the development of the river water quality over a 7-year period 20 years after mine reclamation. The study used statistical methods to investigate the impact of AMD on land use. A similar study was carried out for the Yellow Jacket River (South Africa) 2 years after mine site remediation, based on monitoring data (Nyamadzawo et al., 2007). For the St. Kevin Gulch water quality observations were used to model the chemical reactions within the river and the interactions with the river bed (Broshears et al., 1996). The impact of AMD on the Almond catchment (Great Britain) was investigated based on estimates of pyrite oxidation rates and the subsequent development of the corresponding mining affected groundwater composition. Based on these data the impact of groundwater discharges on the hydrochemical composition of the river water was modeled with PHREEQC (Chen et al., 1999). For the West Allen catchment (Great Britain) the current point and diffuse sources of AMD were monitored for different run-off conditions and the corresponding cumulative zinc loads (Gozzard et al., 2011). Attempts were made to estimate the total pollution burden from abandoned non-coal mines on rivers in England and Wales, using data collected over the last 100 years at various discharge zones of the most important abandoned mines within these regions (Mayes et al., 2010). All these studies remained restricted to descriptions of the past or current status, based on monitoring data and experiments. Where predictions of the future river water quality over longer periods were made, they were generally not constrained by mass-balance considerations for the source terms and did not take into account the physico-chemical processes that control the long-term evolution of groundwater discharges to surface waters.

In contrast to the aforementioned studies, for the present investigations we developed and tested a deterministic model framework for simulating the pollution pathways within the groundwater system on a regional scale. We use a reactive transport modeling approach to track the pollutant fate and mass balances within the sources, i.e., the mining dumps, as well as their passage across a time variant groundwater regime towards the Kleine Spree River and the Spree River. With this approach we resolve the individual effects of single mining dumps and, more importantly also the cumulative effect within the study area. As in many previous studies (Canovas et al., 2007; Chen et al., 1999; Kim and Chon, 2001; Sarmiento et al., 2009; Schemel et al., 2006), we use sulfate as an indicator for mining induced contamination. Sulfate, while migrating at faster rates compared to acidity and, e.g., trace metals, is generally considered as a useful proxy for AMD and has the advantage that it was and is included in essentially all monitoring programs within the study site and elsewhere in AMD-affected regions. Based on the developed model we predict the evolution of the groundwater quality in the mining area and the mass discharges into the river system until 2100.

2. Material and methods

2.1. Study site

The area investigated in this study is part of the former open pit lignite mining district Lausitz. It is located in the Eastern part of Germany about 100 km south of Berlin. The district has been mined since 1940, with a climax in the 1980s. The mining activities resulted in large dumps of deposited quaternary and tertiary overburden material. Dewatering has caused a substantial groundwater drawdown affecting an area of approximately 1350 km^2 in 1990 and leading to a groundwater deficit of about 7 billion m^3. Meanwhile, rising groundwater levels have eliminated more than 80% of the groundwater deficit, accompanied by the formation of an extensive pit lake system with over 100 new lakes until 2020. With a total area of 170 km^2 the mining district will become the largest artificial lake system in Europe.

Our study site is situated in the South-Eastern part of the district and it comprises an area of about 540 km^2. The study site includes 8 decommissioned mining dumps of different size that were expected to have the greatest influence on the water quality