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Review

Wastes in Underground Coal Mines and Their Behavior during Mine Water Level Rebound—A Review

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Abstract: Backfill materials of various origin and composition, abandoned machinery, oils, PCB, gallery support material and cables are the main wastes occurring in underground coal mines during the period of their abandonment. Bearing in mind that under increasing societal pressure most if not all underground coal mines are going to close sooner rather than later, it is important to understand the interactions of these waste materials with rising mine water during mine water level rebound to prevent adverse environmental effects, especially on surface and groundwater. To this end, the composition of mine water at decant points as well as the hydrogeochemical, temporal and spatial dynamics of mine water during rebound requires quantification. In the first part of this paper, an overview of waste materials in underground coal mines is presented. The second part focusses on the experiences gained in the Ruhr area, a closed underground coal mining region in western Germany, where mine water rebound has been ongoing for decades. In this regard, the mine water modeling program Boxmodell was applied during regulatory approval procedures to predict the hydrodynamics and hydrogeochemical development of the water rebound. The results of these investigations allow deep insights into the interactions of rising mine water with wastes as well as the complex chemical evolution of mine water and potentially occurring contaminants (e.g., PCB). The experiences regarding wastes in underground coal mines and the geochemical evolution of rising mine water gained in the Ruhr area can be utilized to support the planning of mine closure in currently still active underground coal mining areas worldwide.

Keywords: coal mine; waste material; Boxmodell; reactive transport; mine water level rebound

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1. Introduction

Mining is finite, yet the consequences of mining may be enduring if they are not addressed with the necessary diligence. Both the mining method and the extracted material may have adverse environmental consequences, e.g., when coal is used as a fuel. Coal mining and the use of coal are therefore considered unattractive from a wider societal point of view, even when they are economically attractive in some cases. Consequently, many scientists and policy makers consider a (global) termination of coal mining [1,2].

Currently, approximately 3200 active coal mines exist worldwide, and more than 50% of these are underground operations [3]. From the analysis of 97 closed underground and open-cut coal mines in Europe, Gombert et al. [4] conclude that the quantitative influence of coal mine water discharges is "substantial" and that "evaluating their environmental impact on surface and underground water is important". Therefore, this article intends to shed light on the processes during mine water level rebound in underground coal mines, whereby an emphasis is set on the interaction of mine water with waste materials. In this regard, the Ruhr area, a closed underground coal mining region in western Germany, provides an exceptionally well-documented example. The results of investigations related to this topic since the early 1980s until today are presented. In the Ruhr area, it is estimated that a total of ~70 Mm^3/a of mine water will be pumped and discharged into surface waters [5]. Here, mine water rebound is carried out in a controlled manner, i.e., to protect the drinking reservoir in the region. The importance of knowing the potential contaminant sources and understanding the transport processes involved to mitigate the environmental impact caused by mine water is underlined. Typical questions arising in this context are related to the duration of mine water level rebound, the flow volumes and geochemical evolution of mine water during the rising period and its final composition at decant points.

During operation, mine water is pumped to the surface and discharged, with or without treatment, into receiving waters. In the period of underground coal mine closure, mine water drainage systems are no longer required, pumping is stopped and, consequently, the mine water rises until it reaches a natural or a planned level, whereby the final goal will be to reach a chemically and physically stable state of mine water discharge into receiving waters. During this phase, mine water constitutes a potential link between wastes in underground coal mines and receiving surface waters or shallow groundwater.

For the prediction of mine water development, the mine water modeling software Boxmodell was applied in the Ruhr area. Further, the importance of water management concepts in the water rebound phase is highlighted, which effectively prevents harmful environmental effects. Notably, the quality of surface water in the Ruhr area has significantly improved over recent decades [6]. Achieving these outcomes requires an in-depth understanding of several pivotal aspects:

(1) The composition and quantity of waste materials within the underground mine(s).
(2) The extent and geometry of the underground workings and, in the case of neighboring mines, their connections.
(3) The transport mechanisms of various types of contaminants.
(4) The utilization of specialized mine water modeling software as predictive tools.

2. Waste Materials in Underground Coal Mines

Within the European Union (EU), mine wastes are defined as "[. . .] substance or object which the holder discards or intends or is required to discard" [7–9]. Usually, materials like fractured (waste) rock, topsoil or tailings are associated with this definition. Mine water is not explicitly mentioned as a waste in these directives. As described in Vogt [10], a court decision from 1992 determined that mine water is not classified as wastewater in Germany. Even the dissolution of contaminants does not make mine water wastewater (see also [11]). However, for the purpose of this article, mine water plays a central role since it may leach and transport contaminants from deposited wastes in underground coal mines. Further, mine water geochemistry may pose an environmental risk, regardless of the interaction with wastes, since possibly substantial amounts of mine water may need to be discharged into receiving waters or may infiltrate surface-near groundwater resources.

Wastes associated with coal mining are dealt with or are even the central theme in, e.g., Agboola et al. [12], Garbarino et al. [9], Lee [13] and Gombert et al. [4], amongst others. However, these publications do not specifically focus on wastes in underground coal mines. The study by Agboola et al. [12] discusses mine dump pollution monitoring and mine dump management strategies and is thus not focused on underground mining. Lee [13] points towards potential adverse economic effects of poor waste management and provides an example where a coal slurry leak into the Athabasca River in Canada caused severe damage associated with >100 million Euro clean-up costs. In Garbarino et al. [9], the main focus is set on the effect of waste material above ground, e.g., landscaping of waste rock piles or spontaneous combustion of waste rock piles. In addition, the quantity and potential for reuse of (coal mine) waste material is discussed. The document also mentions that wastes may influence the composition of mine water, whereby the authors introduce the term "Extractive Waste Influenced Water (EWIW)". In the proposed techniques to model emissions from wastes to water, a reference is provided which deals with mainly open-cast coal mines in South Africa [14]. An example for techniques applicable to underground coal mines is, however, lacking in [9].

In the Ruhr area in western Germany, underground coal mining ceased in 2018 after an approximately 150-year-long industrial extractive phase [5]. Due to the presence of surface-near groundwater, which is exploited as a drinking water supply, strict and complex regulations apply for a controlled mine water level rebound to a target level, approximately 150 m below the drinking water aquifer [5]. In order to obtain permission to let the mine water level rise to this particular target level, the responsible mining company RAG needed to submit extensive (modeling) reports to the competent authority, proving that the mine water will not contaminate the surface-near drinking water and that contaminants such as, e.g., PCB (polychlorinated biphenyls) are not discharged into receiving waters at unacceptable concentration levels. In order to ensure the highest level of transparency, the extensive reports submitted to the competent authority are publicly available and provide excellent documentation of waste materials contained in underground coal mines. The reports are described and cited in Section 3 in this paper.

In the following sections, an in-depth exploration of the primary waste materials and potential contamination sources in underground coal mining is provided. Several wastes and contamination sources, relevant during mine water level rebound following mine closure, are presented in Table 1. The compilation of Table 1 has been performed based on the personal experience of the authors and has to be regarded as nonexhaustive, since the occurrence of wastes in underground coal mines varies from case to case.

Table 1. Overview of waste material and potential contamination sources in underground coal mines.

Waste Type/Contamination Source	Types of Contamination
Exposed rock in shafts and galleries	AMD (acid mine drainage), metals
Fractured rock and residual coal in goaf areas, coal mine spoil	AMD, metals, coal fines and dust
Abandoned equipment and machinery	Oil, solvents and lubricants, PCB, PCDM (polychlorinated diphenylmethanes), plasticizers from the conveyor belts (mostly remained underground) and many more
Backfill materials of various origin, e.g.,: - Fly ash. - Coal processing wastes, tailings or slurry.	Metals, salts, etc.
Gallery supporting material/cables	Metals (Fe, Cu)
Mine water and EWIW	Salts (Cl, NO_3, SO_4, etc.), metals, radionuclides

2.1. Backfill Material

The deposition of mining waste material as backfill material in underground mines is a common worldwide practice. The reasons for backfilling are related to geotechnical concerns, e.g., to decrease ground subsidence effects [15,16]. In addition, backfill materials can be of other origins. In the Ruhr area, for example, waste materials from municipal waste combustion were used as backfill material next to wastes from flue gas desulphurization (see [17,18]). These materials are usually mixed with cement prior to deposition [19]. According to Szczepanska-Plewa et al. [20], the use of the extracted material in coal mining as backfill material can have an immediate and significant adverse impact on groundwater quality. This impact can be characterized by sulfide oxidation, sulphate generation and the leaching of metals and chloride, resulting in groundwater concentrations that often exceed locally specified maximum contaminant levels. In cases where insufficient or no documentation on deposited wastes exists, the risk magnitude cannot be directly assessed. The monitoring of water quality and quantity, however, can be helpful to distinguish the source of contamination (see [5,21]). The challenges posed by backfilling with waste material in underground coal mining continue to drive research and innovation in the field. Researchers are investigating novel techniques for waste management, including the development of backfill materials that are less prone to leaching harmful substances [22]. Additionally, advancements in monitoring and control systems enable real-time tracking of water quality and quantity changes, facilitating rapid response to any detected contamination.

2.2. Abondoned Waste Materials, Lubricants, Oil, PCB and PCDM

Lubricants and oils have been used in underground coal mines, as has diesel oil as fuel in the locomotives of most mines [23]. These materials play essential roles in powering machinery and ensuring the smooth operation of mining activities. Such mineral oil products, which may have leaked into the mining gallery ground, are, to a large extent, complex mixtures of mineral oil hydrocarbons with different properties. In particular, diesel oils have relevant water-soluble fractions. Water solubility of these hydrocarbons decreases with increasing molecule length.

In addition, hydraulic fluids in coal mines require attention as a relevant source of contamination due to their widespread use. Since the discovery of PCBs as nonflammable components in hydraulic fluids in the 1960s, they have been used extensively in underground coal mines, e.g., in hydraulic systems, transformers and gearboxes, amongst other materials. In the US, the production of PCBs was prohibited in 1978 by the Environmental Protection Authority (EPA) due to their toxic properties [24]. However, in mining, PCBs were still used in machines, which were then abandoned in underground mines. In Germany, the use of PCB in mining was discontinued in the mid-1980s, and instead, the PCB alternative PCDM was used beginning in 1984; however, this proved to have toxic properties similar to PCB. Finally, in the early 1990s, the application of PCDM entirely ceased [19].

The presence of lubricants, oils and PCB in underground mines poses a risk to surrounding ecosystems and therefore requires detailed investigation in relation to the substances' emission and spreading potential during mine water level rebound. PCBs, in particular, are notorious for their long-lasting toxic effects on wildlife and aquatic environments. These chemicals can accumulate in the food chain, leading to bioaccumulation and potential harm to organisms at higher trophic levels [25]. Additionally, the release of mineral oil hydrocarbons from underground mines, which may include water-soluble fractions, can lead to water pollution, adversely affecting aquatic life and local water quality. It is crucial to highlight that fractions insoluble in water can also be transported, potentially contributing to water pollution. In Section 3, an example is provided where, through diligent mine water management planning informed by predictive modeling and verified through monitoring, adverse effects on the environment were avoided.

2.3. Mine Water and Acid Mine Drainage

The naturally occurring material inside underground coal mines may cause mine water contamination, e.g., acid mine drainage (AMD). Coal deposits often contain sulfide minerals because the depositional environment favored the precipitation of these minerals [26]. Mining activity alters the reducing environment and brings sulfide minerals (e.g., pyrite) in contact with oxygen and water (oxidizing condition). The resulting oxidization of pyrite produces sulphate, iron and substantial amounts of acid when not buffered by carbonate rocks. A higher content of sulfide minerals in coal deposits results in higher concentrations of the potential pollutants, sulphate and iron [22]. The resulting AMD is a concern related to waste materials since acidic mine water has a higher potential to leach contaminants from waste material.

AMD has been reported for many coal mining areas worldwide; Wang et al. [22] reviewed acid mine drainage due to coal mining in the US, India, South Africa, China and Europe. Their review focuses on abandoned mines including underground and open-pit mining at small scale, i.e., isolated coal mines and large-scale mining regions such as the Shanxi Region in China with interconnected underground mines. A further case regarding AMD in underground coal mines is reported by Wood et al. [27] for abandoned mines in Scotland. Here, the authors conclude that AMD resulted in water contamination within the first decades after mine closure, whereas after 40 years of mine closure, water pH increased, and consequently, contaminant levels decreased. Lambert et al. [28] conclude that for underground coal mines in the Uniontown Syncline, Fayette County, PA, USA, the rate and degree of water quality improvement is dependent on the mine flooding rate, whereby mines with a high degree of flooding show a significantly higher degree and rate of water quality improvement. Abandoned coal mines in Shanxi, China, form an interconnected underground mining region where shafts and connections were not effectively blocked to prevent water exchange between deeper contaminated mine water and near-surface groundwater. This has resulted in a drinking water safety crisis [22]. Notably, for the underground mining regions in Germany, i.e., the Saar area and Ruhr area, AMD is not an issue because of sufficient carbonate content in the host rock [17].

3. Case Study: Wastes in the Ruhr Area—Germany

3.1. Underground Mining in the Ruhr Area

To shed light on the processes and prediction methods relevant to contaminant transport during mine water rebound, the underground mining region of the Ruhr area constitutes an exceptionally well-documented example [5,10,17,19,29–43]. The Ruhr area is located in North Rhine Westphalia in western Germany (see Figure 1). Approximately 5.1 million inhabitants live in the Ruhr area, which covers an area of 4439 km^2, resulting in ~1140 inhabitants/km^2, making it one of the most densely populated regions in Germany [44]. A recent description of water management challenges in the Ruhr area, together with an overview of mine water chemistry, geology and hydrogeological setting, is provided in Jasnowski-Peters and Melchers [5]. For better understanding, a brief overview of key aspects is presented here.

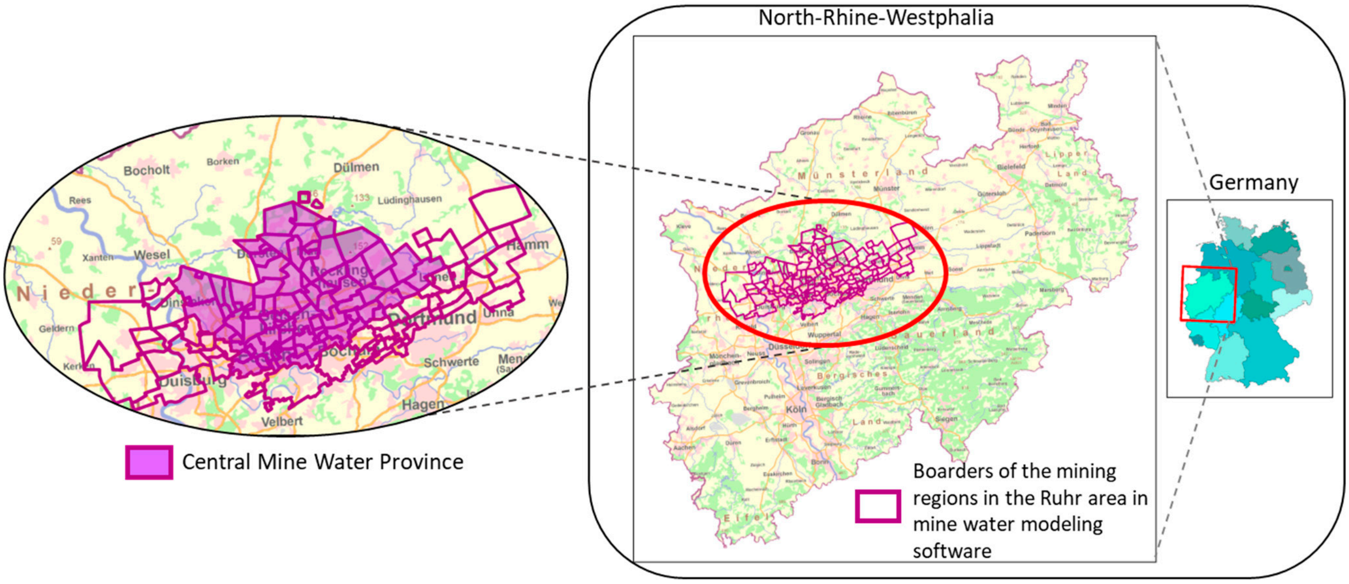

Figure 1. Location of the Ruhr area in North Rhine Westphalia and Ruhr mining region with boarders of the mining regions (volume elements) in the mine water modeling software Boxmodell. The Central Mine Water Province is highlighted on the left.

Almost the entire geographic extent of the Ruhr area has been mined in dozens of neighboring and interconnected underground coal mines. Coal mining finally ceased in 2018 with the closure of the last operating coal mine, Prosper Haniel. However, most coal mines closed in the 1960s [45]. Due to proximity of underground mines and hydraulic interconnectivity through roadways, water management had to be maintained in closed underground mine workings to prevent water intrusion into neighboring (active) mines. As a result, the Ruhr area has been subdivided into several so-called water management provinces with central water pumping stations. In 1969, 25 drainage systems were used to drain mine water and protect the active mines [43]. In the following years, as coal mine closure continued, the number of drainage systems was decreased and therewith optimized. The current water management plan for the Ruhr area envisages the use of six drainage installations for the entire Ruhr area (see Table 2) [10,18].

Table 2. Mine water provinces and pumping stations [6,36].

Main Water Province	Mine Water Pumping Stations in 2006	Mine Water Pumping Stations after Implementation of Mine Water Management Plan
West	West, Walsum, Concordia	Walsum
Central	Amalie, Emschermulde, Zollverein, Prosper-Haniel, Lohberg, Carolinenglück, Lippe, Auguste Victoria	Lohberg
East	Hansa, Haus Aden, East	Haus Aden
Ruhr	Robert Müser, Friedlicher Nachbar, Heinrich	Robert Müser, Friedlicher Nachbar, Heinrich

The mines in the Ruhr area are represented in a comprehensive numerical mine water model with hydraulically effective connections between the mines, their overburden and deep mine water tributaries. As simulator, the software Boxmodell, is used, which is based on the finite volume method [46]. Since voids (e.g., shafts and galleries) within a mine are hydraulically interconnected (hydraulic short-circuit system), the mines are usually discretized as one element. However, mines can also be divided into several elements if required or requested. The finite volume method offers very good flexibility especially for this. The Ruhr mine water model has a three-dimensional structure. The vertical node spacing is about 50 m. Both steady state and transient flow can be calculated. For mass and heat transport, only transient calculations apply.

In the numerical mine water model of the Ruhr area, all relevant input data such as pump performance curves, a complicated network of roadways, drifts and shafts, shortcuts between mining fields, hydraulic connections and many more properties can be considered. Moreover, it allows for the simulation of parallel multicomponent transport, modified sorption potential per transport unit, geochemical reactions and microbiological degradation.

Following the mine water model setup, it was calibrated with regard to both the mine water flow and the associated mass and heat transport. For this purpose, the mine water level measurement values available since 1988 were compared with the model calculation values. Calibration to the measured values was achieved by varying the model parameters, especially the conductance value. The same applies to mine water qualities and temperatures.

From a regulatory point of view, the measures related to a controlled mine water level rebound to implement the overarching water management plan require approval from authorities. To obtain approval, predictive water rebound modeling for individual coal mines and for certain interconnected, neighboring coal mines needed to be prepared. In addition, the complex approval procedures were supported by scientific and feasibility studies for certain regions within the Ruhr area. For example, an extensive feasibility study for the so-called "Central Water Province" was compiled in 2020 by RAG (see [10]) prior to the actual closure management plan, which received approval from the authorities in 2021. For this feasibility study, and many other previously conducted approval procedures, the mine water modeling software Boxmodell [46] was used to investigate and predict several aspects of the planned mine water level rise, including substances mobilization, transport and discharge. The main results of the feasibility study in this regard are summarized in Sections 3.3–3.5 below.

The feasibility study [10] of the Central Water Province also demonstrates the complexity of the mining situation in the Ruhr area. The water management plan foresees the reduction of the eight former pumping stations to one central pumping station at the Lohberg subprovince, which can control the mine water level within the entire Central

Water Province (see Figure 1 and highlighted box in Table 2). The spatial and temporal evolution of mine water for the Central Water Province is presented in Section 3.3.

3.2. Backfill Material

The deposition of backfill material in the underground mines of the Ruhr area has been intensively studied [19,37,40–42,47]. Between the mid-/late 1980s up until 2006, approximately 1.6 million tons of waste was deposited in underground coal mines of the Ruhr area [19]. The backfill material consists of combustion residues from coal, sewage and communal/household waste. Further, foundry sand and sludges from the chemical industry were used as backfill material (see Reisinger [39]).

Following a decision from the respective mining and water authorities in 1987, residues from coal combustion were classified as "emission neutral". The deposition of these residues covered many mine fields, i.e., Haus Aden/Monopol, Hugo/Consolidation, Walsum, Ewald/Schlägel & Eisen, Lippe, Fürst Leopold, Auguste Viktoria, Blumenthal/Haard and Lohberg (see Figure 2). In total, approximately 1.2 million tons of emission neutral wastes were deposited until 2006.

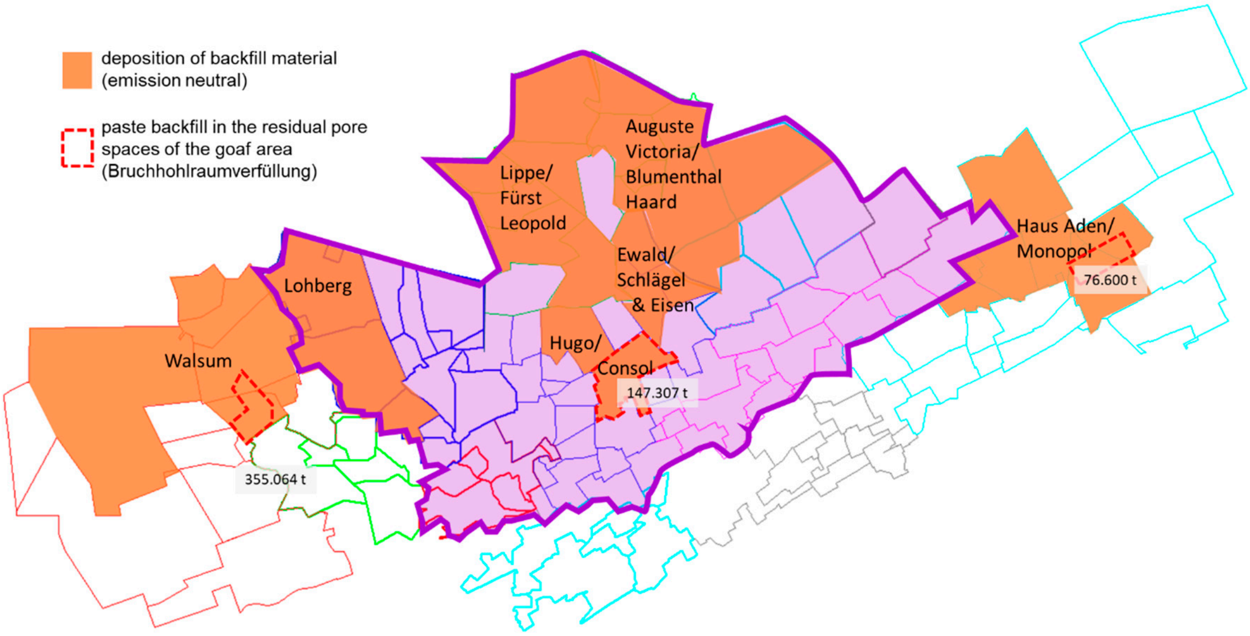

Figure 2. Overview on the backfill material in the Ruhr area based on [20].

Since the early 1980s, the Bergbau-Forschung GmbH (today DMT) tested and developed methods for depositing potentially harmful wastes (i.e., combustion residues from communal waste facilities containing PCDD/F, dioxins and furans) in underground coal mines in a series of R&D projects. The studies developed a methodology of paste backfill in the residual pore spaces of the goaf area, the so-called "Bruchhohlraumverfüllung". Following these studies, Jäger et al. [40] concluded in a feasibility study that under certain conditions, the deposition of potentially hazardous waste is environmentally acceptable. Next to the actual methodology of hydraulic paste backfill in the goaf area, these conditions are, e.g., a depth >800 m below the surface and the presence of clay-rich host rock, forming hydrogeological and hydrogeochemical barriers. The adequacy of these barriers was further investigated and substantiated by, e.g., [37]. Following the approval from the respective authorities, ~0.58 million tons of potentially harmful wastes were backfilled in the mines Haus Aden/Monopol (~0.08 million tons), Consolidation (~0.15 million tons) and Walsum (~0.36 million tons). The feasibility study [40] also contained discussions on mine water level rebound. The deposition took place between 1993 and 1998.

In 2013, a second wave of intensive investigations related to the backfill material was triggered due to a complaint, raised by local inhabitants, concerning possibly near-surface groundwater contamination through uprising mine water [19]. Consequently, the focus of these investigations was on the interaction of rising mine water and the wastes. In this context, the risk potential (i.e., mass, composition and geographic distribution of the backfill material), emission and distribution potential were determined. For the emission potential, hydrogeochemical modeling was performed, and for the distribution potential, hydrodynamic simulations were set up. The results indicated that the emission and distribution potentials were negligible for the following reasons (see [19]):

- The paste material reacts with the rising mine water, causing elevated pH values. The heavy metals, however, require low pH values for dissolution. At least 1000 years are required for the pH value to decrease to a point where Zn and Pb could possibly be leached. More realistic scenarios, where more backfill matrix material reacts with the mine water, indicate that the geochemical barrier will remain active for several tens of thousands of years.
- If heavy metals such as Zn, Cd or Pb are leached from the backfill material, sorption processes to the clay-rich formations take place, thus further decreasing the contaminant load. It is stated that within 100 m from the deposited backfill material, the potential concentrations of heavy metals Zn and Pb would decrease to a minimal percentage of the original value.
- In case mine water pumping is still maintained after the initial release of Pb or Zn, i.e., once the geochemical barrier is no longer active, the increase in Pb and Zn in the pumped mine water would increase in the per mill range and is therefore negligible.
- Without pumping, the flow paths would be ~2000 m long, which would take ~0.8 million years to reach the next receiving waters. Along this flow path, sorption processes will take place, which will reduce the concentrations to negligible values.

For dioxin, only, particle transport is possible and thus also negligible. With respect to the backfill material, it was therefore concluded that no actions were required to further reduce risk of emissions to the biosphere (Denneborg et al. [19]).

Mainly due to the persistence of the geochemical barrier, but also due to the other abovementioned reasons, the backfill material is not considered for modeling the spatial, temporal and geochemical evolution of mine water during rebound. However, due to the environmental risk potential of the mine water itself, the temporal, spatial and geochemical evolution is described in Sections 3.4 and 3.5 below.

3.3. PCB

In addition to studying the backfill material, the behavior and the mobilization of PCB in underground coal mines in the Ruhr area has been intensively researched [10,19,23,31,38,39].

PCB was used in the Ruhr area between 1964 and 1986. As noted earlier, Germany ceased all applications of PCDM in the early 1990s, so that the total amount of PCDM used in German coal mines is small in comparison with PCB. However, due to leakages, defects or the hosing down of the machines for safety reasons, PCB accumulated in underground mines. Ultimately, it can be stated that PCBs are almost always present in old mines, especially in the vicinity of shafts, due to driving, later use and carryover. Only peripheral mine workings excavated before and after the PCB period of use can be considered PCB-free. On a global scale, the extent of the contamination can hardly be quantified today. Denneborg et al. [19] state that approximately 15 thousand tons of PCB were used in the Ruhr area. Vogt [10] estimate that between 1964 and 1984, roughly 5–7 thousand tons of PCB were deposited in underground mines only in the western part of the Ruhr area. In [6], a detailed description of PCB mobilization modeling at the Lohberg site is presented.

Like other high-molecular-weight organic compounds, PCB has a very low solubility in water and a high tendency to bind to particle surfaces; therefore, it can be transported in a high content on particles. Numerous investigations have shown that this is also the case in mine water [23,24,31,48,49]. The mine water program was therefore upgraded for the particulate transport of PCBs. According to this modeling approach, PCB content in mine water is essentially a result of the suspended matter concentration in the water and the PCB content of this suspended matter [31]. In contrast to soluble species, the solid particles are eroded in turbulent water flow and accumulated again at low flow velocities.

The concentrations calculated from inflows, water flow in the mine and finally the mine water rebound are influenced by the mining conditions and the corresponding inputs of the different particle types:

- Particles containing PCB, whose mobilization and transport behavior are considered in the model.
- PCB-free particles, which are also taken into account with fractions in the model.

The relationship between the two particle types results from the area ratio of the respective mining elevation (PCB use or PCB-free) in the various hydraulically connected levels.

The model calculates a mixture of the particle contents transported by the inflows during the water rebound and mobilized by additional erosion. In general, the reduction in the particle content after flooding by diminution of turbulent flow and increased laminar flow in the water-filled galleries leads to a significant decrease in PCB concentration [24].

Figure 3 shows the PCB concentration in the Central Water Province Lohberg. Results show a wide spread in PCB contents in the mine waters in the subprovinces. This is due to the spatial heterogeneity of the remaining PCB in underground mines from the active mining period.

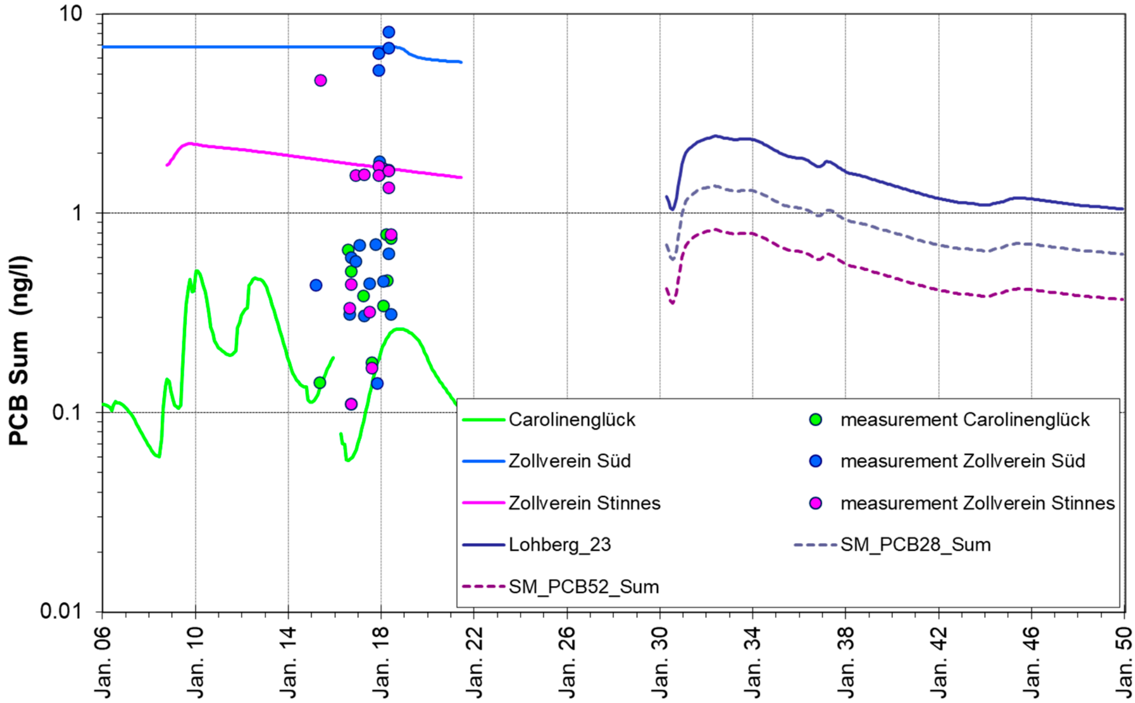

Figure 3. Simulated (lines) and monitoring data (symbols) of particle-bound PCB concentrations in the Central Water Province Lohberg.

Generally, seven PCB congeners are determined as representative indicator compounds (see [50]), which are also considered as separate substances in the mine water modeling. The chemical properties of the different PCB congeners depend on the chlorine content [10]. Due to the nature of the PCB-containing liquids used in mining, besides the sum of the seven congeners, PCB-28 and PCB-52 as main components are of particular importance. The analytically determined particle concentrations of a few mg/L and PCB levels in the solid of several 100 µg/kg result in particle-bound concentrations of a few ng/L.

3.4. Spatial and Temporal Evolution of Mine Water in the Central Water Province

In order to demonstrate the complexity of the spatial and temporal mine water evolution during rebound and how mine water management can be optimized, the feasibility study for the Central Water Province of the Ruhr area is used [6]. The Central Water Province has a surface extent of 1155 km^2. The province consists of several mines, which operated in the last decades and were successively merged into larger units. According to the water management concept, mine water level at Lohberg subprovince will be kept at −630 masl. Three pumping stations, i.e., Prosper-Haniel, Lippe and Auguste Victoria, closed in 2021. The remaining five pumping stations, which were still active in 2022/2023, are shown in Figure 4 with the subprovinces delineated in color.

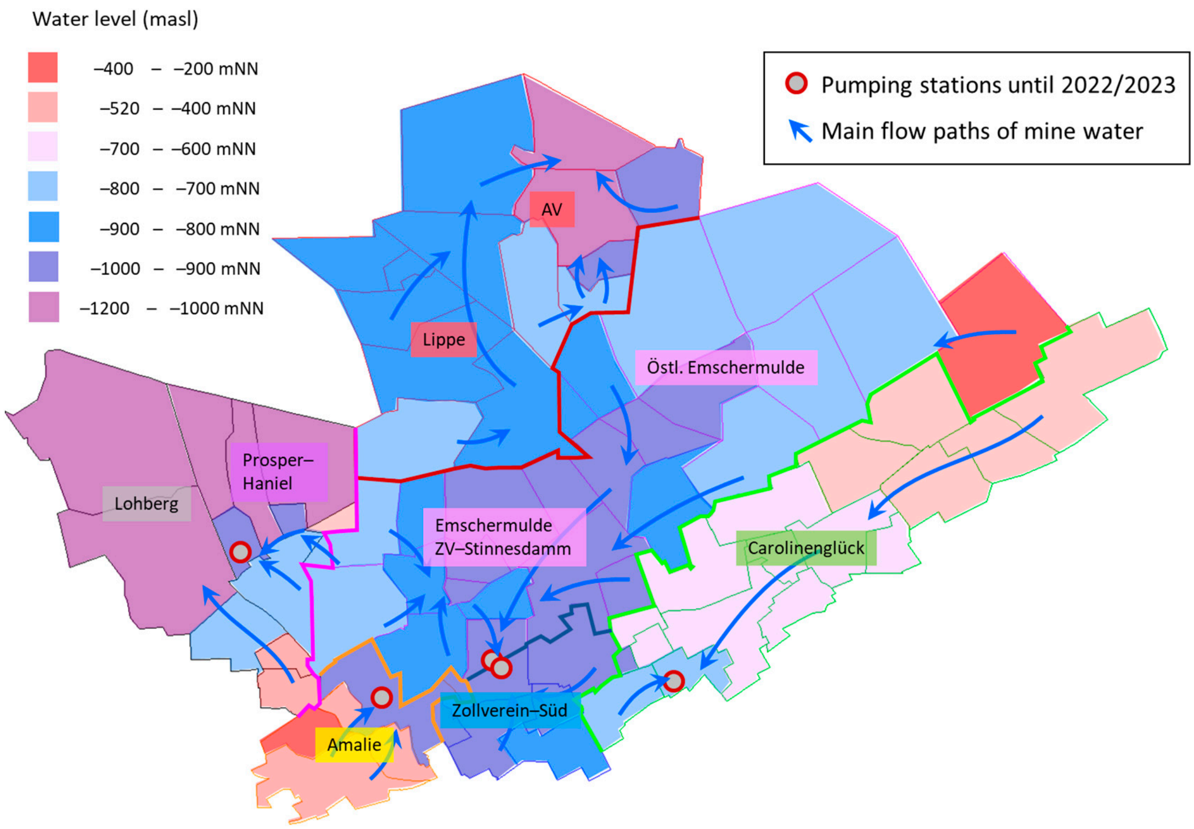

Figure 4. Simulated water levels and the water flow directions in the Central Water Province after termination of pumping in 2022/2023.

Figure 4 shows the flow paths and water levels derived with the mine water modeling software. The very complex flow paths have been simplified here into the main flow paths, which indicate the direction of flow from the individual mine areas to the pumping stations. At the beginning of 2023, the plans of the feasibility study [6] were implemented, and the pumping operations at Zollverein, Carolinenglück and Amalie were halted. As a result, the water level is now rising in these subprovinces.

In Figure 5, the predicted time-dependent water level rise is presented for the remaining five pumping stations, which were still active in 2022/2023, i.e., Zollverein III and South, Carolinenglück and Amalie. The current hydraulic situation is characterized by different water levels in the subprovinces. In most of the subprovinces, the water level currently is in rising phase with different rising speeds. This is due to the different dewatering termination times and the distinct excavation/void volumes in the mining areas. In addition, naturally occurring inflow rates are variable, which are mainly concentrated in the southern area. The monitoring data show seasonal fluctuations that are not considered in the mine water modeling software. The model forecasts the beginning of pumping in Lohberg in mid-2030, as the mine water level will then have reached the corresponding level of −630 mNHN. The water level further east (Carolinenglück, Zollverein), however, continues to increase under the model conditions, so that an additional pumping in Zollverein at −600 mNHN must be activated. The predicted water levels in Carolinenglück are always slightly above the −600 mNHN level at Zollverein. This is a result of the hydraulic gradient, required for water flow. The water level will be temporarily maintained at this stage in this region, which represents an intermediate stage towards finding an optimal mine water rebound level. This optimal mine water level serves to protect the regional drinking water reservoirs in the northern Ruhr area.

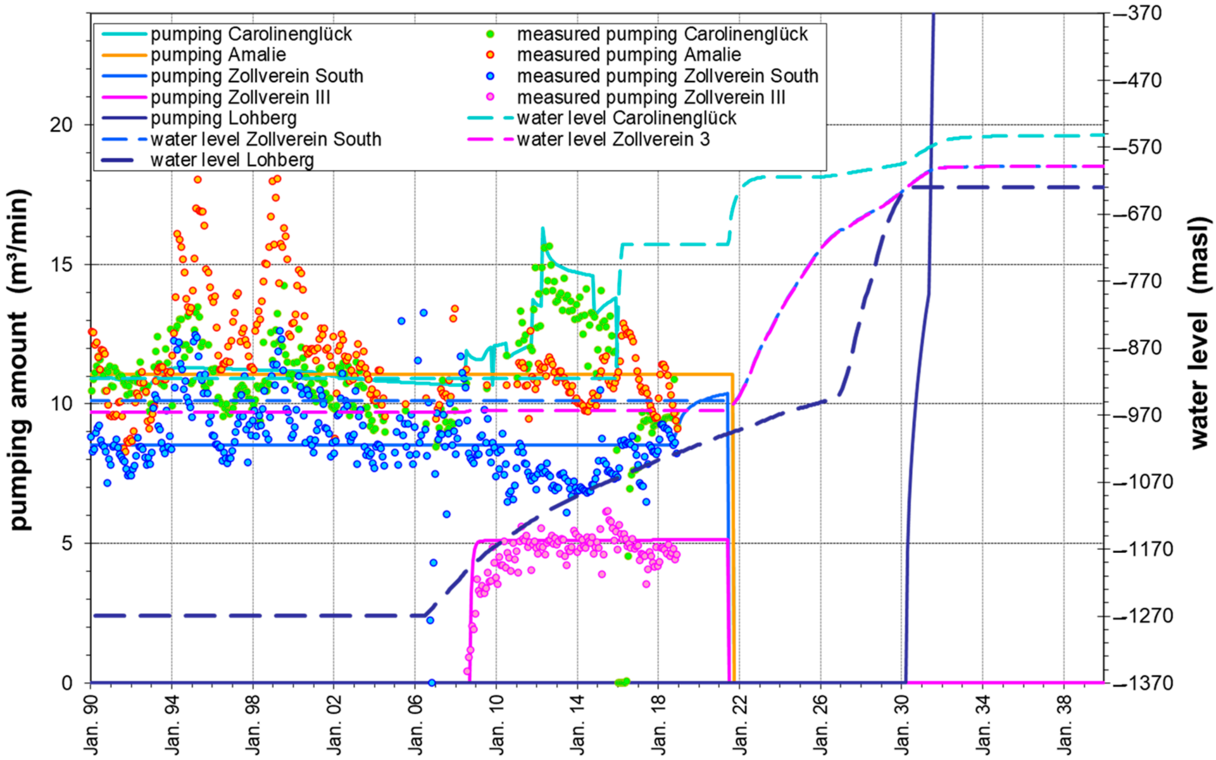

Figure 5. Simulated (lines) and monitoring data (symbols) of the pumping rate (solid) and water levels (dashed) in the Central Water Province. Note that between 2022 and 2030, mine water pumping is stopped.

Figure 6 shows the predicted water levels in the Central Water Province with the predicted water flow directions (blue arrows) after installation of the Lohberg pumping station in the expected balance state in 2050. The flow paths were again derived by modeling with the mine water modeling software. The very complex flow paths were simplified into the main flow paths, which illustrate the direction of flow from the subprovinces towards the main pumping station in Lohberg.

Following this initial prediction of the mine water rebound dynamics, mine water quality predictions can be evaluated accordingly in order to investigate if the mine water at the target elevation requires treatment prior to discharge into receiving waters.

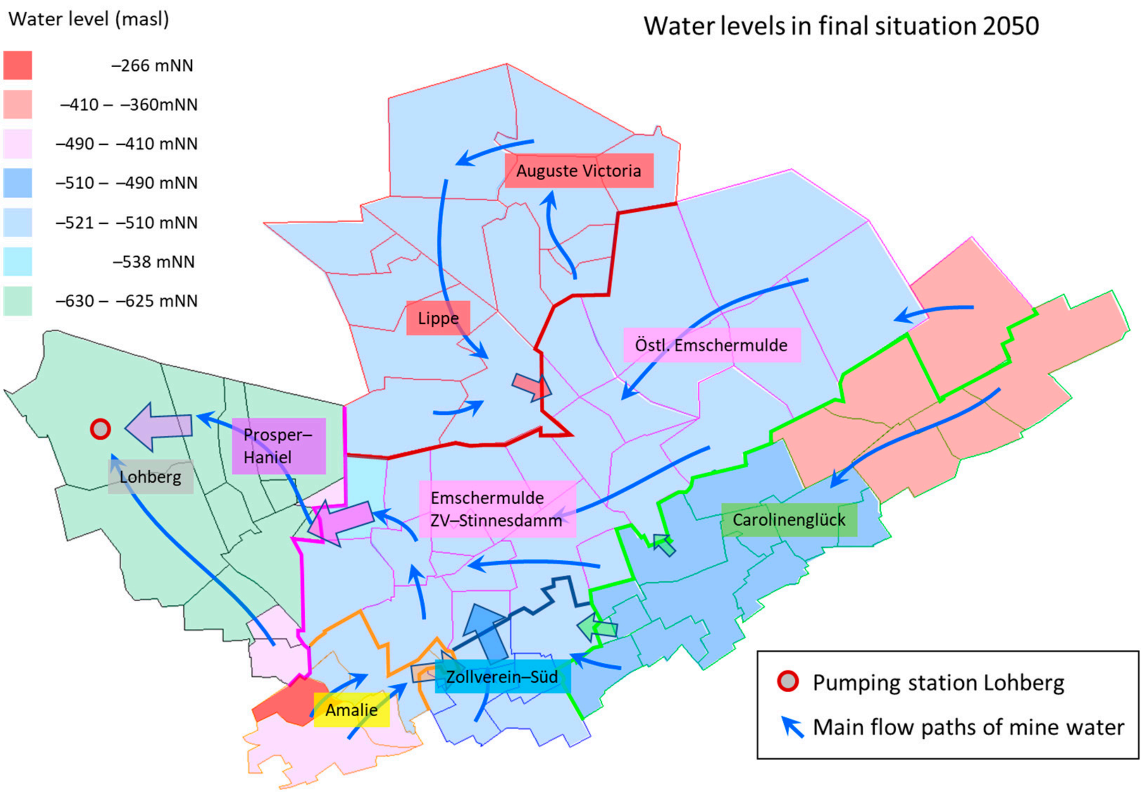

Figure 6. Predicted water levels and water flow directions in the Central Water Province in final situation in 2050.

3.5. Chemical Evolution of Mine Water

Analogous to the temporal and spatial evolution of mine water, the prediction of its chemical composition is very complex. A general understanding of the behavior and interactions of several components is, however, required in order to assess potential contaminant leaching from wastes or the possible environmental impact of mine water discharge, regardless of any interaction with waste materials. The findings made during the simulation of solute transport for the feasibility study of the Central Water Province [10] demonstrate this complexity.

The mine water composition in the underground coal mines of the Ruhr area, as for coal mining regions in general, is mainly influenced by the chemical composition of the inflow waters, pyrite oxidation, mineral precipitation (e.g., through mixing of waters with different hydrogeochemical composition) as well as microbial activities (e.g., sulfate-reducing bacteria). The interaction with waste material can potentially also contribute to mine water geochemical composition, which is, however, negligible for the case of the Ruhr area (see Section 3.2).

The development of the mine water in the Central Water Province of the Ruhr area is explained below using chloride, sulfate, iron and zinc. Their behavior also represents other groups of substances in mine water.

3.5.1. Chloride

Chloride, a highly soluble component representing the total salinity of mine water, serves as a valuable parameter for comprehending flow dynamics and water mixing processes. Chloride is less susceptible to chemical precipitation reactions, making it akin to a tracer element. In this context, it acts as a reliable indicator of water movement without undergoing significant alteration. This characteristic renders chloride concentrations particularly informative for understanding complex hydrological interactions within the mining environment.

The temporal variations in chloride concentration within the Central Water Province are depicted in Figure 7, illustrating its dynamic behavior over time.

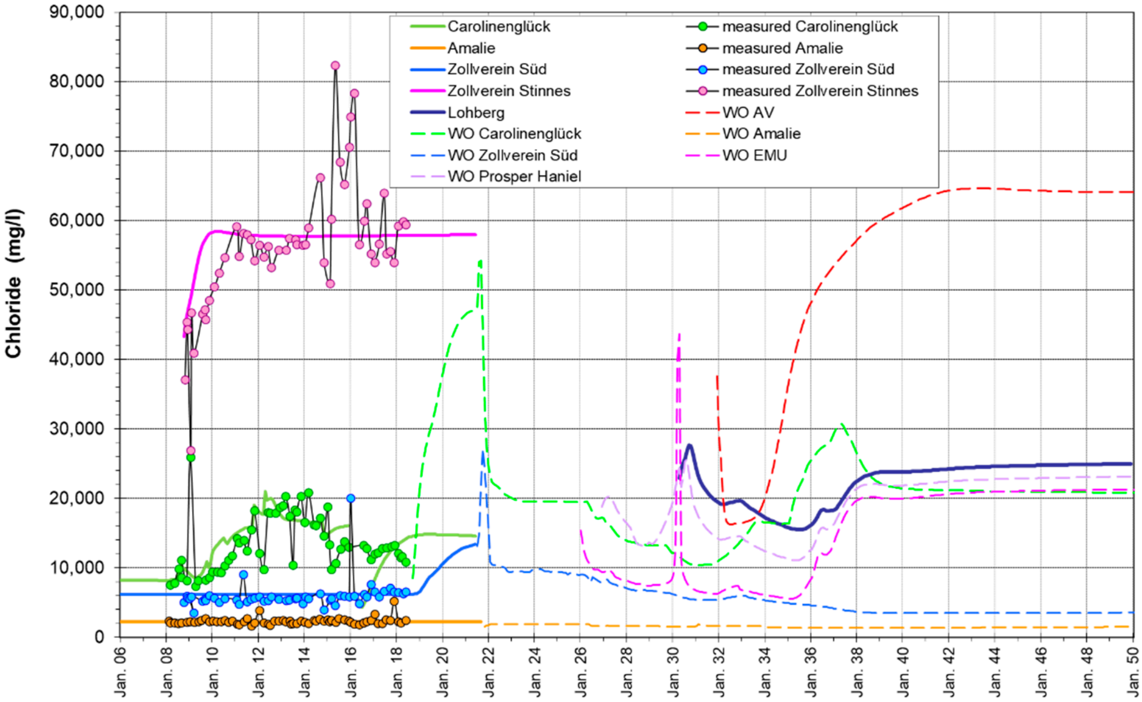

Figure 7. Simulated (lines) and monitoring data (symbols) of chloride concentration in the Central Water Province.

The investigations during the compilation of the feasibility study for the Central Water Province [10] revealed distinct salinity patterns within the study area. The central region, encompassing the deep Emschermulde (Zollverein Stinnes), exhibits a notably elevated chloride concentration, approximating 57 g/L. In contrast, the southern zone, characterized by the Water Overflow Amalie (WO Amalie), exhibits significantly lower chloride levels at around 2 g/L. The southern region demonstrates elevated flow rates, indicating the presence of significant groundwater movement.

Following the water level rise within the Central Water Province, an interplay of inflowing waters culminates in a blended mine water composition containing approximately 20 g/L of chloride. It is noteworthy that the introduction of highly saline mine water from the northern Auguste Victoria/Lippe subprovince, specifically the WO AV source, also contributes to this intricate mixture.

There is no clear evidence for a relevant mobilization of chloride during water rise, as one might assume from the dissolution of the salt deposits in drifts. Probably these amounts are too small compared to the total amounts in the mine water. Therefore, chloride mobilization from rocks and backfill materials is not considered in the model, and all the chloride discharge comes from the various inflows of the mine water. This also applies for numerous other salts such as ammonium, boron, sodium, potassium, etc., whose content is usually also closely correlated with chloride.

3.5.2. Sulfate

In order to study contaminant mobilization and transport, the development of sulfate in mine water plays a major role, since the mobilization of sulfate from pyrite oxidation in the ascending mine water has a crucial effect here. Sulfur, as the main component of pyrite (FeS_2), is especially affected by the oxidation process and the subsequent dissolution of the resulting salts and appears as sulfate in the mine water.

Figure 8, shows the concentrations of sulfate within the Central Water Province, unveiling intriguing insights into the behavior of this essential anionic component.

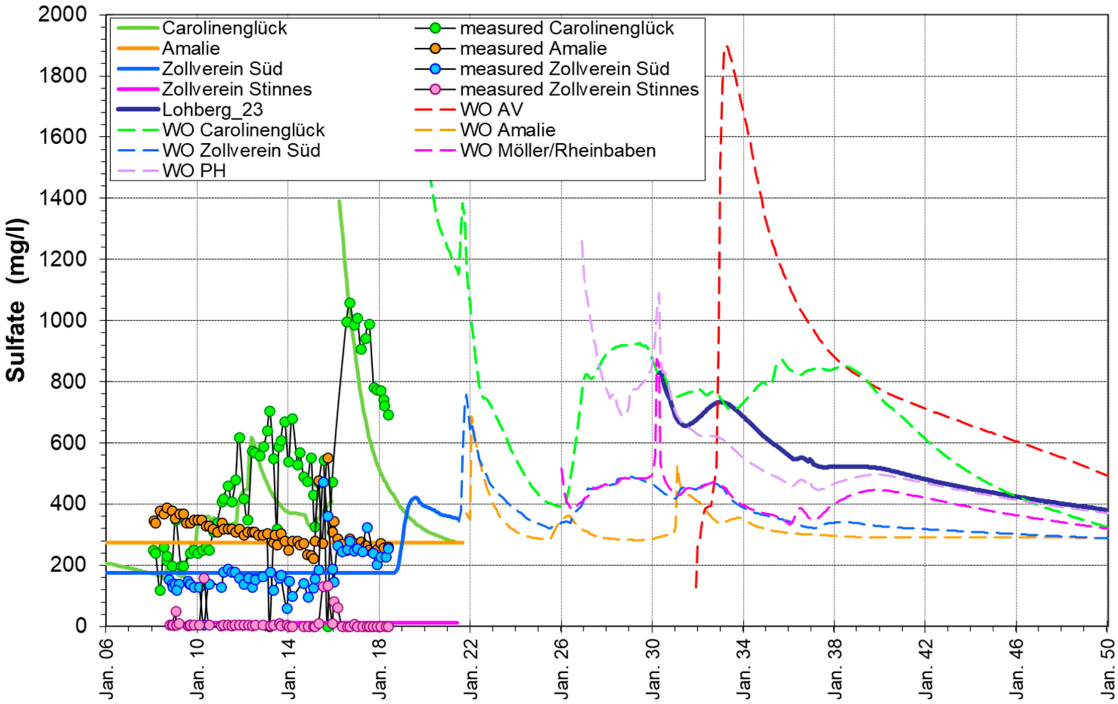

Figure 8. Simulated (lines) and monitoring data (symbols) of sulfate concentration in the Central Water Province.

Following flooding events, prominent peaks in sulfate concentrations are observed. This phenomenon can be attributed to the rapid introduction of water into the mining environment, triggering a surge in sulfate levels due to the dissolution of sulfide-bearing minerals present within the mine area. This initial increase is followed by a gradual exponential decline in sulfate concentrations. This decline is primarily due to the exchange of mine water in the post-water-rising phase.

Remarkably, Figure 8 highlights a spatial heterogeneity in sulfate distribution, wherein the southern region, particularly the WO Carolinenglück, exhibits the highest sulfate content, reaching approximately 2 g/L. This marked disparity can be attributed to the distinctive hydrogeological conditions prevailing in this area. Seepage waters from the surface, enriched with pyrite oxidation byproducts, contribute significantly to sulfate levels. The elevated occurrence of pyrite oxidation in this region results in the release of sulfate ions into the groundwater, thereby increasing sulfate concentrations. This finding underscores the substantial influence of surface interactions on the geochemical composition of mine water.

Conversely, the central Emschermulde (Zollverein) registers notably lower sulfate concentrations, measuring below 50 mg/L. The rationale behind this trend is linked to a complex interplay of factors. Notably, the deep inflows in this region carry a considerable load of barium ions. Consequently, within the confines of the mine water system, barium ions interact with sulfate ions, leading to the precipitation of barite (barium sulfate). This precipitation process effectively removes sulfate ions from the aqueous phase, thereby driving down sulfate concentrations. The prevalence of barium-associated mineral precipitation within the central Emschermulde illustrates the intricate balance between multiple chemical reactions within the mine water environment.

Barium is a constituent of saline waters, accompanied by radium in a very constant ratio. Barium sulfate precipitates quite rapidly when water containing barium mixes with sulfate-containing waters. Barium can also serve as an indicator of radioactive contamination by radium 226 and radium 228, which are also present in the precipitated barium sulfate due to correlated coprecipitation of radium. Conversely, sulfate-containing waters in the south are barium-free. The overall balance after mine water level rise is a sulfate-containing, barium-free mine water with no treatment requirements for barium/radium.

3.5.3. Iron

Iron, the second principal component released through pyrite oxidation, reveals a vital facet of the hydrochemical system in response to changing mine water levels. Comparable to sulfate, iron is mobilized during mine water rise, contributing to the evolving composition of mine water. This process exhibits analogous trends and can be prominently observed in regions such as Carolinenglück and Auguste Victoria/Lippe provinces, as illustrated in Figure 9. While the primary focus can be directed towards these regions, it is essential to recognize that iron mobilization is a phenomenon that is likely to occur universally across the mining environment.

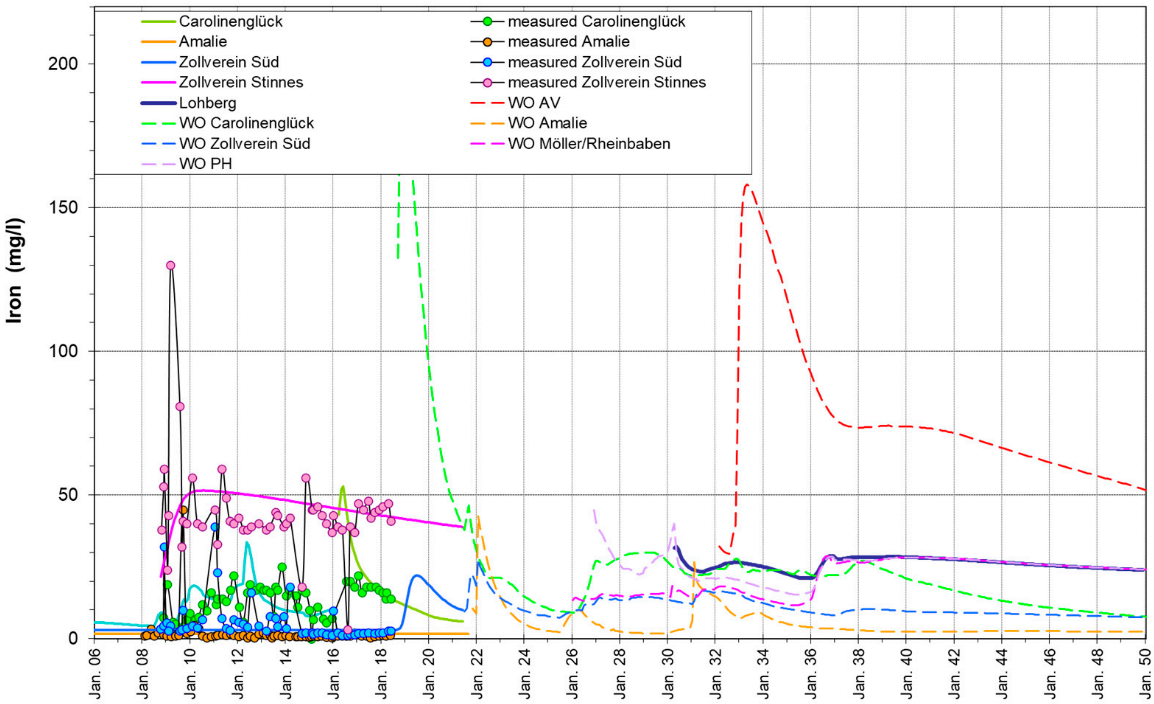

Figure 9. Simulated (lines) and monitoring data (symbols) of iron concentration in the Central Water Province.

The flushing of iron present in the mine water after the water rise in the deep Emschermulde is very slow (the inflowing water contains 13 mg/L in the mixture). A large volume of impounded water is replaced from small-inflow-rate quantities here. The Carolinenglück province reacts disproportionately more dynamically to hydraulic changes. The highest primary iron concentrations come from the Auguste Victoria mine. In addition, due to the mineralization, high material inputs are also to be expected from this area in the future.

In the overall mixture, iron levels between 20 and 40 mg/L can be expected upon discharge. The extent to which this is compatible with the limit values in the receiving river Rhine must be the subject of corresponding investigations.

3.5.4. Zinc

Metal contamination within mine water stands as one of the significant challenges faced by mining regions across the globe, garnering attention due to its widespread occurrence and adverse environmental effects [51]. Among the plethora of metals that taint mine waters, zinc takes center stage as one of the most prominently abundant.

The variances in zinc concentrations within the Central Water Province exhibit a striking dynamism, as illustrated in Figure 10. This continuous fluctuation provides an invaluable window into the intricate behavior of this metal in response to evolving hydrochemical conditions.

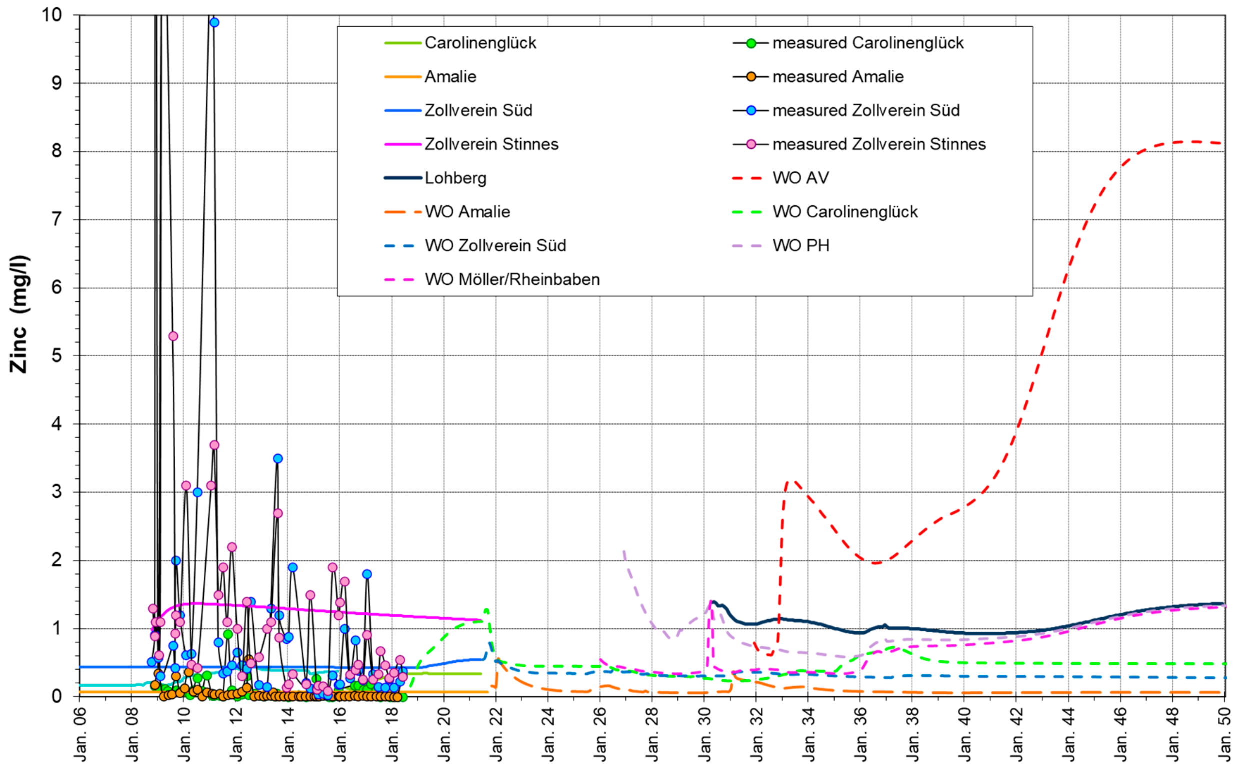

Figure 10. Simulated (lines) and monitoring data (symbols) of zinc concentration in the Central Water Province.

Drawing parallels with iron mobilization resulting from pyrite oxidation, a similar process can be inferred for zinc during mine water level rebound. The observed fluctuations in zinc concentrations are primarily driven by the interplay of geological and hydrological factors. These insights suggest that, akin to iron, zinc is mobilized as a result of pyrite oxidation or direct release from pyrite-rich minerals present within the mine workings.

Further analyses reveal that the mobilization of not only zinc but also nickel and copper is closely tied to the pyrite oxidation process. As pyrite oxidizes, the liberation of these metals occurs either directly from pyrite itself or as a secondary consequence of the oxidation reactions. This intricate interdependence underscores the multifaceted nature of the metal mobilization mechanisms within the mine water system.

Of particular interest is the Auguste Victoria mine (WO AV), as it shows notable differences in its geochemical conditions. Here, the pH value is notably lower compared to other provinces. This divergence can be primarily attributed to the influx of waters from an occurring Pb/Zn ore mineralization, which is already partially in contact with mine water. This lower pH environment creates a conducive setting for the release of zinc, culminating in an unusually elevated concentration of the metal. This localized "zinc hot spot" within the Auguste Victoria mine serves as a focal point of metal contamination, exerting a significant influence on the overall water quality within the area.

The intricate behavior of zinc in response to hydrochemical variations stands as a testament to the multifaceted nature of metal mobilization within the mine water system. The fluctuating concentrations underscore the dynamic interplay of diverse processes that govern metal release and migration. The presence of a zinc hot spot in the Auguste Victoria mine area, while detected, does not significantly impact the overall quality of the pumped mine water.

4. Conclusions and Suggestions

As coal mines reach the end of their operational life cycle, effective and responsible mine closure strategies become vital in safeguarding ecosystems, protecting water resources and mitigating pollution risks. Following the principle of continuous mine closure, these strategies should ideally be developed and reinvestigated throughout the entire mining life cycle (see [52]). This includes comprehensive planning, monitoring, predictive modeling and, based on this, management of mine water rebound to prevent or minimize contamination of surrounding surface and groundwater. Understanding the interaction of different types of waste materials in underground mines with rising mine water is of great environmental importance. Uncontrolled mine water level rebound may result in the pollution of drinking water resources (see [22]) due to mine water contact with wastes or natural substances that impair water quality. On the contrary, it has to be mentioned that, e.g., sorption processes or decreased flow velocities in flooded mining voids reduce particle transport (PCB) and therefore inherently decrease the risk of water contamination (see [19,31]).

In the Ruhr area's underground coal mines, flame-resistant material PCB/PCDM together with backfill materials of various natures are wastes which interact with mine water during the rebound phase. The showcased case study of the Ruhr area illustrates that extensive research, coupled with meticulous documentation of waste materials (i.e., backfill, potential, PCB characterization), along with a profound comprehension of the mining and hydrogeological systems, led to a comprehensive understanding of mine water risk assessment, in this case with negligible effects. This example from the Ruhr area unequivocally underscores the essential nature of documenting wastes in coal mines.

Simulations of hydrodynamics and hydrogeochemical processes occurring during the mine water rebound enable informed decisions to reduce the risk of surface and groundwater contamination and prevent environmental hazards. Next to the application of conventional modeling approaches, the mine water modeling software Boxmodell was developed and applied by DMT for the Ruhr area and significantly contributed to achieving approval for water management plan activities from the local authorities. So far, the RAG and the RAG foundation have successfully undertaken substantial efforts to ensure drinking water quality.

Monitoring and accurately characterizing the qualitative and quantitative aspects of inflows and pumped mine water play a pivotal role in model calibration and the prediction of long-term mine water drainage. This empirical foundation is indispensable in acknowledging that mine water inherently contains diverse contaminants, including potential pollutants, when discharged into surface water bodies. Consequently, considerations regarding potential water treatment strategies become imperative during the planning of controlled mine water level rebound. In conclusion, the integration of monitoring, calibrated models and the delineation of treatment strategies for mine water contaminants is a complex yet vital endeavor. Our findings reinforce the linkage between empirical insights and the emphasis of theoretical frameworks on predictive modeling for effective treatment planning. The broader implications extend to the transformation of mine water management practices within the coal mining landscape, where historical practices are being reshaped to meet modern regulatory demands while fostering environmentally responsible water treatment practices. Real-time monitoring allows water authorities to react promptly to potential issues and proactively take measures to maintain water quality and ensure water supply safety. This significantly contributes to the long-term sustainability of our water resources and plays a crucial role in safeguarding the environment and public health.

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