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Full-scale bioremediation and long-term monitoring of a phenolic wastewater disposal lake*

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Abstract: A former open-cast pit was used as the disposal site for wastewater from lignite low-temperature coking processes. The formed lake with a strong stratification had a final volume of 2×10^6 m^3. Its water was highly toxic and of dark brownish color (transparency about 3 cm). Traditional remediation methods were too expensive, and it was therefore necessary to develop a new, ecologically and economically acceptable remediation strategy. This strategy is based on a first precipitation step of the dark-brown-colored macromolecules as iron-humate flocs at pH 4.0. These macromolecules are similar to fulvic/humic acid and are formed abiotically by autoxidation of polyphenolic compounds. Thus, the dissolved organic matter (DOM, about 280 mg/l) was decreased by about 50 % and transparency improved. After a neutralization step with 2200 m^3 of 20 % calcium carbonate suspension and a subsequent "fertilization" with 0.75 % phosphoric acid (3×0.8 m^3) "enhanced natural attenuation" was initiated. Due to higher transparency, intense growth of algae ("blooming") and bacteria started in the upper zone of the lake. In winter, algae sedimented and one year after the precipitation step, the water of the surface zone was almost clear (transparency 1 m, after 10 years over 3 m) and odorless.

Keywords: bioremediation; enhanced natural attenuation; iron-humate; lignite; polyphenolic compounds; pyrolyzed lignite.

INTRODUCTION

Until the reunification of Germany in 1990, the former East German state was the biggest producer of lignite, or brown coal, worldwide (in 1978: 253 264 t a^{-1}) [1]. Lignite was not only used for the production of energy, but also formed the basis for a whole branch of chemical industry, namely, carbochemistry. This sector thrived until 1990, but caused considerable environmental damage [2]. Nearly a third of the mass of pyrolyzed lignite is water (reaction and inherent water) which is heavily loaded with volatile organic substances (mainly phenols and lower fatty acids) and ammonia [3].

Only a part of the wastewater was treated in aerobic wastewater treatment plants; the rest was passed into rivers untreated [4]. In some cases, the wastewater was pumped into layers of Upper Permian Zechstein [5] or was "stored" in former open-cast mines, forming lakes with a potential danger for groundwater [2,6].

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After 1990, a government program initiated extensive remediation work. In some cases, however, technical solutions or established methods and procedures were not applicable for economic reasons. One such case was the Vollert-Süd lake (so-called “phenol lake” in allusion to the contaminants in the water), a wastewater reservoir for process water from lignite pyrolysis in a disused open-cast mine. Process waters from a nearby low-temperature coking plant were discharged into a disused open-cast mining pit between 1950 and 1968 [7], which led to the development of a highly polluted meromictic lake [8]. Its water was of a very dark brownish color (transparency only about 3 cm), caused by the polymeric autoxidation products of polyphenolic compounds (such as catechol, resorcinol, etc. [4,9]) as constituents of effluents of low-temperature lignite carbonization [7]. These polymeric artificial fulvic/humic compounds are very recalcitrant concerning bacterial degradation [10–13]. The concentration of dissolved organic carbon (DOC) was 263–690 mg/l, depending on the water depth [7].

Because of this special water composition, traditional lake remediation methods were not feasible and it was therefore necessary to develop a new, ecologically and economically acceptable remediation strategy which considered special aspects of limnology, microbiology, and chemistry of fulvic/humic compounds.

MATERIALS AND METHODS

Site description

The Vollert-Süd lake is located in the state of Saxony-Anhalt in Germany (N51°06'15.8", E12°03'28.0", WGS84). It has surface area of 9 ha, a maximum depth of 27 m, and a volume of 2×10^6 m^3 [2,14]. The depth profile was measured by echograph (Type Krupp Atlas DESO 20 by 210 and 33 kHz) in 1990. The lake has an oval funnel shape. The maximum depth was in the center of the lake. Table 1 shows the relation of depth and theoretical surface area of the lake.

Table 1 Relation between depth and resulting theoretical surface area of the disposal lake.

Depth (m)	Lake surface area (%)
0	100
10	40
15	22
20	22
24	11
25	5

A discontinuity in the analytical data of the water body was found in the depth between 5 and 10 m (see Table 3). The upper layer down to 10 m (surface water) was calculated to contain a water volume of about 1×10^6 m^3, and the deeper layer down to 27 m also contained about 1×10^6 m^3 of water.

Sampling and analytical methods

Water samples were taken from the center of the lake at the depths of 0.5, 2, 5, 8, 15, and 27 m. Some sediment samples were taken using the “freeze-core” method [15].

Standard analytical methods

Samples were prefiltered using a polyether sulfone filter with a pore size of 0.45 µm (Filtron Technology Corp., USA). DOC and IC (inorganic carbon) were analyzed using TOC 5000 and TOC 5050 (both Shimadzu Corporation Kyoto, Japan). DO (dissolved oxygen) was analyzed using Oxi 196

(WTW, Weilheim, Germany). BOD (biochemical oxygen demand) was measured manometrically using the WTW BOD-analyzer and COD (chemical oxygen demand) using the micro method with chromosulfuric acid.

Steam-volatile–distillable phenolic compounds were measured according to [16]. Nitrate, nitrite, and sulfate were determined by ion-chromatography (DX 100 Dionex Corp., USA) using the AS4A-Sc-column with AG4A-Sc-pre-column. Sulfide was measured by the Ag 500 ion-selective silver sulfide electrode and the R 502 reference electrode (WTW). Ammonium was determined electrochemically using a NH_3-electrode (Ingold, Germany). The light absorption in the ultraviolet and visible range was measured with a spectrometer (DMS 100, Varian). The absorbance at 436 nm measured the typical brown color of the water caused by humic substances. Light absorption at 270 nm presents a typical local maximum of the absorption curves of the water. Light adsorption in this part of the spectrum is also typical of phenolic compounds and delocalized electron systems of the nucleus of humic matter [17].

Microbiological analysis

All details of the determination of the total cell counts, viable cell counts, colony forming units (CFUs), bacterial identification, and degradation potential can be found at Becker et al. 1997 [18].

Gas-chromatic determination of organic compounds and sorption/desorption studies

Details of the gas chromatography (GC) for the determination of the spectrum of the organic compounds were described in details by Pörschmann and Stottmeister [19,20].

Solid-phase microextraction for determining both conventional and headspace mode was used for determining the freely available and reversibly bound fraction of phenols in contaminated water rich in dissolved polymeric organic matter (DOM) [21,22].

Methods of in situ experiments in enclosures

Enclosure systems were used for in situ experiments. Thus, it was possible to document the effects of the external conditions of the water body (water temperature, wind, dilution due to precipitation, etc.) and, most importantly, to investigate the changes in the water during the experiments. Mixing with the water outside of the enclosure was eliminated.

The enclosure system that was used for the pilot studies consisted of a ring-shaped metal construction to which a tube-like welded foil (HD-PE, 2 mm) was attached. Foamed plastic material was tied to the metal ring and kept the enclosure system floating. It had the following dimensions: 33 m diameter; 25.5 m depth; volume about 20 000 m^3, water surface about 855 m^2, fitted with 4 working platforms (Fig. 1) [2].

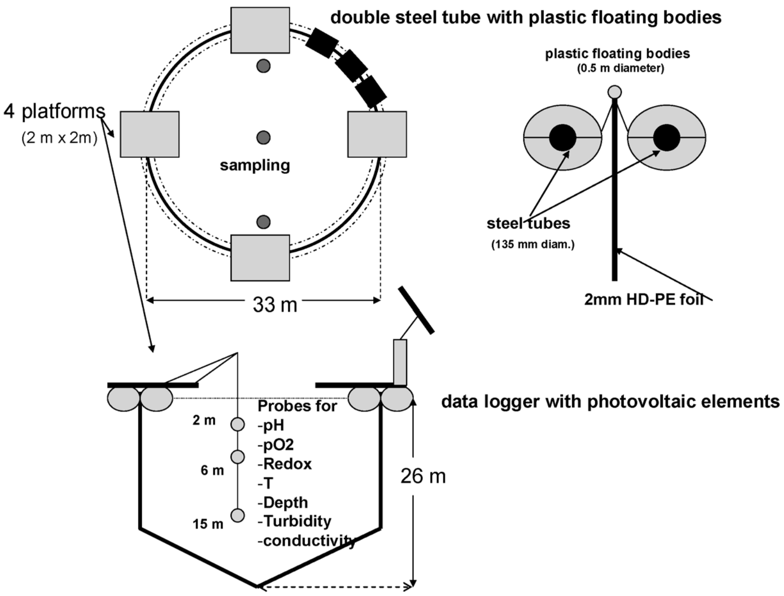

Fig. 1 Experimental design of the enclosure for flocculation and bioremediation experiments.

The enclosure system was filled with water pumped from the deposited lake from a depth of about 10 m. The analytical data of the water inside the enclosure after its filling are shown in Table 2.

Table 2 Analytical data of the water from the disposal lake in the enclosure for in situ flocculation and bioremediation experiments.

Depth (m)	pH	Conductivity mS cm^{-1}	Sulfide mg/l	NH_4–N mg/l	Phenols mg/l	COD mg/l
0	7.80	1.52	19.0	94.5	21.1	993
10	7.64	1.55	34.3	99.2	21.5	1009
20	7.66	1.59	35.4	105.7	22.3	1093

Developing the remediation strategy

Analytical characterization of the water of the deposited lake before the remediation

The analytical values of the main parameters of the water are summarized in Table 3.

Table 3 Analytical data of the water before the remediation depending on depth (samples from 2 June 1992 [7,14]).

Depth (m)	Phenols (steam-volatile) mg/l	Ammonium mg/l	COD mg/l	DOC mg/l
0	7	79	964	263
5	23	85	980	274
10	86	134	1350	465
15	202	221	2200	627
20	228	239	2290	689
25	219	250	2245	690

A detailed analysis using gas chromatography/mass spectrometry (GC/MS) coupling provided information on the individual phenolic components (Table 4).

Table 4 Analytically detectable main monomeric compounds of the water of the disposal lake at a depth of 24 m (before the remediation) [7].

Group	Compound	Concentration (mg/l)
Phenols	Phenol	135
	m-cresol	22
	p-cresol	57
	Sum C_2-phenols	53
	Sum C_3-phenols	20
	Sum C_4-phenols	7
n-alkanes	C_{10}….C_{42}	210
Alcohols	Sum hydrogenated indenols	18

Altogether, over 100 different organic compounds from different categories of contaminants were detected using thermoanalytical methods, applied in combination with highly efficient microcolumn GC/MS [23]. The concentration of different polycyclic aromatic hydrocarbons (PAHs) in water and sediment was described by Wiessner et al. [7] and Stottmeister et al. [14].

Even at the surface of the lake, the oxygen concentration was below the detection limit of about 0.1 mg/l. Experiments in oxygen consumption cells (Sapromat) confirmed that the filtrated water without microorganisms still showed oxygen consumption [7], which means that there were still not fully autoxidized polyphenolic compounds in the water with a high oxygen affinity.

The investigation of the ground of the disposal site showed a sediment layer with a thickness between 0.1 and 1.2 m (measured from freeze cores). There was no sharp transition between water and solids, so it would be more accurate to speak of a "sedimentation zone". The analysis of samples from this zone showed phenol concentrations of up to 240 mg kg^{-1}.

In comparison to the process waters as originally produced [4,9], the deposited water showed significant differences in its chemical composition:

- Low-molecular fatty acids had disappeared completely and had probably been metabolized by microorganisms despite the high concentrations of toxic phenols.
- Aromatic compounds, in particular the polyphenols, had reacted with each other and with the oxygen from the air to form macromolecules.
- Increasing aggregation of molecules had led to the formation of colloids and solid particles which were able to sediment.

A decrease of concentration of the mono-phenolic substances at the surface was detected. Ammonium was still present in high concentrations.

Microbial degradation experiments at laboratory scale

Aerobic degradation experiments [13] showed a DOC decrease of only 60 % and a decolorization between 30 and 60 % in comparison to the original water. The relation between degradation efficiency and molecular size alteration during the aerobic microbial treatment was shown by Martius et al. [17]. Eismann et. al. [24] investigated the degradation of phenols and phenolic polymers under nitrate and sulfate reducing and methanogenic conditions. They proposed the injection of nitrate into the sediment of the deposited lake. Adsorption experiments using activated carbon and activated coke [25] demonstrated the possibility of the decolorization of the water from different depths.

In summary, it was shown that the aerobic degradation of low-molecular compounds is possible. However, macromolecular fractions of the water have a toxic effect on the microorganisms. Furthermore, intensive aeration caused a formation of new macromolecules and was the reason for the measured nonbiological oxygen demand.

Aging of brown coal processing wastewater in open air disposal sites

Because the pyrolysis wastewater was stored at the disposal site over a longer period of time, significant chemical and microbiological processes had taken place. Therefore, knowledge about the wastewater treatment of fresh process water could not be applied to this already "aged" deposited water.

During the lignite low-temperature coking (pyrolysis) process, steam was generated which carried away different steam-volatile compounds. These are low-molecular phenolic and polyphenolic compounds, which were formed from the aromatic structures of the coal, low fatty acids, and sulfur and nitrogen compounds. The concentration of the compounds varies depending on the source of the coal and on the technical process that is applied. After condensation, the hot process water has a slightly red color. After cooling down, the pH is in the range of 7.5 and 10 [4].

Under contact with air, the water quickly changes to a darker color, which intensifies the longer the water is in contact with the air [26].

Reactions with atmospheric oxygen by radical formation are well known in phenol chemistry [27]. The phenolic components in the "fresh" pyrolysis wastewaters form new macromolecules upon contact with atmospheric oxygen. These can generally be compared to humins, as they behave in a similar way chemically and physically [28,29]. We proposed that these newly formed macromolecules be termed AHS—anthropogenic humic substances [11,12,23].

Applying this knowledge on the phenolic wastewater from lignite pyrolysis, the following can be concluded:

- The aromatic compounds in wastewater, particularly the numerous phenol and especially polyphenol derivates, react in the slightly alkaline wastewater under contact with air to form macromolecular structures which resemble humins.
- Thus, all findings from the chemistry of humic systems, such as sorption and formation of bond residues, can be applied to the conditions in aged lignite low-temperature coking (pyrolysis) wastewater [11,12]. Analogies between the heterogeneous system "soil contaminants" and the homogeneous deposited wastewater are possible regarding the incorporation of contaminants, masking, etc. [20–23,30].

Deduction of individual steps for the development of a holistic remediation strategy

In consultation with public authorities, it was the aim of the remediation to obtain a nonhazardous ecosystem by activating natural degradation processes. This was to be done with a minimum of technical effort and energy consumption (concept of enhanced natural attenuation).

In order to activate the complex system of the chemistry of humic compounds and the microbiological degradation processes in the surface water layer of the lake, aspects of chemistry, microbiology [28,29,31], and hydrology have been included in the development of a holistic remediation strategy, which combines individual strategies from laboratory and field studies.

Chemical remediation strategy

The dissolved AHSs were to be flocculated by adding cations as humates. Optimum humate flocculation is only achieved in slightly acidic conditions (lowering of the pH value to pH 4–5) (Step C1: acidification; see Table 5). Laboratory studies showed that iron(III)-salts had the best effects (Step C2: addition of $FeCl_3$; data not shown). An additional flocculation effect was to be expected due to destruction of colloids. From knowledge of the general behavior of humic matter systems, it could furthermore be expected that "bond residue" molecules would be released from their macromolecular matrix. These remain dissolved in the water phase after the settling of the insoluble flocs (Step C3: settlement of the flocs).

Table 5 Steps of the chemical remediation strategy of the contaminated water of the disposal lake.

Step	Measure	Expected results
C1	Acidification pH 4.0	Colloid destruction, formation of insoluble humates
C2	Addition of $FeCl_3$	Flocculation of insoluble Fe-humates
C3	Settlement of flocs	Low-molecular organic compounds are soluble in the water phase

Microbiological remediation strategy

The deposited wastewater contained up to 10^5 bacterial cells per ml depending on the water depth [18]. However, it appeared that these were inhibited in their ability to degrade the water contaminants. It was assumed that the reasons were:

- a lack of oxygen
- non-bioavailability of some organic compounds (masking by the AHSs)
- toxicity of some oligomeric chinoid autoxidation products of polyphenolic
- components (particularly for the oxidation of ammonium)

From the viewpoint of microbiology, the chemical remediation strategy steps C1–C3 offer the following advantages:

- presence of oxygen (no new oxygen-depleting formation of humates)
- decrease of toxicity
- increase of bioavailability

Arguments against these strategies were:

- Acidic pH values do not provide optimum conditions for bacterial degradation.
- The amount of dissolved phosphates decreases when iron salts are added.

Regarding the application of the chemical remediation strategies, the following measures were recommended:

- neutralization to pH values between 7 and 8 for activation of the bacterial community (M1)
- in case of signs of phosphate limitations for the growth of bacteria, addition of phosphates (M2)

Hydrological considerations

The Vollert-Süd with the deposited wastewater could not be treated as a “normal” lake with seasonal changes of the water density, stratification, and the mixing due to wind and waves [8].

Analytical investigations showed stratification of contaminant concentration and of electrical conductivity (data not shown), particularly between depths of 5 and 10 m (Table 3). The prevalent concentration profile of contaminants with higher concentrations in the deeper zone had developed over the years. Calculations that took into account the shape of the disposal site and the water depth showed that this concentration stratification was practically identical with the natural boundary layer that would be expected in an equivalent natural lake. According to these calculations, water layers below 8 m should not be affected by seasonal mixing (temperature, wind). Therefore, it was necessary to keep the natural concentration stratification intact and to avoid a mixing of the water (aspect H1). At the boundary layer, a zone for the exchange of deep water and surface water will develop mainly only due to diffusion. This behavior of the deposited wastewater of the Vollert-Süd led to the assumption that iron humate flocs that have sedimented in the deep will remain there (aspect H2).

Overall remediation strategy for the disposal lake

The individual strategies of the different disciplines and their resulting requirements were combined into one overall concept.

Step 1
- Addition of acidic iron(III)-chloride solution. The deposited water must reach a pH value of 4–5 (C1, C2).
- The natural stratification of the water must be kept intact when adding the flocculation agent into the water (H1).
- The flocculated humates should remain at the bottom of the deposited water (C3, H2).

Step 2
- Neutralization of the acidified deposited water (pH 7–8) (M1).

Step 3
- Addition of a calculated amount of phosphate for an optimum growth of microorganisms and algae (M2).

Methods for the full-scale treatment of the disposal lake

A new system for the injection of acidic iron(III)-chloride solution and lime suspension had to be developed (SIDRA Wasserchemie, Bitterfeld, Saxony-Anhalt, <http://www.sidra.de/>). An engine-driven raft was connected to the banks of the lake with a floating pipeline and navigated the water according to a grid system that was marked by buoys.

The solutions were injected layer-by-layer—first into the lower layers and then in the same way into the upper layers—while the raft moved forward. Thus, the stratification remained but horizontal mixing of the flocculation agent and the lake water was achieved (see below). The 40 % $FeCl_3$ solution was supplied continuously with the help of a tank. The lime suspension was stored in stirred containers at the banks of the lake (see below for volumes). Phosphate was added as H_3PO_4 (75 %), diluted 1:200 (CWP Chemische Werke Piesteritz GmbH).

EXPERIMENTS AND RESULTS

Experiments at pilot scale (in situ enclosure experiments)

The behavior of the deposited wastewater and the microbial transformations in the enclosure could be determined easily and quickly using the four main parameters DOC, transparency, steam volatile phenols, and water temperature. For the realization of steps 1–3 of the overall remediation strategy, 45.7 t of 40 % $FeCl_3$ solution were used for the flocculation, and 41 t of 20 % lime suspension were used for the neutralization. The progress of the experiment over the first two years is illustrated in Fig. 2.

Day zero signifies the start of the dosage of iron(III)-chloride solution with simultaneous acidification to a pH value of 5. The immediate reaction is indicated by a decrease of DOC by about 50 %, caused by the settling of humate flocs, and an increase of transparency from 3 cm to about 80 cm.

During the acidic phase of about three months (winter), mixing and secondary flocculation occurred without external influences and these could be identified by a decrease in the analytical values (up to day 80).

After this period, the content of the enclosure was neutralized with lime suspension and adjusted to a pH value of about 7.5. This neutralization meant an initial increase of all parameters, especially as a mixing due to diffusion was made easier in spring when the density of the surface water and that of the deeper layers within the enclosure were equal. The substantial increase of the transparency of the water to 2 m was remarkable. In this upper layer (down to a depth of 2 m), concentrations of DO of about 1 mg/l were measured.

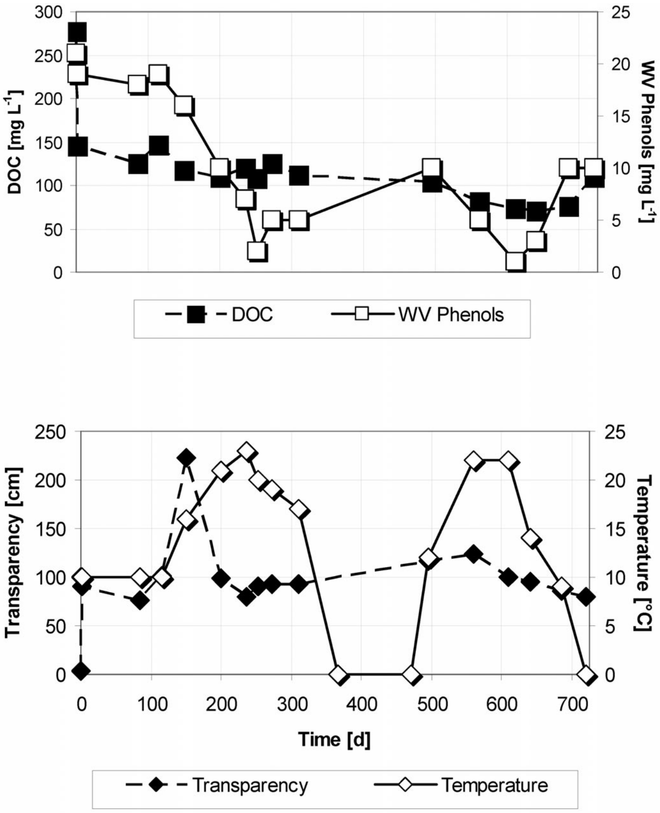

Fig. 2 In situ pilot study for the realization of the remediation strategy in the enclosure (Flocculation: at day zero, 14–15 December 1994, 36 t $FeCl_3$ solution (40 %); 22 February 1995: 9.7 t $FeCl_3$ solution (40 %); resulting in a pH of 4.5. Neutralization: at days 80/81, 2–3 March, 41 t $Ca(OH)_2$, 20 %, resulting in a pH of about 7.5).

After three more months (approx. days 100–200), the concentration of steam-volatile phenols decreased significantly, at the same time as the water temperature increased. In late summer (days 240–260), a concentration of just 2 mg/l was measured in the surface water (initial value: 21.5 mg/l). The transparency returned to a value of about 80 cm.

Seasonally dependent mixing was again noticeable in autumn and winter of the following year (days 300–500). In summer (up to day 620), the concentrations of the steam-volatile phenols decreased again. DOC, a measure for the sum of all organic dissolved compounds, also decreased slowly and steadily.

Microbiological investigations that were carried out at the same time as the chemical analysis confirmed the microbiologists' hypothesis. According to this hypothesis, only a low degree of microbial degradation was to be expected after the flocculation, the acidic phase, and neutralization. However, bacterial recolonization was to be expected with the favorable pH conditions for bacteria present immediately after the neutralization. The microbiological results are published in detail by Becker et al. and Stottmeister [18,32–36].

These new microorganisms found the following optimum conditions:

- low-molecular aromatic compounds as a utilizable carbon source, as well as sufficient nitrogen present as ammonium and
- high concentration of DO.

Because of the growth of bacteria connected with the degradation of contaminants, the low-molecular components of DOC decreased steadily over the years. In the following two years, phosphate limitation became evident in the form of a decrease in the growth of bacteria. This was resolved by adding phosphoric acid (7.2 l 75 % H_3PO_4, diluted 1:200, injected at a depth of 0.5–1.0 m). The average concentration of phosphate was about 1.4 mg/l.

The full-scale treatment—Induction of enhanced natural attenuation

Based on this successful pilot study in the enclosure within the lake, the conditions for the full-scale remediation were deduced.

The whole lake was treated exactly like the enclosure with a diameter of 33 m by:

- flocculation with iron(III)-chloride solution,
- adherence to necessary time intervals of about 80 days,
- neutralization with lime suspension, and
- addition of phosphate

The calculated concentration was 4.2 mol $FeCl_3$ m^{-3} for the upper (surface) zone and 7 mol $FeCl_3$ m^{-3} for the deep water zone. On this basis, the total amount for the application in the upper zone was calculated to be 1200 m^3 and for the deeper zone 2000 m^3 $FeCl_3$ (40 %).

The injection of the acidic $FeCl_3$ solution started on 1 October 1996 and finished 6 December 1996.

The neutralization with 20 % lime suspension (2200 m^3) started in the following year (23 April 1997) and finished on 29 August 1997. The input of the stirred suspension was carried out from the surface into deeper layers and was a technical challenge.

The time of the phosphate addition was determined in response to the biological development and the addition was carried out in three steps (28 April 1998, 19 August 1998, 6 May 1999; 3 × 0.8 m^3 of 75 % phosphoric acid, diluted 1:200).

The water behaved as in the pilot study with regard to the recolonization and activation of the microbial community and the subsequent degradation of steam-volatile phenols. As a visible success of the remediation process, the water lost its color and had a transparency between 60–200 cm, depending on the season.

The results of the water analyses after 5 years and 10 years are shown in Table 6.

Table 6 Summary of the main water parameters of the disposal lake in 2004 (July) and 2008 (August, 10 years after initiation of the enhanced natural attenuation).

Depth m	Phenols (steam-volatile) mg/l		NH_4–N mg/l		COD mg/l		DOC mg/l	
	2004	2008	2004	2008	2004	2008	2004	2008
0	<0.01	<0.01	43	33.9	250	80.9	29	27.1
5–6	0.8	<0.01	54	36.7	275	80.9	38	28.0
10	62	<0.01	70	37.8	420	82.9	83	29.8
15–16	8	0.45	79	72.9	456	n.d.	106	96.9
20	11	0.64	86	82.1	728	n.d.	152	136
24	22	0.78	90	91.8	1204	n.d.	302	143

DISCUSSION AND FINAL REMARKS

As far as we know, this is the first successful and comprehensively documented full-scale remediation of a wastewater disposal site of the chemical industry of such dimensions according to the principle of enhanced natural attenuation. The successful realization was made possible by combining the expertise of chemists, microbiologists, and hydrologists.

Ten years after the initiation of the natural degradation of the phenolic and aromatic components, the concentrations of these components even in the deeper zones of the lake have been significantly reduced or—as in the case of steam volatile phenols—completely removed. Ammonium removal, facilitated by a neutral pH and probably due to ammonium oxidation, which is very sensitive to toxic influences and denitrification, also proceeds slowly.

The stratification (mixolimnion and monolimnion) of this meromictic lake was not disturbed by the remediation processes [8]. An aerobic ecosystem at the surface and an anaerobic ecosystem in the deep zone of the lake coexist stably side-by-side. The biology of the surface water had developed almost identically to that of unpolluted surface water close-by (comparable bacterioplankton) [36]. Water birds have accepted the new lake, and are breeding. However, no fish have their habitat there because of the still relatively high ammonium content. Before flocculation, there was a strong smell noticeable in the surrounding villages, but this was no longer the case directly after flocculation.

Degradation processes also take place in the anaerobic deep zone, and become evident in occasional localized gas eruptions (methane and carbon dioxide formation) at the water surface.

Over the course of the study, further unexpected positive effects were observed which had not been part of the original remediation strategy. After elimination of water toxicity by flocculation and subsequent phosphate dosage, large amounts of algae started to grow. Photosynthesis in these algae produced additional oxygen, which led to high DO concentrations (up to 12 mg/l). This has a positive influence on aerobic microbial degradation even in deeper water zones. Dying algae sink into deeper water zones and provide easily utilizable carbon for anaerobic processes.

In addition to the successful remediation, the whole project also provided new findings for fundamental research. Particularly worthy of mention are publications on the analysis of waters containing humic matter [7,19–21,30,37–39], behavior of AHS, the theory of sorption to humic matter [10–12,22,23,40–46], and studies in microbiology [13,17,18,24,32–36,47] and hydrogeology [5,6,8]. Phytoremediation studies at a pilot-scale constructed wetland on site led to fundamental findings regarding the annual cycle in nitrogen removal [48].

We did not investigate the processes of "humic matter respiration" [49] which are very likely to have occurred.

The existing experimental data will also provide a basis for new findings in future. The lake will continue to be observed. New analytical documentation is envisaged at intervals.

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