Testing Iron Removal in a Trifurcated Pilot Plant for Passive Treatment of Circum-neutral Ferruginous Mine Water

Joscha Opitz[1,2], Matthias Alte[2], Martin Bauer[2], Stefan Peiffer[1]

[1]*University of Bayreuth, Department of Hydrology, Universitätsstraße 30, 95447 Bayreuth, Germany, joscha.opitz@uni-bayreuth.de*

[2]*BASE TECHNOLOGIES GmbH, Josef-Felder-Straße 53, 81241 Munich, Germany*

Abstract

A trifurcated pilot plant was implemented at an abandoned mine site for passive treatment of circum-neutral, ferruginous seepage water. Purpose of the composite system was to investigate the suitability of serially connected settling ponds, wetlands and sediment filters for passive iron removal in accordance with the strict compliance limit of 1 mg/L. After demonstrating reproducibility at equal flow rates in the three identical lines, hydraulic variation was used to develop comparison data for sizing of a full scale passive system. The multi-stage setup proved successful, reducing relatively low iron concentrations of app. 10 mg/L in the influent to compliance level in the effluent. System monitoring provided evidence that physico-chemical removal of suspended ferric hydroxides is a critical step in natural iron removal and may even become the rate-determining step for thoroughly oxygenated, circum-neutral mine waters. Area-adjusted removal rates were relatively low because of low hydraulic loading. Dependence of both treatment efficiency and area-adjusted removal on hydraulic loading was demonstrated during the variation period, showing direct and inverse relationships at graded flow rates, respectively. Ultimately, a more extensive and higher resolution database is necessary for pilot plant upscaling to full scale.

Keywords: passive treatment, iron removal, wetlands, hydraulic variation

Introduction

Contamination of water resources through mining activities is one of the most complex and costly environmental problems worldwide. Most prominent is the occurrence of acid mine drainage (AMD) as a result of sulphide mineral oxidation, which causes ferrous iron, acidity, sulphate and associated metals to be released into aqueous environments. Subsequent aeration and neutralisation lead to precipitation of ferric hydroxides, resulting in compromised usability of water resources and/or complete destruction of aquatic ecosystems. Ferruginous mine water can be treated in passive systems by utilising and enhancing natural (geo)chemical, biological and physical processes (Skousen et al. 2017). However, the scale of passive components (settling ponds, aerobic wetlands) is currently based on an empirical, area-adjusted removal rate by Hedin et al. (1994) that doesn't reflect the kinetic background of respective biogeochemical and physical iron removal mechanisms (Sapsford and Watson 2011; Tarutis Jr. et al. 1999).

A trifurcated pilot plant was installed for passive treatment of ferruginous seepage water from a former open-pit lignite mine. Whereas most multi-line (pilot) systems were constructed to test different materials or setups (e.g. García et al. 2004; Nyquist and Greger 2009; Whitehead and Prior 2005), the innovative system in this study consists of three identical lines with multiple successive treatment stages each for natural iron removal. The system was designed to collect comparable hydraulic and hydrochemical data to investigate performance, kinetic relationships and critical influencing factors as a basis for upscaling. After providing evidence of reproducibility at equal flow rates in the three lines, hydraulic variation was used to investigate iron removal efficiency as a function of flow rate. Furthermore, monitoring data is used to investigate kinetics and connexions of natural iron removal mechanisms with a focus on physico-chemical ferric iron removal processes such as aggregation and sedimentation/filtration.

Pilot Plant

The study site is located in the historic lignite district of Upper Palatinate (Germany) (fig. 1). For protection of surrounding aquifers, seepage water from a former open pit is pumped to a conventional treatment plant for iron removal via flocculation and coagulation. A small amount of the raw water is split off via a bypass and diverted to a distribution tank preceding the pilot plant.

The pilot plant for passive treatment of the mining influenced seepage water consists

Figure 1 Study site

of three identical, parallel lines with multiple successive treatment stages each (fig. 2):

1. Preliminary treatment in settling ponds,
2. fine treatment in aerobic wetlands and
3. purification in sediment filters.

The composite setup was chosen to meet the very low compliance limit for total iron of 1 mg/L by successive iron removal with both treatment efficiency and maintenance requirements of the passive treatment components increasing along the flow path.

Roll-off containers (7.00 × 2.35 × 1.25 m) were used for settling ponds and wetlands. Sediment filters are liner-sealed, approximately trapezoidal trenches (ca. 4.0 × 0.5 × 0.5 m) filled with granite gravel (8-16 mm). A total of ten sampling/measuring points numbered MP01-MP10 were installed in protected manholes with MP01 as inflow of the three parallel system lines, MP02-MP04 behind settling ponds, MP05-MP07 behind wetlands and MP08-MP10 behind sediment filters (fig. 2). The outflow of one component thus corresponded to the inflow of the subsequent component, with discharge from sediment filters (MP08-MP10) corresponding to system line outflows. Flow rates were adjusted by way of ball valves equipped with flowmeters in the three feeding pipes branching off MP01.

Materials and Methods

Monitoring of the pilot plant includes fixed sensors for continuous measuring of pH, conductivity and dissolved oxygen at the central line and all wetlands (MP01, MP03, MP05-MP07, MP09) and turbidity at all measuring points. Automatic measurements are double-checked weekly with hand-held meters (pH, SC, O_2, turbidity, redox potential). Water samples are collected once

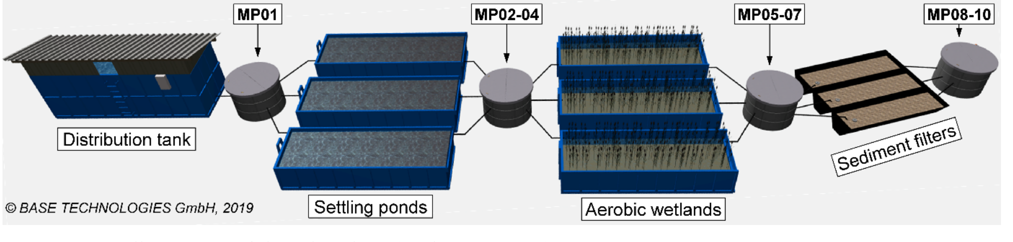

Figure 2 3D-illustration of the pilot plant (without terrain) or twice a week at all measuring points. Laboratory analysis includes dissolved (<0.45 µm) ferrous and ferric iron, particulate (>0.45 µm) and total iron (Butler et al. 2008) as well as mayor elements. Operation of the pilot plant started in November 2017 and the first months were used as a test and adjustment period. In a first step, system inflow was distributed homogeneously across the three system lines to confirm similar performance at equal flow rates. A reference dataset was obtained between July and November 2018 (113 days) with evenly distributed flow rates ("reference period"). In a second step, comparison data was obtained between November 2018 and March 2019 (113 days) with flow rates incrementally varied (1st "variation period"). It should be noted that hydraulic fluctuations were inevitable in at least one system line (usually line 3) subject to filling level variations in the gravitationally drained distribution tank. Resulting average flow rates were 269±38 L/h, 277±8 L/h and 262±59 L/h during the reference period and 178±12 L/h, 276±16 L/h and 344±76 L/h during the variation period in system lines 1, 2 and 3, respectively.

Performance of individual treatment components was evaluated using typical performance indicators such as treatment efficiency (%-removal) and area-adjusted removal (areal mass removal per time interval) (Hedin et al. 1994, Tarutis et al. 1999). Flow rate and hydraulic (iron) loading in the pilot plant were directly proportional because of equal inflow iron concentrations in all system lines and may thus be used synonymously in the context of performance evaluation.

Results and Evaluation

Results of the entire operation period (including the test period) showed excellent system performance with effluent iron concentrations consistently below the compliance level of 1 mg/L (fig. 3). High-resolution monitoring of hydrochemical parameters revealed environmental factors (esp. seasonal and diurnal cycles) had a strong effect on ecology and hydrochemistry of the aerobic wetlands. For instance, plant growth and algae blooms in spring and summer resulted in substantial diurnal cycles of pH and dissolved oxygen in the wetlands. Besides, turbidity and conductivity were affected by discontinuous pump operation and concomitant filling cycles of the distribution tank, decreasing substantially during pump intermissions (e.g. weekends).

Inflowing iron was predominantly composed of suspended (particulate) ferric hydroxides (>90 %). As a consequence, iron removal occurred predominantly by way of physico-chemical processes such as aggregation and sedimentation/filtration. Iron analysis during the reference period (n=30) showed an overall iron removal efficiency of 97.8 % with most of the iron retained in settling ponds (72.8 %), followed by wetlands (19.6 %) and sediment filters (5.3 %) (tab. 1). Although the bulk of iron is retained in settling ponds, treatment efficiency is similarly high in subsequent treatment components despite substantially

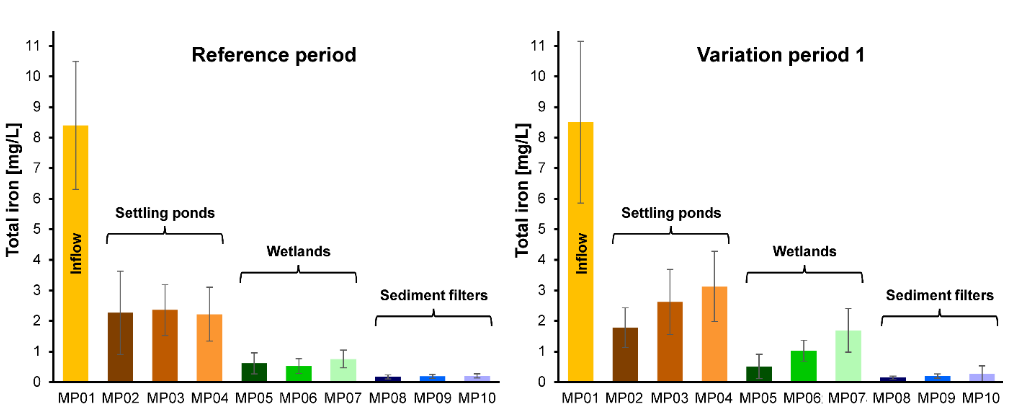

Figure 3 Iron monitoring in the pilot plant

Table 1 Key figures for treatment components during the reference period

Key figures	MP01	MP02	MP03	MP04	MP05	MP06	MP07	MP08	MP09	MP10
Outflow c(Fe) [mg/L]	8.4±2.1	2.3±1.4	2.4±0.8	2.2±0.9	0.6±0.3	0.5±0.2	0.8±0.3	0.2±0.1	0.2±0.1	0.2±0.1
Treatment efficiency [%]	-	73.0	71.9	73.6	72.9	77.8	65.9	72.4	63.6	72.7
Area-adjusted removal [$g/m^2/d$]	-	2.4	2.4	2.4	0.6	0.7	0.6	1.0	0.7	1.2

Table 2 Key figures for treatment components during variation period 1

Key figures	MP01	MP02	MP03	MP04	MP05	MP06	MP07	MP08	MP09	MP10
Outflow c(Fe) [mg/L]	8.5±2.6	1.8±0.7	2.6±1.1	3.1±1.1	0.5±0.4	1.0±0.3	1.7±0.7	0.1±0.1	0.2±0.1	0.3±0.3
Treatment efficiency [%]	-	79.0	69.1	63.1	71.2	60.9	45.9	71.1	80.4	83.4
Area-adjusted removal [$g/m^2/d$]	-	1.7	2.4	2.7	0.3	0.6	0.7	0.5	1.8	3.9

decreased inflow concentrations (tab. 1), confirming expected superior treatment efficiency of wetlands and sediment filters.

Iron analysis during variation period 1 (n=30) also showed decreasing concentrations, yet with clear gradation according to flow rates (fig. 3, tab. 2). As expected, this corresponded to an inverse relationship between treatment efficiency and hydraulic loading in settling ponds and wetlands. The effect was, however, mostly offset by sediment filter efficiency, resulting in only marginally graded overall iron removal in system effluents during variation period 1 (fig. 4).

Whereas cumulative iron removal along the flow path of the three system lines is almost identical at similar flow rates during the reference period, distinct gradation according to flow rate is in evidence during variation period 1 as illustrated by fig. 4.

Area-adjusted removal based on estimated total mass removal over the reference period was similar in the three system lines at similar flow rates. Dependence of not only iron removal efficiency, but also area-adjusted removal on hydraulic loading was demonstrated during variation period 1. Line 2 showed similar results compared to the reference period at a similar flow rate, whereas lines 1 and 3 showed substantially decreased and increased area-adjusted removal at decreased and increased flow rates, respectively. Dependence of area-adjusted removal on hydraulic loading is illustrated for settling ponds in greater detail in fig. 5. Boxplots are based on all iron analysis with available flow measurements for both

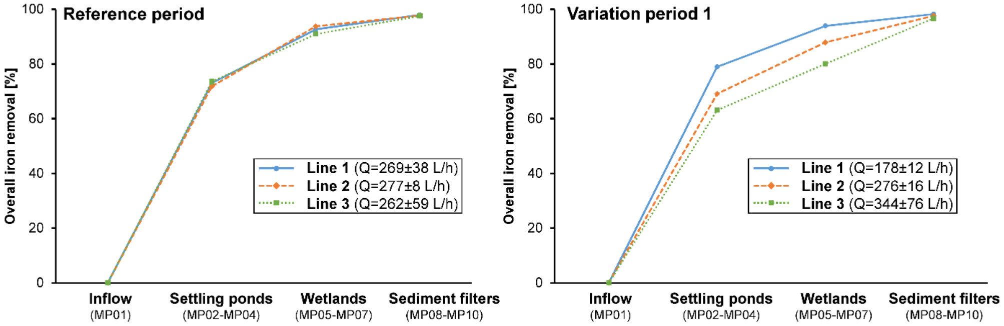

Figure 4 Iron removal along the flow path with flow rates (Q)

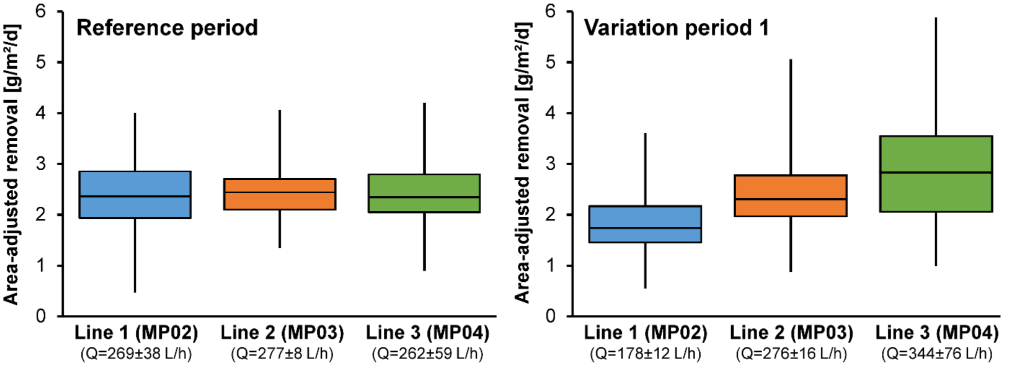

Figure 5 *Area-adjusted removal in settling ponds with flow rates (Q)*

reference and variation period with 26 and 29 data pairs, respectively. Flow rates were averaged over an interval of ±24h around the sampling time to compensate for hydraulic fluctuations as best as possible.

Discussion and Outlook

It should be noted that the datasets described above were subject to varied environmental influences with reference and variation period set in autumn and winter, respectively. Differences in temperature and vegetation development as well as variation of hydraulic retention time subject to pump operation or ice thickness were inevitable, yet potential effects on iron removal processes are difficult to quantify wherefore higher resolution data is necessary.

The composite system showed excellent treatment efficiency with iron removal consistently >95 %. Settling ponds showed considerably higher area-adjusted removal rates than wetlands due to much higher hydraulic loading on the one hand and area-adjusted removal not correcting for lower hydraulic retention time due to lower volume at same surface area on the other hand. Ranges of area-adjusted removal for settling ponds were in accordance with >50 % of passive systems investigated by Tarutis et al. (1999). Nevertheless, area-adjusted removal for both monitoring periods in the pilot plant was still orders of magnitude lower compared to (optimally) expected ranges for wetland sizing of 10-20 $g/m^2/d$ (Hedin et al. 1994). However, it is well-recognised that area-adjusted removal does not adequately reflect treatment performance with regard to non-linear overall iron removal kinetics as critically discussed by Tarutis et al. (1999) and Sapsford and Watson (2011). Low area-adjusted removal at relatively high pH in the pilot plant and particularly in wetlands is attributable to generally low hydraulic loading of the system. At the study site, catalytic processes such as heterogeneous oxidation of ferrous iron and homoaggregation of particulate ferric hydroxides take effect in the preceding seepage sump and water distribution system, where high (ferrous) iron concentrations in the range of 50-100 mg/L are effectively reduced to <15 mg/L before reaching the pilot plant. Therefore, little to no catalytic effects are to be expected within the actual pilot plant. Main purpose of the treatment system is the removal of residual suspended and, if present, dissolved iron for compliance purposes. Importance of efficient treatment components such as wetlands and sediment filters is clearly demonstrated by high treatment efficiency at (or despite) low inflow concentrations. Physico-chemical removal of suspended ferric iron clearly is the rate limiting step in the pilot plant and, based on the results of this study, there is a good cause to believe that the same may be true for other oxygenated and circum-neutral mine waters as previously suggested by Flanagan et al. (1994) and Mayes et al. (2009).

Available datasets clearly show decreasing treatment efficiency yet increasing area-adjusted removal with increasing hydraulic loading in accordance with findings of Tarutis et al. (1999). Future work will include further variation periods with intermediate, higher and lower flow rates to expand the database. In addition, high-resolution mass balances will be generated using continuously monitored turbidity as a proxy for (particulate) iron in the pilot plant. Finally, overall data will be used for statistically adequate performance evaluation and calculation of suspended ferric hydroxide removal kinetics as a basis for upscaling from pilot to full scale.

Conclusions

- Combining multiple treatment components with successively increasing efficiency to a composite system proved successful, reliably reducing iron concentrations from about 8.4 mg/L in the influent to compliance level (<1 mg/L) in the effluent;
- Treatment efficiency and area-adjusted removal showed inverse and direct relationships with hydraulic loading, respectively. Area-adjusted removal is relatively low because of low hydraulic loading (concentration dependence of natural iron removal processes);
- System monitoring provided evidence that physico-chemical removal of suspended ferric hydroxides is a critical step in natural iron removal and may even become the rate-determining step in treatment of thoroughly oxygenated, circumneutral mine waters;
- Hydraulic variation of the identical system lines provided excellent comparison data, yet a more extensive and higher resolution database is necessary for upscaling of the pilot plant to full scale.

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