IMPLEMENTATION OF AN IN SITU BUBBLE WALL FOR OXIDATION AND BIOREMEDIATION OF AN AQUIFER CONTAMINATED WITH AMMONIUM

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Summary

A field scale in situ oxygen bubble wall was implemented in a contaminated sandy aquifer for aerobic biotransformation of ammonium. Technology developments and tests referred to installation of gas-tigth sealed gas injection probes, flow controled gas injection regimes, and low volume and pressure conserved groundwater sampling. Estimated effective gas storage capacities and effective water-gas oxygen transfer rates from pilot scale experiments were taken into account for planning oxygen mass injection at the test site using a kinetic oxygen consumption mass balance model. The field experiment demonstrated the suitability of installation of an aerobic oxidation zone for complete in situ ammonium biotransformation. Chemical oxidation of sediment material and groundwater was induced simultaneously with autotrophic microbial degradation. No nitrification limitations by high dissolved oxygen concentrations were found and pH-buffering by calcite stabilized the nitrification process.

Introduction

Groundwater within the subsurface drainage area of the Berlin-Friedrichshagen waterworks (southeast of Berlin, Germany) is in part highly contaminated with ammonium due to former disposal of domestic and industrial sewage and drainage from the unsealed sludge-drying places of the Muenchehofe wastewater treatment plant. The contaminated aquifer has a volume of about 200 million m³. The ammonium concentration in the groundwater spans a range of 10-90 mg/L NH_4-N. Nitrate was detected only beneath the former irrigation fields in the upper part of the aquifer [1].

Former research and technology development activities of the Berlin Water Works focused on in situ oxidation of the contaminated aquifer by infiltration of highly oxygenated water and simultaneous pumping of contaminated groundwater (pump-and-treat, about 15 thousand m³/day during 20 years). It was shown by laboratory and field test investigations that oxygen concentrations of 100-150 mg/L in the infiltration water inhibited nitrification of ammonium by subsurface bacteria but only led to oxidation

of reduced iron and manganese and organic matter at the solid phase. There is a desorption limitation for ammonium at the sediment surface and there also exists a mixing problem of oxygenated infiltration water and groundwater which can take place only by hydrodynamic dispersion [1].

The idea is to build up a low-cost permeable bio-oxidation-zone within the drainage area of the waterworks to replace the time-consuming and cost-intensive pump-and treat decontamination concept. Oxygen gas is sparged into the aquifer to induce a fine dispersed, high surface residual trapped oxygen gas phase in the bubble wall zone which is able to supply suficient oxygen to the passing groundwater flow for stimulation and control of microbial oxidative degeneration of ammonium. The bubble wall will continuous or periodically be reloaded with oxygen gas when dissolution of residual oxygen is advancing and/or dissolved oxygen level in the sparging zone is falling [2, 3]. Recent modeling and pilot scale experimental work on fundamentals of gas sparging processes [4, 5, 6] needs to be evaluated and to further developed.

Reactive zones for in situ bioremediation of ammonium are the gas injection zone (bubble wall) and the downstream aquifer zone towards the wells of the waterwork. Thus effective mixing of oxygenized water with anaerobic water is generated using heterogeneous gas distribution in the wall and downstream dispersion without exceeding a critical dissolved oxygen concentration for nitrification in the bulk flow. Advantages of the aquifer as a reactor for nitrification are constant temperature and pH-buffering. Because of the large volume of the aquifer and low flow velocities of groundwater, required reaction rates of nitrification for effective in situ remediation can be small.

Bubble wall construction and monitoring program

A 15 month preparation and planning period was used to develop and test gas injection technologies (field scale), to investigate and estimate dynamic gas storage and water-gas oxygen transfer processes (pilot scale), and to test sensitivity of on line and in situ measurable parameters for spacial oxygen gas distribution detection (field and pilot scale). As a result of these tests it was concluded:

- to use a hydraulic pressing and a low-diameter drilling (with multilevel sealing injections against vertical gas channeling in the borehole zone) technique for installation of gas injection probes,
- to perform a flow controled gas injection regime for mass balancing and to interprete gas injection pressure behaviour as a parameter for dynamic spacial gas distribution processes,
- to use a combined geophysical borehole logging procedure for the quality check of borehole sealing and for detection of local gas saturation changes in the subsurface
- to estimate spacial gas storage capacities of the sandy aquifer material by gas saturations of 0,5 .. 5 % of pore volume,
- to estimate the dynamic water-gas oxygen transfer process being completed after passing 3 pore volumes of oxygen-free groundwater through the gas storage zone and to estimate this process being mainly finished (70 .. 80 %) yet after passing 1 pore volume of groundwater,
- to use dissolved oxygen sampling in the groundwater zone (by in situ pressure conserving and low volume sample techniques) and soil air monitoring for controling the oxygen gas supply process,
- to estimate gas injection mass flow using a kinetic oxygen consumption balance model with parameterization as given in [1].

The test site dimensions for site construction, hydraulic control and balance calculation were initially estimated of about 15 meters width, 35 meters length, and 25 meters depth. Groundwater surface was about 4 m below land surface. The aquifer was built of layered glacial sediments (fine to coarse sands) down to about 30 m below land surface. It could be divided schematically into a deeper (27 ..

20m below surface) and a upper (18 .. 8m below surface) layer which were separated by a highly compacted fine sand layer.

The gas injection field consists of 10 injection elements at 4 different depths (probes BIL / DIL at 10, 15, 20, 27 m below surface). Horizontal distance between the injection probes was 4 m. Groundwater flow was forced througth the test site by slow pumping (well EB located about 30 m downstream of the injection zone). A total of 16 sample points of low-volume pumping (MDP), 8 sample points of pressure conserved sampling (BAT) and up to 8 multilevel observation wells were included in the groundwater process monitoring. There were also integrated 10 soil gas sampling probes (GS) and 4 water level detectors.

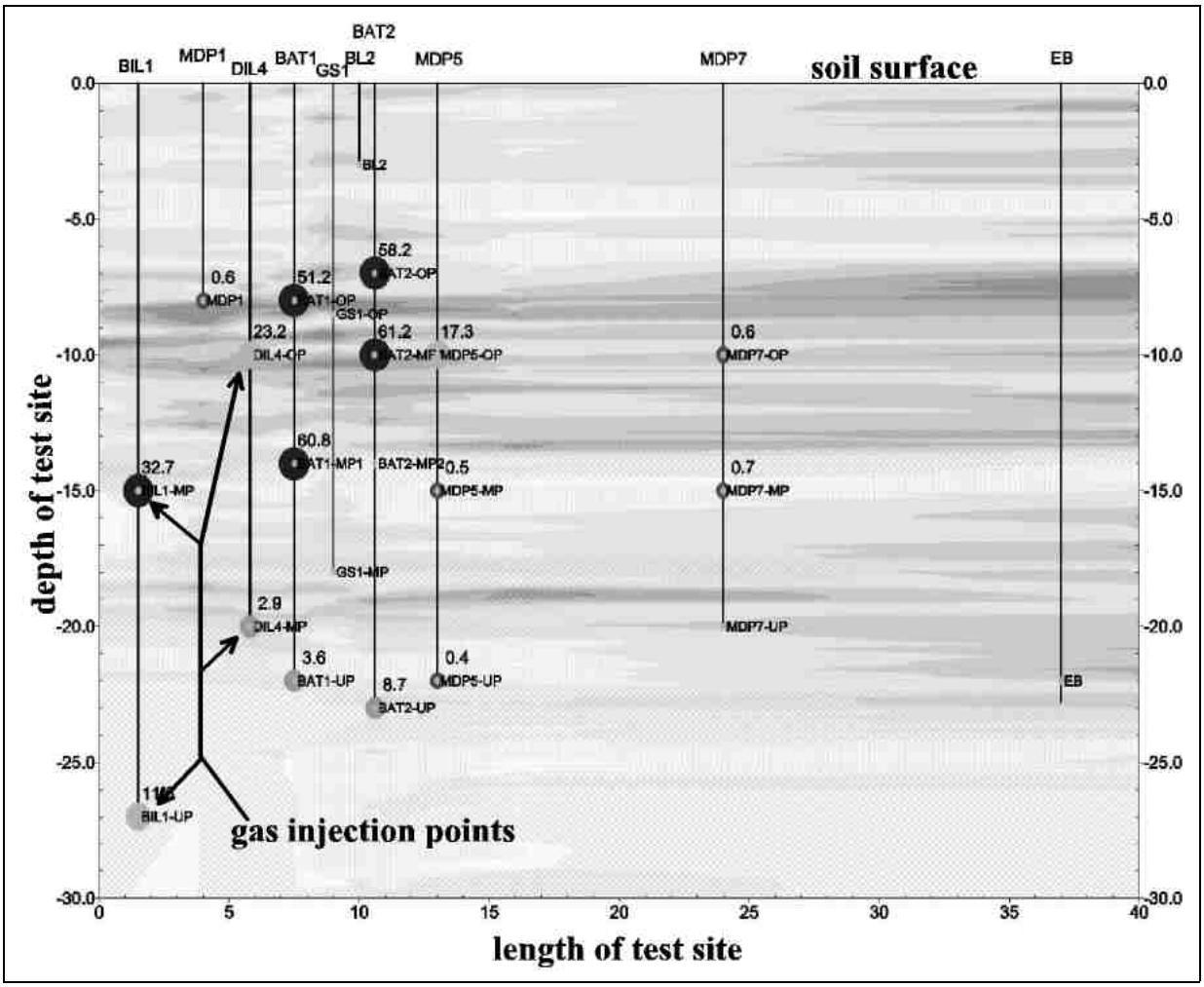

Fig. 1 Cross section of the constructed test site. Implementation of the in situ bio-oxidation wall.

A scheme of ammonium biotransformation processes is given in [1, 7]. The main process monitoring program consisted of measurements of dissolved and gaseous oxygen and carbon dioxide in the groundwater and soil air, of nitrogen components (dissolved nitrogen, ammonium, nitrate, nitrite), DOC, cations (K, Na, Ca, Mg, Fe, Mn) and anions (carbonates, sulfate, chloride) in the groundwater and aqueous environmental parameters (pH, Eh, EC, T). Sampling intervalls ranged from 1 to 2 weeks.

Field experiment

The field experiment started on July, 1st, 2002, after a reference state measurement at all sampling points. There were neither dissolved oxygen nor nitrate in the aquifer. Groundwater ammonium concentrations spanned from 10 to 20 mg_{NH4-N}/L_W. The parameter Eh was at about +150 .. +250 mV and pH at 7,1 .. 7,2. The goal of the initial gas injection (period I) was to achieve a high level of oxidation of solid aquifer material in the bubble wall zone to generate a stable aerobic environment for iniciation of nitrification processes. A discontinuous oxygen gas injection regime at all 10 injection points was chosen according to pilot scale experiments for oxygen gas storage and gas-water oxygen transfer. A kinetic oxygen consumption balance model was used for gas injection mass estimations at the test site. Injection period I was stopped after 33 days. About 800 kg of oxygen gas were supplied (34 kg_{O2}/d) and aerobization was detected in the intermediate (18 .. 12 m below land surface) and upper layers (12 .. 8 m below land surface) of the aquifer by elevated redox potentials (Eh > 350 mV) and dissolved oxygen in the groundwater (5 .. 65 mg_{O2}/L_W). Spreading of dissolved oxygen up to 12 m far from the gas injection points was only possible to explain by gaseous oxygen transport in horizontal subsurface layers. Elevated oxygen concentrations of about 30% indicated to losses of injected gas into the soil air. A very slow beginning of nitrification (<0,5 mg_{NO3-N}/L_W) at single sample points was also detected.

No oxygen gas was supplied to the aquifer in period II during 19 days and it was found that aerobization processes ceased rapidly. Variable tailing behaviour of dissolved oxygen in the pore water was explained by heterogeneous oxygen gas transport and local storage processes. A more effective oxygen supply to the western part of the test site was observed where disturbed zones around groundwater monitoring wells are located.

Reiniciation of oxygen gas supply in period III was performed by continuous injection at one of the deepest (27 m below land surface) injection points. It was intended to stabilize the existing oxidation zone by a single point injection, to create an aerobic environment in the deep aquifer layer (27 .. 20 m below land surface) and to minimize gas losses to the soil air. About 1.100 kg oxygen gas were injected during 63 days (17 kg_{O2}/d). It was shown that oxygen gas transport had a ROI of at least 20 m in horizontal (up- and down-stream of groundwater flow) and vertical direccions. While the intermediate aquifer layer showed high dissolved oxygen levels (10 .. 100 mg_{O2}/L_W) over large distances the upper und deeper layers were only partially influenced by the gas injection. Nitrification became significant in the western part of the test site (2,0 .. 10,0 mg_{NO3-N}/L_W) and ammonium in the groundwater subsequently started to lower.

Full scale implementation of the in situ bubble wall was achieved in injection period IV. Continuous oxygen gas supply at the deep injection point was increased to 34 kg_{O2}/d during 48 days and amplification of the nitrification zone was continued at all aquifer layers. Six month after starting oxygen gas injection at the test site ammonium biodegradation became an effective process lowering ammonium concentrations below 1,0 mg_{NH4-N}/L_W. A stable permeable bio-oxidation wall characterized by dissolved oxygen and nitrate was formed at the injection zone. The main process was accompanied by pyrite oxidation (increase of sulfate) and a pH-decrease to 6,7 .. 6,9 where it is stabilized by calcit-buffering (increase of calcium).

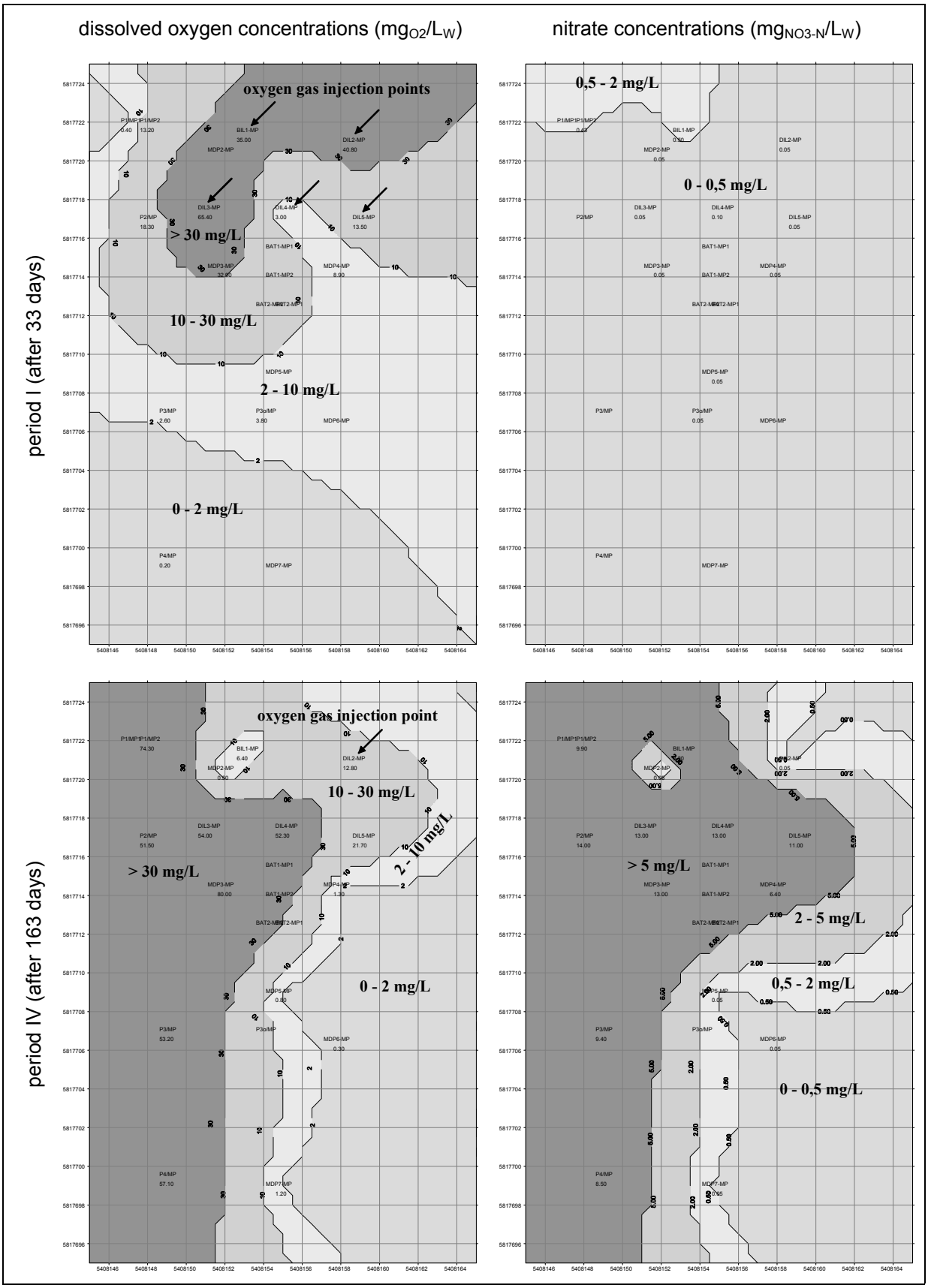

Fig. 2 Development of the in situ bio-oxidation wall in the intermediate layer (18 .. 12 m below land surface), dissolved oxygen and nitrate

Discussion

Observation of gas transport in the aquifer during sparging by dissolved oxygen and soil air oxygen measurements permits a sufficient quick gas injection control. No detection method of sufficient resolution was found for stored gas volumina in the aquifer and/or gas-water interface areas. Long-term oxygen delivery to the groundwater flow from stored (immobilized) gas bubbles was found a generally non-effective oxygen supply process due to storage mass capacity limitations (low gas saturations and heterogeneous gas distribution structures). So a continuous oxygen supply to the aquifer with some losses to the soil air is recommended when there are no stripping problems. Large ROI of gas distribution of single injection points were found caused by field scale heterogeneities. In our case horizontal ROI size was of the same magnitude as vertical distance of the gas injection points to the upper aquifer limit (water table). Deep aquifer basis zones are difficult to manipulate with gas sparging systems (small ROI of gas distribution at injection point depth).

For biotransformation processes no effective nitrification limitation by a maximum dissolved oxygen level (50 mg/L in [1]) was detected. Simultaneous chemical aquifer oxidation and autotrophic microbial nitrification of ammonium were established in the in situ bio-oxidation wall. The lag-phase of the nitrification process was of about 30 .. 50 days. Stable nitrification was achieved at pH < 6,95 and Eh > 350mV. The problem of long term pH-buffering of in situ nitrification processes needs further attention. Large reaction and mixing zones (hunderds of meters) are needed for application of the in situ bio-oxidation wall technolgy to effectively protect the drinking water wells of the Berlin-Friedrichshagen waterworks.

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