Saph Pani

Enhancement of natural water systems and treatment methods for safe and sustainable water supply in India

Project supported by the European Commission within the Seventh Framework Programme Grant agreement No. 282911

Deliverable D 1.3

Concept for application of BF in aquifers contaminated with nitrogen species

– Delhi as an example for BF at sewage influenced surface waters

SAPHPANI

Work package	WP1 Bank Filtration
Deliverable number	D 1.3
Deliverable title	Concept for application of BF in aquifers contaminated with nitrogen species
Due date	Month 30
Actual submission date	Month 37
Start date of project	01.10.2011
Participants (Partner short names)	KWB, FUB
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Dissemination level: (**PU**blic, Restricted to other **P**rogrammes **P**articipants, **RE**stricted to a group specified by the consortium, **CO**nfidential- only for members of the consortium)	PU
Deliverable Status:	Final

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1 Introduction

1.1 Bank filtration

Bank filtration (BF), sometimes also called riverbank filtration (RBF), is used worldwide for drinking water production (Tufenkji et al. 2002, Doussan et al. 1997, Grünheid et al. 2005). It has two main advantages: (1) Sufficient quantity of water can be produced independent of the usable groundwater capacity as BF is a form of artificial groundwater recharge (Bouwer 2002, Dillon 2005). (2) Low cost post treatment is often sufficient for the raw water as the process of bank filtration takes advantage of the natural filter capacity of the sediments during the soil passage (Kuehn and Mueller 2000). Usually, there is a significant increase in water quality for the bank filtrate compared to the surface water regarding organic substances, colour, coliform bacteria and faecal contaminants (Singh et al. 2010, Weiss et al. 2005). Additionally, the water quality at bank filtration wells is relative constant, and, therefore, it is easier to treat than surface water, which often shows high variation in many quality parameters (Ray 2004, Tufenkji et al. 2002).

Typically, bank filtration sites are planned according to the local hydrogeological conditions and the wells are constructed to achieve both of the above mentioned effects (Ray et al. 2002). In developing countries there is usually an emphasis on securing sufficient water quantity for drinking. Wells are often constructed along rivers and lakes because alluvial or riparian aquifers generally have good hydraulic properties (Rosenshein 1988). The shallow depths of the sediments make them easy to exploit and help to reduce drilling costs (Doussan et al. 1997). When bank filtration is applied at sewage-contaminated surface waters, which is often the case in developing countries (Ray 2008), a range of problems can arise as contaminated water infiltrates into the aquifer in large quantities and the capacity of the soil to filter the contaminants is often exceeded (Heberer 2002). The resulting contamination of the aquifer can prevail for many decades, making post treatment and/or remediation measures necessary. Such contamination is often caused by nitrogen, especially the species ammonium (Hiscock and Grischek 2002).

1.2 Nitrogen

1.2.1 Occurrence and effects

Nitrogen is a redox-sensitive parameter, which can occur in different species. The most common forms of nitrogen in the water-soil environment are, in order of decreasing oxidation state (Metcalf & Eddy Inc 2014, Stumm and Morgan 1996):

- Nitrate (NO_3^- , +V)
- Nitrite (NO_2^-, +III)
- Nitrogen gas (N_2, 0)
- Ammonia and ammonium (NH_3, and NH_4^+, both -III)
- Organic nitrogen (OrgN, mostly –III)

Whether the reduced form of nitrogen occurs as un-ionized ammonia (NH_3) or in the form of ammonium ions (NH_4^+) depends on the temperature and, to a stronger extent, the pH of the solution:

$NH_3 + H_2O \leftrightarrow NH_4^+ + HO^-$

10%	90%	at pH 8.3 and 20°C (Metcalf & Eddy Inc 2014 p. 94)
50%	50%	at pH 9.25 and 20°C (Metcalf & Eddy Inc 2014 p. 94)

At pH and temperature conditions commonly found in natural waters, ammonium is the principal species (Hem 2005). Sometimes the term “total ammonia” is being used referring to the sum of ionized and un-ionized ammonia.

Nitrogen pollution can cause problems such as eutrophication of surface water bodies (Howarth and Marino 2006), which can lead to toxic algal blooms or decreasing dissolved oxygen concentrations and related issues such as a decrease in animal and plant diversity. Furthermore, ammonia (NH_3, un-ionized) is toxic for aquatic species (Randall and Tsui 2002), but not for humans at low concentrations (GESTIS Substance Database 2014). Nitrite (NO_2^-) is also extremely toxic to fish or other aquatic species (Metcalf & Eddy Inc 2014). For humans, excessive nitrogen intake in the form of nitrate or nitrite through water can result in diarrhoea or methaemoglobinemia (blue-baby syndrome) in infants (Ward et al. 2005). When chlorination is used for the disinfection of drinking water, the presence of ammonium in raw water, even at low concentrations, causes the formation of chloramines (Weil and Morris 1949). Higher chlorine doses are necessary to achieve required minimum residual chlorine concentration at the outlet of the water treatment plants (WTPs) and within the distribution system (Duong et al. 2003).

1.2.2 Guideline values

Guideline values for the different nitrogen species given in the German drinking water ordinance (TrinkwV 2001), the Indian standard (BIS 10500:2012), and the WHO drinking water quality guidelines (WHO 2011) are compared in Table 1. In the German TrinkwV, the guideline value for ammonium is 0.5 mg/L. This low guideline value is due to the fact that ammonium is defined as an indicator parameter of possible bacterial, sewage, or animal waste pollution. The cause of an increase from the usually measured concentrations has to be investigated. The WHO has not established a guideline value for total ammonia because it usually occurs in drinking water at concentrations well below those of health concern. Because the WHO includes the non-ionized form NH_3 and the ionized form NH_4^+ in their definition of ammonia, it is assumed that this is also the case in the Indian Standard BIS 10500, although it is not further defined.

Table 1 Guideline values for nitrogen species in drinking water

Parameter	**Unit**	**TrinkwV (Germany)**	**BIS 10500 (India)**	**WHO**
Nitrate (as NO_3)	mg/L	50	45	50
Nitrite (as NO_2)	mg/L	0.5 (0.1 at the outlet of the water works)	No guideline value	3
Ammonium (as NH_4^+)	mg/L	0.5	---	---
Total ammonia (as NH_3-N and NH_4^+-N)*	mg/L	---	0.5	No guideline value

* Definition of WHO, not specified in BIS 10500

1.2.3 Nitrogen in surface water bodies

In surface water bodies, nitrogen concentrations depend on several factors, mainly land use, sewage disposal and the water balance of the water body, as dilution strongly affects the pollutant concentrations. In European rivers agricultural fertilizers are the main source of nitrogen. NH_4^+-N concentrations range between 0.1 and 0.3 mg/L and median NO_3-N concentrations are around 3 mg/L in large rivers (EEA 2001a). In urban agglomerations in developing or newly industrialized countries nitrogen loads in surface water bodies are expected to be substantially higher: Nitrogen contamination of surface water through untreated or partially treated domestic sewage water is a concern in many of these countries. According to Corcoran et al. (2010), up to 90% of sewage water in these countries is not collected or treated but discharged directly into rivers, lakes and coastal areas or leached into the subsoil. In the Asia-Pacific region alone, this amounts to approximately 150–250 million m^3 per day of untreated (domestic) wastewater from urban areas released to the environment untreated (WWAP 2012). Data on total inorganic nitrogen concentrations (NH_4^+, NO_2^-, NO_3^-) is scarce, as most studies on river water quality in Asian megacities only report concentrations for nitrate and nitrite, e.g. Sikder et al. (2013) and Kido et al. (2009). But because of the high chemical oxygen demand (COD) in sewage-contaminated rivers, the prevailing form of inorganic nitrogen is expected to be ammonium.

1.2.4 Nitrogen in sewage water

In sewage water about 60-70% of the nitrogen is present in the form of ammonia/ammonium, depending on the pH of the solution, while the remaining 30-40% are mostly found as biodegradable or non-biodegradable organic nitrogen (Metcalf & Eddy Inc 2014 p. 712). Fresh domestic wastewater usually contains no more than 1% nitrates and nitrites before it is aerated in the nitrification step at sewage treatment plants (STPs) (Sedlak 1991). Total nitrogen concentrations in wastewater are highly variable depending on the diet of the population (Pescod 1992, Patterson 2003) and the per capita wastewater flow rate (Sedlak 1991). Concentrations between 20 and 85 mg/L are reported as an average for typical domestic wastewater in Metcalf & Eddy Inc (2014), and maximum concentrations can be much higher (sometimes above 150 mg/L – Ammary 2007, Al-Salem 1987 cited by Pescod 1992). The main source of nitrogen in wastewater is urine. The prevailing component of urine is urea, which is degraded by biological hydrolysis (Mobley and Hausinger 1989):

Urea + water $^*\rightarrow$ ammonia + carbamate

$(NH_2)_2CO + H_2O \;^*\rightarrow NH_3 + H_2NCOOH$

* naturally occurring enzyme urease catalyzes the reaction

Carbamate + water → ammonia + carbonic acid

$H_2NCOOH + H_2O \rightarrow NH_3 + H_2CO_3$

The carbonic acid dissociates:

$H_2CO_3 \rightarrow H^+ + HCO_3^-$

Ammonia molecules equilibrate with water:

$2NH_3 + 2H_2O \rightarrow 2NH_4^+ + 2OH^-$

Other sources are faeces (a source of OrgN) and grey water from laundry and personal washing (a source of NH_3/NH_4^+) (Patterson 2003). Products like toilet paper had the lowest nitrogen load contribution (Tjandraatmadja et al. 2010). In countries without a regular waste disposal system, garbage disposal in wastewater is another large source of organic nitrogen.

1.3 Motivation

Increasing ammonium concentrations at one specific well (P3) in a well field located at the Yamuna River in East Delhi are a cause for concern as the well field is used for drinking water production. Elevated ammonium concentrations at the well were reported since 2006 and Groeschke (2013) and Sprenger and Lorenzen (2014) identified the sewage-contaminated river water as the main source of ammonium in the raw water. Ammonium concentrations are already about ten times higher (5.5-8 mg/L) than the Indian guideline value of 0.6 mg/L total ammonia [0.5 mg/L total ammonia-N] as specified in BIS 10500:2012 and are expected to increase further. As water suppliers have to plan several decades ahead in order to be able to develop appropriate water management concepts, it is important to know about the development of future ammonium concentrations at that well field and other well fields along the Yamuna River to be able to choose appropriate treatment and/or remediation options.

Numerous studies of ammonium contaminations in groundwater have been conducted, mostly focusing on contamination from point sources - such as septic tank effluents (Hinkle et al. 2007), leachate from sewage farms (Hamann 2009), leachate from coking plants (Haerens 2002a) or chemical companies (Clark et al. 2008) - and from contaminations resulting from the infiltration of treated sewage water (LeBlanc 1984, Ceazan et al. 1989, Böhlke et al. 2006, DeSimone and Howes 1996 and 1998). A comprehensive review about published literature on ammonium retardation is given by Buss et al. (2004). Doussan et al. 1997 and Doussan et al. 1998 studied the transport of nitrogen species at a RBF site at the Seine (France), where the river water was a main source of nitrogen – in the form of nitrate. Reducing conditions prevailed in the aquifer owing to the decay of organic matter and the nitrate was reduced to ammonium during the soil passage, while the mineralization of organic matter was an additional source of ammonium. In central Delhi, reducing conditions in the aquifer are caused by the infiltration of reducing surface water. It is expected that without infiltration of reducing surface water, a redox sequence from oxidized conditions to iron-reducing conditions further away from the river would prevail - as reported by Lorenzen et al. (2010a) for a field site upstream Delhi.

In order to understand the behaviour of ammonium in aquifers at BF sites with surface waters highly polluted by untreated sewage, field data have been collected and laboratory column studies have been conducted with aquifer material from Delhi. Results of the analyses and experiments are summarized in chapter 2 and were used as the basis for recommendations about the application of BF in nitrogen-contaminated aquifers.

1.4 Objective

The objective of this report is to give recommendations for remediation and treatment concepts for BF sites in aquifers contaminated with inorganic nitrogen species. The focus lies on the reduced species ammonium, which occurs at BF sites located at sewage contaminated surface water bodies. The authors will give a comprehensive overview of existing and emerging remediation and treatment options and discuss them critically regarding the site-specific requirements and the challenges arising in the Indian context.

2 Description of the Delhi case study

2.1 Overview

The well fields in East Delhi are an example of unintended RBF use by placing wells along a sewage-contaminated river. Delhi is a megacity with a population of currently 16 million people (Census of India 2011) located in central India in the Indo-Gangetic Plain (Fig. 1). The Yamuna River, the largest tributary of the Ganges River, flows through Delhi in north-southerly direction. As many rivers in Asia, the Yamuna is a braided river system characterized by high sediment loads and a constantly changing riverbed. Although numerous dykes and embankments were constructed within the city to control the flow, the river still has the opportunity to shift within certain limits (Fig. 2). Thus, with or without embankments, the location of the riverbank frequently changes. The river is dammed up by two barrages within the city area, Wazirabad barrage in the North and Okhla barrage in the South, and the river stretch between the two barrages is highly polluted by sewage water.

The floodplain extends up to a width of about 2.5 km on the eastern and western banks along the entire stretch of the river in Delhi. It is mostly undeveloped and mainly used for agriculture. The floodplain sediments are mostly medium grained sands with a thickness of up to 70 m in the north of Delhi (Shekhar and Prasad 2009) and about 20 m in the southern part of the city. They constitute what is known as the floodplain aquifer or Newer Alluvium. Compared to other groundwater sources in Delhi, the groundwater of the Newer Alluvium is found at shallow depth and is characterized by only minor water table fluctuations of about 0 - 2 m throughout the year and over decades - as opposed to about 4 - 20 m in other aquifers in Delhi (CGWB 2012 and Shekhar et al. 2009).

Numerous tube wells and about 20 Ranney wells (radial collector wells) were constructed on the floodplain in Delhi, tapping the Newer Alluvium. The wells are not arranged parallel to the riverbank but were constructed across the complete width of the upper floodplain. Owing to losing stream conditions (Lorenzen et al. 2010a) it can be assumed that the wells situated close (200 – 500 m) to the river draw a high share of bank filtrate, although they have not been specifically designed for bank filtration.

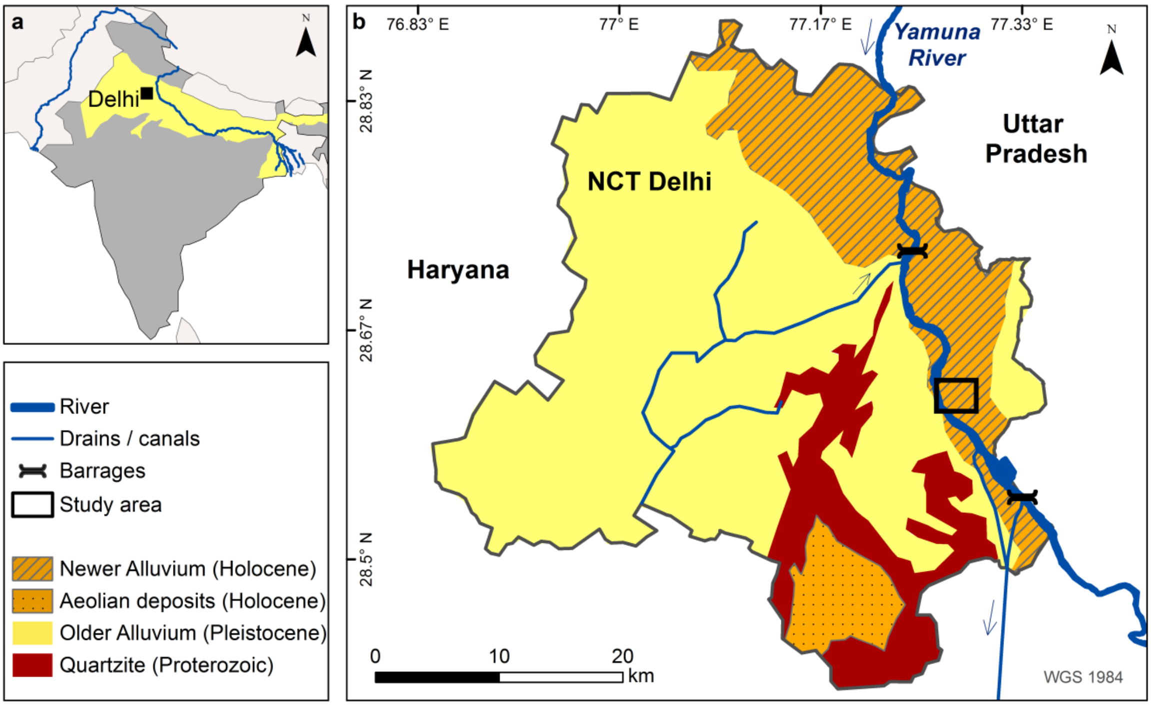

Figure 1 a. Location of Delhi in the Indo-Gangetic plain (yellow). Data source: Natural Earth (2011) b. The study area is located on the East bank of the Yamuna River, where sewage-influenced river water infiltrates into the sediments of the Newer Alluvium. Geological Map modified after Geological Survey of India (2006)

2.2 The study area

2.2.1 Sampling points

The study site covers an area of about 2.5 km^2 on the east bank of the Yamuna River in East Delhi near the Akshardham Temple between 720800 and 722800 m E and 3165900 and 3168200 m N (UTM, WGS84 Zone 43 N). Here, the undeveloped floodplain is 2.4 km wide. Four Ranney wells operated by the Delhi water company Delhi Jal Board (DJB - P3, P4, P5, P6) and one Ranney well operated by the Indian Railways (NH24) as well as numerous hand pumps and bore wells used by the local population are located within the area and were partially used for water sampling. Additionally, four observations wells were installed at a distance of about 500 – 550 m to the riverbank and two hand pumps were installed at distances of 35 m and 250 m to the river in the frame of the Saph Pani project. The locations and type of sampling sites are shown in Figure 2.

2.2.2 Previous research

Previous research at this location was conducted 2006 in the frame of the feasibility study IDB India (International development of bank filtration: Case study India) and 2007-10 in the frame of the TECHNEAU project (Lorenzen et al. 2007, Pekdeger et al. 2008, Sprenger et al. 2008, Lorenzen et al. 2010a, Lorenzen et al. 2010b, Lorenzen 2011, Sprenger 2011, Sprenger and Lorenzen 2014).

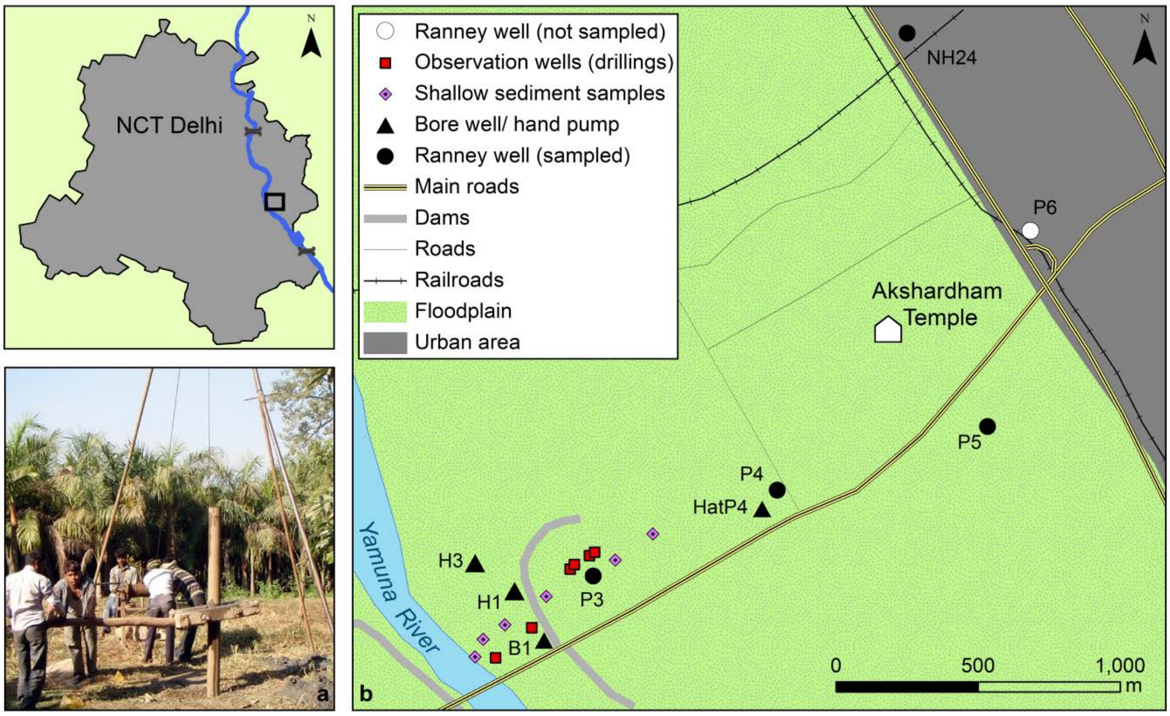

Figure 2 a. Drilling of observation well. b. Location of the hand pumps, Ranney wells and of the shallow and deep drillings conducted during the field work (modified after Groeschke 2013)

2.2.3 Geology and hydrogeology

In the study area, like in the entire flood plain in Delhi, the Holocene alluvial sands of the Yamuna floodplain (Newer Alluvium) are underlain by finer grained Pleistocene sediments of the Older Alluvium. At 38 mbgl (meter below ground level) Precambrian bedrock was encountered (Sprenger 2011 p. 70). Hydraulic conductivities of the Newer Alluvium are in the range of 2×10^{-4} to 7×10^{-4} m/s (Chatterjee et al. 2009) and Sprenger (2011, p.66) reported an average travel time of 0.9 m/d for this unit at the field site. The hydraulic conductivities of the Old Alluvium are between 3×10^{-5} and 5×10^{-5} m/s (Chatterjee et al. 2009). According to Lorenzen et al. (2010a) losing stream conditions prevail on the East bank of the river. Sprenger (2011 p.66) reported infiltration rates of 6.4×10^{-7} $m^3/m^2/s$ for monsoon times and 4.2×10^{-7} $m^3/m^2/s$ for non-monsoon times. For the west bank of the river at this location no information is available.

2.2.4 Description of the production wells

The Ranney wells of the Delhi Jal Board in the study area were constructed in 1973 and started operation in 1975. They are between 15 and 20 m deep (personal communication DJB 2012) and thus tap the floodplain aquifer. Each well has ten laterals, which are each about 30 m long. The recorded discharge of the wells is about 150 – 300 m^3/h (Chatterjee et al. 2009). The wells are typically operated about 8 hours every day, except for well number P3, which is sometimes not operating because of elevated ammonium concentrations in the groundwater. The water of well P4 is directed to the Common Wealth Games Village WTP while the water of wells P3, P5, P6 is supplied to the Okhla

WTP. Well NH24 of the Indian Railways is constructed similar to the DJB Ranney wells, but the water is not used for public water supply.

2.3 Field studies

2.3.1 Water and sediment sampling

During the SAPH PANI project, 72 groundwater samples were taken from the sampling points shown in Figure 2. In addition, eleven regular river water samples and two samples of the flood event in July 2013 were taken and analyzed for NO_3^-, NO_2^-, NH_4^+, main cations and anions, pH, ORP, EC, dissolved oxygen, trace elements. Sediment samples were taken from depths up to 28 m by manual auger drilling (Fig. 3a) at six locations at distances of 35 m, 250 m and 500 m to the riverbank. In addition, seven shallow drillings (up to 4 m) were done at distances of 5 m, 75 m, 200 m, 375 m, 500 m, 600 m, and 800 m to the river using an Eijkelkamp hand drilling device (Fig. 3b). To sample the river bottom sediments and to measure water depths, three profiles were taken across the Yamuna River in December 2013, using a Van-Veen-Grab Sampler (Fig. 3c).

Figure 3 a. Manual auger drilling b. Shallow drilling with Eijkelkamp hand drilling device c. Sediment sampling of river bottom sediments

2.3.2 Ammonium concentrations at the field site

Varying ammonium concentrations were found in the aquifer close to the river. In 2012, the trend in ammonium concentrations at the three sampling points B1, H1, and H3 was similar with values between 4.5 mg/L in June 2012 and 26 mg/L in December 2012 (Groeschke 2013). In 2013, ammonium concentrations still fluctuated between 6.4 and 35 mg/L, but no trend could be discerned. In the Ranney well P3 at a distance of 500 m from the river, ammonium concentrations varied between 5.5 and 8 mg/L in 2012 and 2013. In wells farther away from the riverbank, ammonium concentrations remained below 1.65 mg/L in both years. In the river water, ammonium concentrations up to 20 mg/L were measured in 2012 and up to 16 mg/L during the field campaigns in 2013. Maximum and minimum concentrations at the field site are summarized in Figure 4. Data obtained from water samples were further used to set-up the 1D model described in section 2.5.

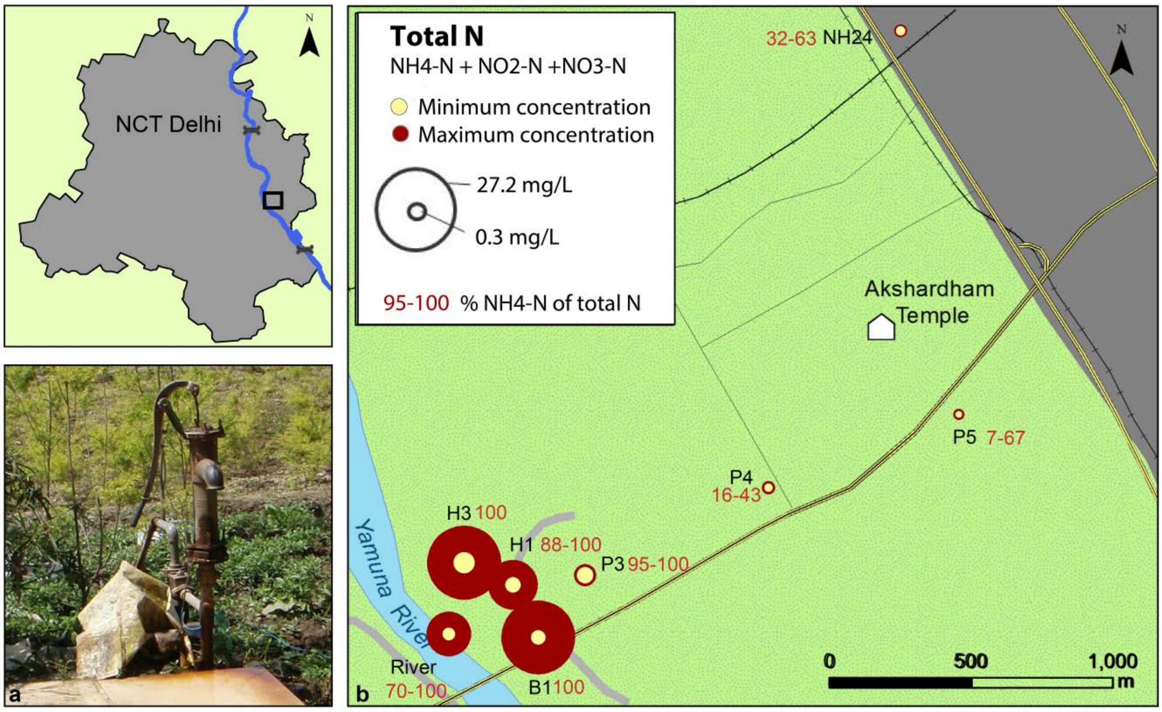

Figure 4 a. Hand pump H1 b. Minimum and maximum total nitrogen concentrations in the water samples taken between March 2012 and December 2013

2.4 Laboratory studies

2.4.1 Sediment analyses

To characterize the alluvial aquifer, a total of 14 sediment samples were taken from one shallow (3 m) drilling at a distance of 5 m from the riverbank and from three deeper drillings with depths between 15 and 33 m at a distance of 500 - 550 m from the riverbank (Fig. 5). The water contents varied from 5% to 38% increasing with decreasing particle size, which is in accordance with respective literature (e.g. Genske 2006, Schultze and Muhs 1967). An increase of water contents with higher organic portions could not be clearly identified. This contradicts respective literature (e.g. Schultze and Muhs 1967), but could well be due to dewatering during the long transport of material from the field site to the laboratory.

The sediment's main components ranged between silt and gravel. The particle sizes increase with increasing sampling depths. Silty fine sands are predominant in the unsaturated zone, while the saturated zone is dominated by well-sorted medium sands. At a depth of about 13.5 m a gravelly layer has been encountered, which is underlain by silty clays, presumably of the Old Alluvium at a depth of about 16 m. In the Newer Alluvium, the transition between beddings is smooth and graded. The hydraulic conductivity (k value) ranged between 2.6×10^{-6} m/s and 2.3×10^{-4} m/s if calculated according to Hazen (1893) and between 7.7×10^{-5} m/s and 1.6×10^{-4} m/s if calculated according to Beyer (1964). The hydraulic conductivities increase with increasing particle size and thus with depth. The gravelly layer consists of real gravel particles (grain size of > 2 mm) and of concretions > 2 mm, which dissolve into sand and silt when shaken with sodium pyrophosphate ($Na_4P_2O_7$) for 24 hours.

Organic contents and carbonate contents were measured through loss on ignition. Organic contents of the sediments range between 0.5% and 2.8%. Samples with higher portions of fine grains also contain the highest share of organic substances. The carbonate contents of the sediments range between 1.5% and 18%. The highest carbonate contents were found in the gravel layer, which contains the concretions.

The cation-exchange capacity (CEC) was determined using a $BaCl_2$ percolation method for the sands and gravels and a $BaCl_2$ batch method for the fine grained sediments. The results range between 1.2 meq/100 g sediment in the saturated zone and 37.2 meq/100 g sediment in the unsaturated zone. Calcium had the highest share in the CEC, leading to the assumption that the carbonates consist mainly of calcium carbonate and the carbonatic concretions are probably the typical calcite concretions locally known as kankar (Eybing 2014).

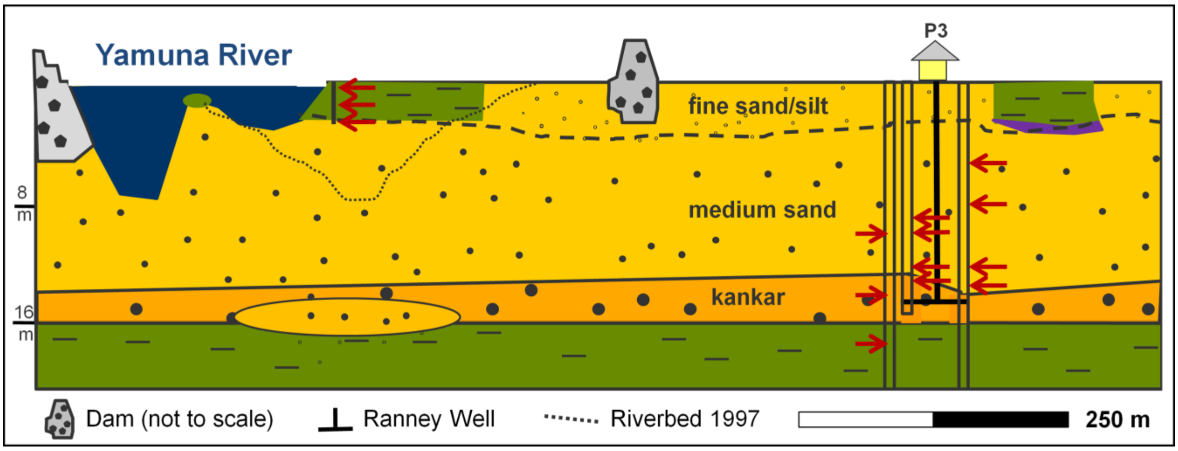

Figure 5 Sampling locations of the 14 sediment samples (arrows) used for the sediment analyses

2.4.2 Column experiments

The transport and fate of ammonium in the sand and kankar aquifer materials from the Yamuna floodplain was further investigated in laboratory column experiments at Freie Universität Berlin. Such experiments are a common method in hydrogeology to determine specific sediment parameters. The goal of these series of experiments was to provide data regarding sorption, degradation and fixation of ammonium under field site conditions. The data were later used to set up a reactive transport model of the field site to predict the future development of the ammonium plume. The set-up of the experiments is shown in Figure 6.

The experiments were conducted under suboxic or anoxic conditions, whereby the latter prevail in the aquifer. To achieve laboratory conditions similar to those at the study site, the following parameters were adapted:

- The model water was adjusted for the main cations to be comparable to the concentrations in the groundwater.
- Anoxic conditions were established by using argon balloons to create an oxygen- (and nitrogen-) free atmosphere above the model water container and the effluent sampling flasks. Glass and gas-tight tubing materials were used to inhibit gas exchange at the connections. The oxygen concentration was monitored with

chemical optical oxygen sensor spots (PreSens) at six points during the entire duration of the experiments. Redox potential was measured using ORP-probes at the outlet of the columns.

- A peristaltic pump maintained a flow rate of ~0.17 mL/min, which correlates to a flow velocity at the field site of about 0.9 m/d (Sprenger 2011 p.66).

The glass columns had an inner diameter of 45 mm and a sediment filled length of 146 mm and were flushed upflow with the model water. Three sets of experiments were conducted with this set-up: pre-tests, adsorption experiments and desorption experiments.

- During pre-tests, the freshly filled columns were flushed with nitrogen-free model water until nitrogen concentrations in the effluent were sufficiently low.
- In the adsorption experiments, the columns were flushed with model water with ammonium concentrations of either 20 mg/L or 10 mg/L until the ammonium concentrations in the effluent were equal to the concentrations in the model water.
- During the subsequent desorption experiments, the columns were again flushed with nitrogen-free model water until nitrogen concentrations in the effluent were low and did not decrease any further.

Each experiment was conducted with two or three columns filled with the same sediment (duplets or triplets). To check for reproducibility, most experiments except for the pre-tests were repeated one or two times.

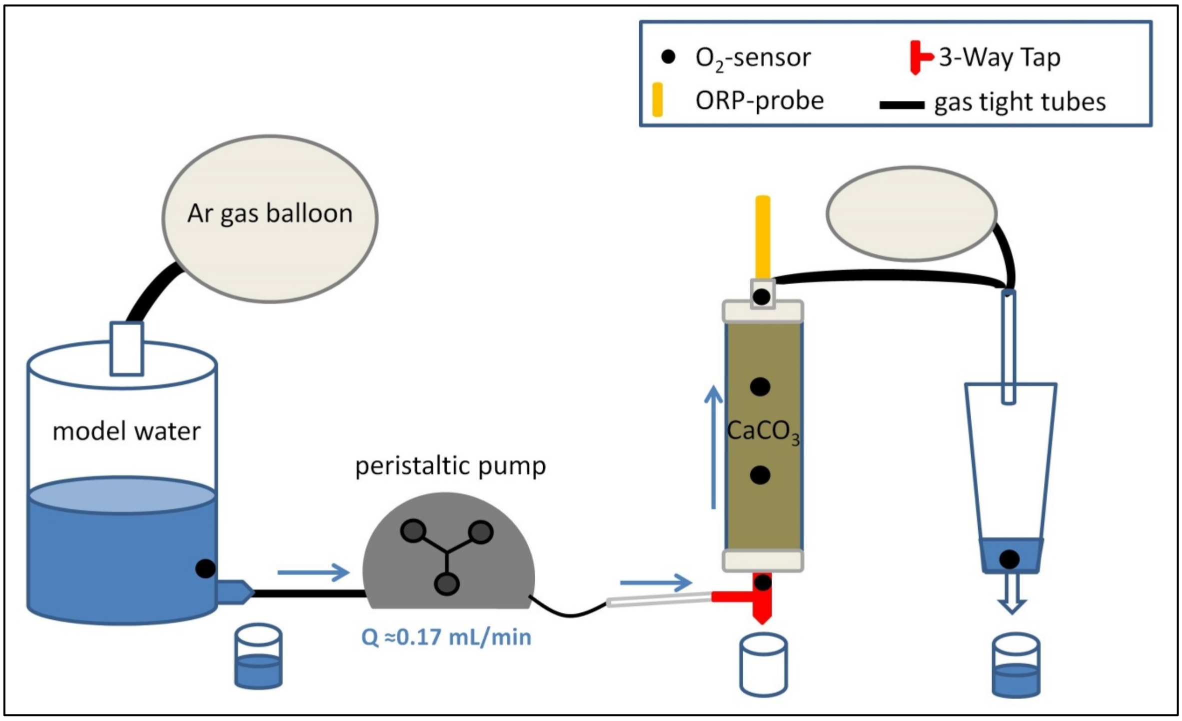

Figure 6 Set-up of the column experiments

The column experiments indicate that there is some degradation or fixation of ammonium in the sediments of the unsaturated zone and no or very little natural degradation potential in the sediments of the saturated zone. The transport of ammonium is, therefore, mainly controlled by cation exchange. In the sand, it took 10 - 12 pore volumes to have a 100% breakthrough of ammonium in the columns and about 15 pore volumes to flush the ammonium out of the sediment. In the gravel, 30 - 35 flushes were necessary to observe the 100% breakthrough in the adsorption experiments and the flushing of the ammonium in the desorption experiments took about 40 pore volumes (Groeschke et al. 2014). The breakthrough curves of the sand and the kankar-gravel samples are shown in Figure 7.

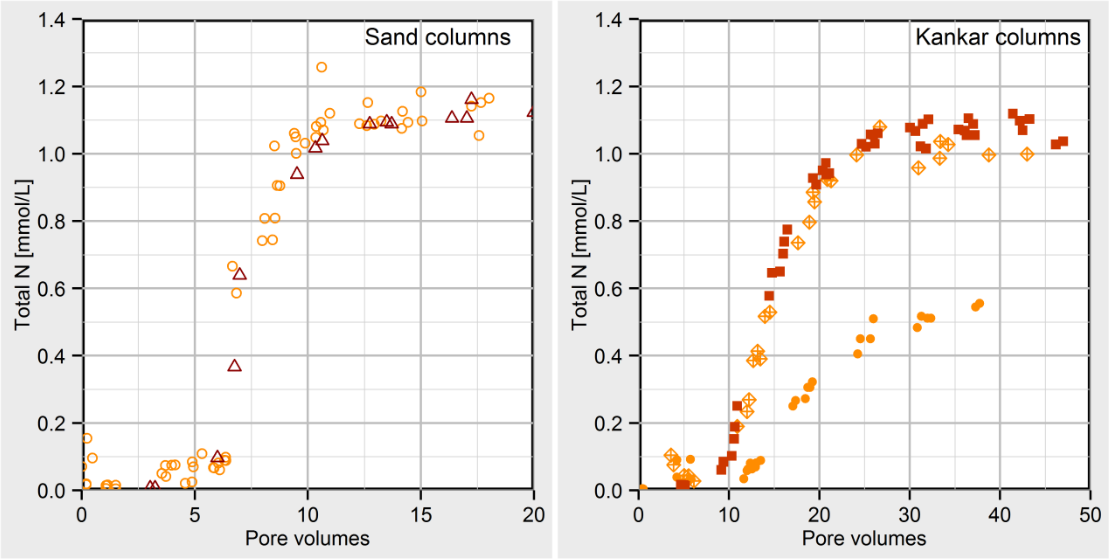

Figure 7 Ammonium breakthrough curves in sand and kankar samples. (Modified after Groeschke et al. 2014)

2.5 1D transport modelling

With the aim to predict the future concentrations of ammonium at well P3, 1D reactive transport models (Haerens 2002b) were set up for the field site. Based on the results of the 1D reactive transport column models of Groeschke et al. (2014), two flow paths in representative aquifer sediments were modelled with PHREEQC v3 (Parkhurst and Appelo 2013; Fig. 8). One flow path comprises 500 m distance from the riverbank to Ranney well P3. These 500 m were set up as a column, which is divided into 139 cells with a cell length of 3.6 m each. The time step was set to 4 d, resulting in the average linear velocity of 0.9 m/d as determined by Sprenger (2011). Transport parameters (effective porosities, number of exchange sites, and selectivity coefficients for the cation exchange) were taken from Groeschke et al. (2014) without any further adjustments and are summarized in Table 2. Dispersivities were adjusted to the model length. Although dispersion is generally higher at field scale than at laboratory scale because of sediment inhomogeneities which are not present in laboratory columns (Gelhar et al. 1992), dispersivities for the field model were adjusted to represent the magnitude measured in the column experiments: the longitudinal dispersivity was set to 5 m (1/100 of the flow path) in the sand and to 50 m (1/10 of the flow path) in the gravel. Because the sediment is carbonatic (Eybing 2014) and most water samples at the field site are slightly oversaturated with calcite, calcite was included as an equilibrium phase in the model. To check for numerical errors, the models were also run with 278 cells (1.8 m cell lengths) and 2 d time steps and with 556 cells (0.9 m cell lengths) and 1 d time steps.

Table 2 Transport parameters used in the simplified 1D model

Parameter	Unit	Sand	Kankar
Effective Porosity (n_e)*	---	0.24	0.175
Number of exchange sites	meq/1L water	0.054	0.21
log_$k_{Na\backslash K}$	---	0.67	0.98
log_$k_{Na\backslash Ca}$	---	0.1	0.18
log_$k_{Na\backslash Mg}$	---	-0.28	-0.09
log_$k_{Na\backslash NH4}$	---	0.55	0.81

* Effective porosities are not explicitly included in PHREEQC models. They are incorporated through the number of exchange sites

To keep the model minimal and straightforward, the following assumptions and simplifications were applied:

- Source water composition (displacing solution) was kept constant, although in reality there is a seasonal variability in the river water due to monsoon – non monsoon compositions.
- Ammonium was decoupled from the nitrogen cycle, meaning it cannot be oxidized to nitrate in the model. This would be representative of anoxic conditions in the aquifer, which by no means must prevail after an improvement of source water quality.
- An average linear flow velocity of 0.9 m/d (Sprenger 2011) was assumed for both flow paths. It is very likely that flow velocities are much higher in the kankar layer, but real data for this layer are not available.

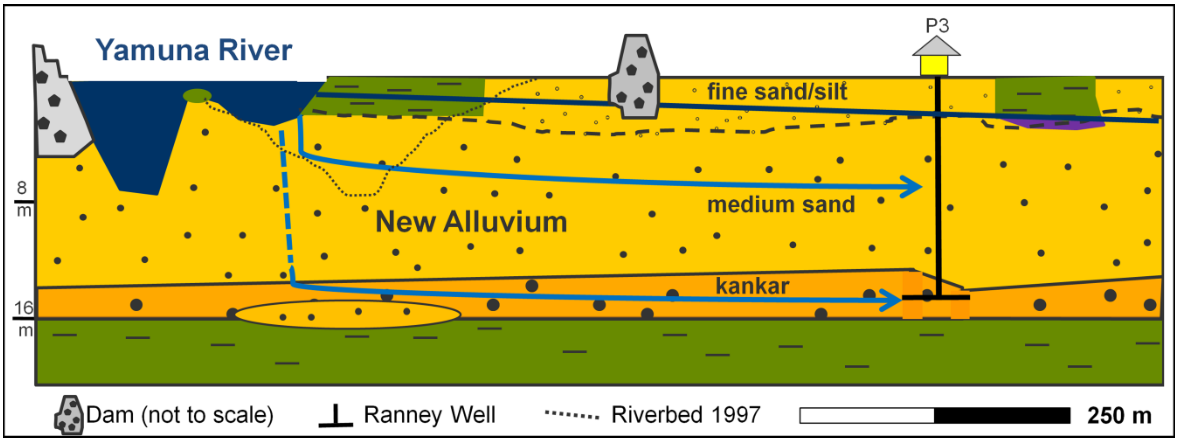

Figure 8 Flow paths from the river to well P3 in the 1D reactive transport models. The vertical flow from the river to the kankar layer was not considered and modelled

2.5.1 Adsorption modelling

To estimate the increase of ammonium concentrations at well P3, the infiltration of sewage-influenced river water into the aquifer was modelled. The cells were equilibrated with water samples taken at sampling points still uninfluenced by the ammonium plume. A sample taken from HatP4 in December was used for equilibrating the sand layer and a sample taken at P4 in December 2013 was used to equilibrate the gravel layer. The cells were then flushed with a displacing solution with the composition of a sewage-influenced river water sample taken at the field site in December 2012 with an ammonium concentration of 20 mg/L. The compositions of the water samples are summarized in Table 3. In the models, it took about 15 years to reach the 100% ammonium breakthrough in the sand layer and 62 years to reach the 100% ammonium breakthrough in the kankar layer (Fig. 9).

Table 3 Composition of the equilibrating and displacing solutions. Water samples were charge-balanced with C(4). Groundwater samples were taken in December 2013. River water sample was taken in December 2012.

Parameter	Unit	Equilibrating solution sand (HatP4)	Equilibrating solution gravel (P4)	Displacing solution (river water field site)
T	°C	26.2	26.4	20.5
pH	pH	7.58	7.4	7.6
E_h	mV	160	175	82
EC	µS/cm	495	893	1588
Na	mg/L	19.9	67.5	171
K	mg/L	5.4	6.8	15.4
Mg	mg/L	14	23	33.7
Ca	mg/L	63.7	80	65.4
Fe	mg/L	0.09	0.1	0.07
Mn	mg/L	0.09	0.3	0.3
HCO_3^-	mmol/L	5.2	5.9	6.5
Cl	mg/L	6	78	218
SO_4	mg/L	2	53	125
S^{2-}	mg/L	0	0	0
NH_4^+	mg/L	0	0.6	20
NO_2^-	mg/L	0.005	0.03	0.02
NO_3^-	mg/L	0	3.5	0

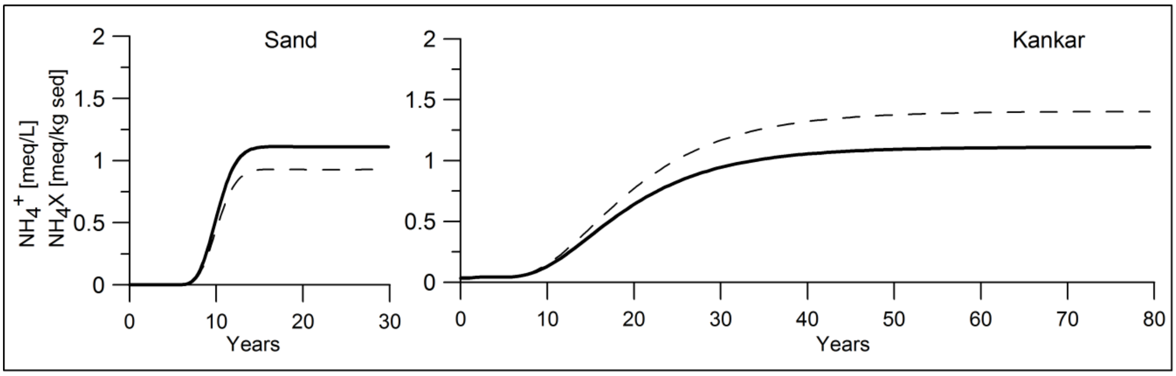

Figure 9 Results of adsorption modelling. Solid line: Ammonium concentration in water in meq/L, dashed line: Ammonium concentrations on the exchanger in meq/kg sediment

2.5.2 Desorption modelling

How long the ammonium contamination will prevail in the aquifer after source water quality improves depends not only on the sediment properties, but also on the extent of the ammonium plume. To model ammonium desorption, it was assumed that

- the ammonium plume has completely reached the well P3 and
- the ammonium distribution within the plume is homogenous at 35 mg/L NH_4^+ for the groundwater in the sand layer and 26 mg/L in the gravel layer.

The cells of the sand flow path were equilibrated with a water sample from hand pump B1 taken in December 2013 and the cells of the gravel flow path were equilibrated with water composition of sample H250 taken in December 2013 (Tab. 4). After equilibration, the column was flushed with a displacing solution with the composition of the river water upstream Delhi at Palla, where the Yamuna is still uninfluenced by sewage water. Ammonium concentrations were below the drinking water limit value of 0.5 mg/L after about 19 years in the sand layer and after about 61 years in the kankar layer (Fig. 10). Because degradation of ammonium was not implemented in the models, the results can only be seen as conservative estimates.

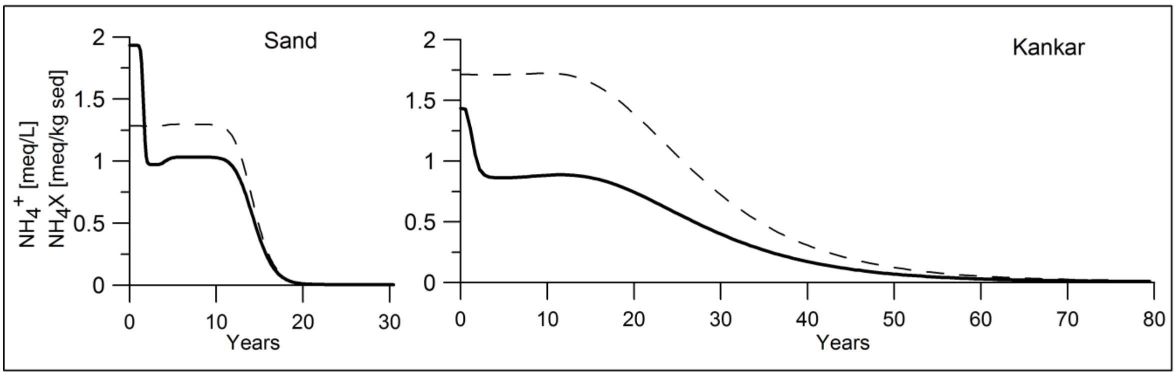

Figure 10 Results of desorption modelling. Solid line: Ammonium concentration in water in meq/L, dashed line: Ammonium concentrations on the exchanger in meq/kg sediment

Table 4 Composition of the equilibrating and displacing solutions. Water samples were charge-balanced with C(4). Groundwater samples were taken in December 2013. River water sample was taken in March 2007 in the frame of the TECHNEAU project.

Parameter	**Unit**	**Equilibrating solution sand (B1)**	**Equilibrating solution gravel (H250)**	**Displacing solution (river water upstream)**
T	°C	25.2	24.3	22.3
pH	pH	6.93	7.23	8.56
E_h	mV	105	84	268
EC	µS/cm	1615	1153	457
Na	mg/L	97	79.7	35
K	mg/L	17.3	13.2	9
Mg	mg/L	38.7	24.8	14
Ca	mg/L	126.5	89.1	44
Fe	mg/L	16.9	5.2	0.62
Mn	mg/L	0.42	0.27	0.05
HCO_3^-	mmol/L	11.9	8.3	2.7
Cl	mg/L	141	115	38
SO_4	mg/L	5	4	46
S^{2-}	mg/L	0.04	0	0
NH_4^+	mg/L	35	26	0.1
NO_2^-	mg/L	0.005	0.005	0.2
NO_3^-	mg/L	0	0.05	6

3 Remediation options

3.1 Source control

3.1.1 General improvement of river water quality

A sewage treatment capacity meeting the demand means that the entire sewage generated will be treated before being discharged into the river. Thus, the source water will meet the local guideline values for STP effluents. The resulting water quality will still strongly depend on the upstream river water quality and the dilution determined by the amount of STP effluents discharged into the river and the flow of the river, but it is almost certain that river water will become oxic. Annual average dissolved oxygen concentrations (mg O_2/L) in European rivers of all sizes show for example that normal oxygen concentration range between 9 to 11 mg/l for most part of the year (EEA 2001b).

The improvement of source water quality will, most likely, change the redox conditions in the aquifer, too. The extent is however depending on the organic content in the river bed sediments. If it is sufficiently low, a redox sequence from aerobic to nitrate-reducing to Fe/Mn-reducing conditions could develop. In areas with geogenic arsenic contaminations, this would have a positive side effect of fixing arsenic as it is less mobile under oxidizing conditions. A summary of the factors controlling arsenic mobility is given below in the grey box (p. 24). Depending on the prevailing redox conditions in the aquifer, the oxidation of ammonium to nitrate could occur.

Even after the ammonium source is removed, the ammonium contamination can prevail for decades, because flushing the aquifer with ammonium-free water will cause desorption of the ammonium from the sediment matrix. Hamann (2009) for example showed from a reactive transport model that in Berlin, Germany, ammonium contamination will prevail over 200 years after the deposition of sewage sludge on a sewage farm was stopped.

The advantages are:

- Achievement of good river water quality
- Decrease of arsenic levels in the bank filtrate (see grey box on arsenic demobilization p. 24)
- Decrease of other problems arising from heavily polluted surface waters like water borne diseases
- Decrease of odour pollution

The disadvantages are:

- Long-term measure: STPs need to be planned and built
- Long-term measure: Depending on sediment characteristics and extent of the ammonium plume, desorption of ammonium can last several decades
- Cost intensive (depending on the sewage treatment capacity needed)

Arsenic mobilization and demobilization

Arsenic can be found in numerous minerals. If those minerals are present in an aquifer, a change of conditions can trigger dissolution thus leading to elevated arsenic concentrations in the groundwater.

Arsenic is classified as a non-metal, and like other non-metals it usually forms oxyanions. Redox conditions and pH control the chemical form and speciation of arsenic. Oxyanions of arsenate As(V) and arsenite As(III) are the most common form of arsenic in the environment: Under oxidizing conditions, As(V) arsenate predominates in the form of $H_2AsO_4^-$ at pH < 7 and in the form of $HAsO_4^{2-}$ at pH > 7. Under reducing conditions, at a pH below 8, As(III) arsenite predominates in the form of non-ionic H_3AsO_3.

The mobility of arsenic is mainly controlled by sorption to iron, manganese and sulfur minerals. Especially As(V) has a strong binding to those, but also the non-ionic form of arsenite can be sorbed through Lewis acid - base interactions. Desorption from hydrous ferric oxide (HFO) can thus be a primary source in aerobic hydrous environments, when conditions change. Besides geochemical processes, biological reductions also play a role in arsenic chemistry in the subsoil.

According to Suthersan and Payne (2005), the three primary triggers that can lead to the release of geogenic arsenic are:

1. Increasing pH in an aerobic groundwater environment. HFO (hydrous ferric oxide) particles are amphoteric ion exchangers depending on the pH of the solution. Above pH 8.5, previously positively charged HFO particles become negatively charged and reject previously bound arsenate and arsenite.
2. Introduction of high concentration of competing anions. Arsenate and arsenite are stronger ligands (electron donors) than chloride and sulphate. Therefore, they are prefentially sorbed by HFO particles. However, phosphate, bicarbonate and silicates in high concentration can decrease arsenic sorption.
3. Development of reducing conditions. At reducing conditions, arsenate As(V) is reduced to arsenite As(III), which is less strongly sorbed. Furthermore, ferric iron Fe(III) is reduced to Fe(II), which goes into solution and releases the sorbed arsenic.

Overall, the biogeochemistry of arsenic and iron are strongly linked and Fe chemistry controls As contamination in shallow groundwater systems. Aerobic conditions reduce the mobility of arsenic as Fe(III) and Mn(IV) oxides remove arsenic from the dissolved phase. Under reducing conditions, sulfate reducing bacteria can remove arsenic by sequestering it in iron sulfides. This process can be triggered by adding degradable carbohydrates, ferrous iron salts and a sulfur source (Suthersan and Payne 2005).

The implementation of river water quality improvement will be complicated in Delhi because of the countless informal distributions into the drains connecting to the river and to the rapid population growth, which leads to constantly increasing sewage generation. According to the General Standards for Discharge of Environmental Pollutants

(Government of India 1986) STP effluents need to meet the guideline values of < 50 mg/L ammonical-N and < 10 mg/L nitrate-N before they are discharged into the river. Compared to the guideline value of 10 mg/L ammonical-N in other countries (Canada, Malaysia - Nova Scotia Department of Environment 2006, D.O.E Malaysia 2009) and 10 mg/L total nitrogen for > 100,000 population equivalent in the European Union (European Council 1991, European Commission 1998), the Indian guideline value for ammonical-N is high and is therefore not a useful tool to solve the problem of high nitrogen loads in surface waters.

In an oxic river, ammonium would not be the predominant nitrogen species as it would be oxidized. Nevertheless, high nitrogen concentrations would have to be expected in the river because the discharge currently is about 3,300 x 10^6 L per day (Anand et al. 2006) and is expected to increase, and the dilution with upstream river water is negligible for most part of the year, because Wazirabad barrage, which regulates the inflow in the North of Delhi, is open only in monsoon times.

The simplified 1D reactive transport models of a 500 m transect show that it can take up to about 60 years to flush out the ammonium that is adsorbed to the sediment matrix in this part of the aquifer. Even then nitrogen concentrations in the groundwater might not meet the drinking water guidelines as the models do not take into account additionally elevated nitrogen concentrations in the source water, which have to be expected because of the high STP effluent guideline values.

3.1.2 Aeration basins at the riverbank

To induce a similar effect as the general improvement of river water quality on a short term basis, large aeration basins could be constructed along the riverbank. River water could be then pumped or diverted into the basins where aeration would be achieved by pressing oxygen gas or air into the water, whereby the first is to prefer due to its higher efficiency. From these basins the aerated water could then either be infiltrated directly into the aquifer or it could be distributed back into the main stream to dilute the river water and increase water quality.

The advantages are:

- Short-term improvement of river water quality
- Low-tech
- Cost-efficient: possibility to treat only a part of the river

The disadvantages are:

- Nitrification is a biologically mediated process dependent on the occurrence of microorganism communities. Therefore, the system is complex and limiting factors are not only oxygen, but also pH, temperature and nutrient concentrations (Pollice et al. 2002**.** System instability is common (Wells et al. 2009)
- Incomplete nitrification can occur (Pollice et al. 2002) leading to elevated nitrite concentrations, which are toxic for aquatic species
- Total nitrogen levels remain high in the river: In STPs, a nitrification step is usually followed by a denitrification step to eliminate the nitrogen from the water. This can’t be achieved in aeration basins at the riverbank.

- Might not be suitable for rivers with highly variable flow volumes and/or frequently changing riverbed
- Difficult to achieve sufficient distribution of the injected oxygen in the water body
- Clogging of the oxygen diffusers could occur, if the water is highly polluted by sewage water

Given the Delhi-context, three of the above-named disadvantages need to be further discussed:

- The chemical and biological oxygen demand (COD and BOD) of sewage-influenced water bodies is very high. Thus, much oxygen is needed to significantly increase dissolved oxygen in the water, which leads to elevated costs of operation.
- As the Yamuna is a braided river system, the river bed is frequently shifting and thus the aeration basins would have to be constantly adjusted.
- The problem of high total nitrogen concentrations in the surface water is not solved by this method as the nitrification is not followed by a denitrification step. Furthermore, studies show that at temperatures above 30°C and pH >7, ammonium oxidisers grow faster than nitrite oxidisers (Hellinga et al. 1998), which would result in incomplete nitrification. In the Yamuna River in central Delhi, a pH between 7 and 8 is common and water temperatures up to 33°C were measured in the summer months.

Because the bacterial contamination in this part of the river is particularly high (average total coliform MPN /100ml: 25’943’333; max. total coliform MPN /100ml: 101’000’000 in 2002 at ITO bridge - Anand et al. 2006; total coliform MPN /100ml: 66’357’500 in 2013 at Okhla Barrage – Rani et al 2013), in addition, occupational health and safety measures would be of utmost importance during installation and maintenance of the facilities.

3.2 In-situ remediation

In the following section, an overview of in-situ remediation concepts and cases is given and the applicability in other settings, especially under the conditions met in India is discussed.

3.2.1 In-situ bioremediation through oxygen gas injection (Bio-Oxidation Wall - BIOXWAND)*

Example: In-situ bioremediation of an aquifer contaminated with ammonium, Berlin, Germany (Giese et al. 2003, Engelmann et al. 2004, Ehbrecht et al. 2004, Horner et al. 2009, BWB 2007, BWB 2012)

Groundwater within the subsurface drainage area of the Friedrichshagen waterworks (southeast of Berlin) is in part highly contaminated with ammonium. The source of the ammonium is the former disposal of domestic and industrial sewage on sewage farms and drainage from unsealed sludge drying places of the Münchehofe sewage treatment plant. The contaminated aquifer has a volume of about 200 million m^3 (1.5 km length, 3 km width and 45 m depth). The ammonium concentration in the groundwater spans a range of 10-

*The BIOXWAND method is a process patented by the Berlin Water Company (Berliner Wasserbetriebe).

90 mg/l NH_4^+-N. In order to protect the downstream drinking water production wells, an in-situ remediation concept was developed on the basis of oxygen injection into the aquifer. In 2007, a 100 m long pilot site was installed. After a successful test phase, it was upgraded in two steps in 2010 and 2012 and has now a length of 800 m.

Technical oxygen and air is pressed 20 and 36 meters deep into the normally oxygen-free subsoil through oxygen gas injection wells every 15 m. The oxygen gas is sparged into the aquifer to induce a fine dispersed, high surface residual trapped oxygen gas phase in the groundwater, the so called *bubble wall* zone. This zone is able to supply sufficient oxygen to the passing groundwater flow for microbial oxidative degradation of ammonium. The bubble wall is continuously or periodically reloaded with oxygen gas, when dissolved oxygen level in the sparging zone is falling. The aerobic zone is followed by an anaerobic zone further downstream, where denitrification takes place.

Secondary processes may include the autotrophic and heterotrophic denitrification reaction involving organic carbon and pyrite (FeS_2) (if present in sediment), ion exchange reactions and dissolution of calcite. Due to these secondary reactions, hardness increases in the groundwater and some acidification of the groundwater takes place. However, monitoring results at the BIOXWAND test site suggested that the acidification is limited by pH buffering by calcite dissolution. Thus, the sulphate release causes only a moderate drop in the groundwater pH by about 0.3 pH units (from 7 to 6.7; Horner et al 2009).

The effect of pyrite is limited to the initial phase due to the high reaction rate of pyrites. After complete pyrite consumption by oxidation and denitrification reactions, a successive dilution to pristine sulphate concentrations was demonstrated by reactive transport modelling. This prognosis is confirmed by the long-term monitoring performed by the Berlin Water Company (Horner et al. 2009).

For biotransformation processes, an effective nitrification limitation by a maximum dissolved oxygen level (50 mg/L) was not 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 to 50 days. Stable nitrification was achieved at $pH < 6.95$ and $Eh > 350mV$ (Giese et al. 2003).

After eliminating the contamination source at the sewage field, the contamination plume itself is expected to be flushed out without any attenuation remediation measure after about 80 years (Ehbrecht et al. 2004). Therefore, the oxygen bubble zone has to be managed at least over this time span to provide security for the production wells (Horner et al. 2009). Air sparging is potentially effective in homogeneous, highly permeable aquifers and with compounds that are easy to volatilize (Beckmann 2006).

The advantages are:

- Maintenance and operation is low-tech
- Maintenance is cost-efficient as it only involves injection of oxygen
- Constant temperature of the aquifer as a reactor helps in the nitrification process (Giese et al. 2003)

The disadvantages are:

- Oxygen is also consumed by the oxidation of reduced iron and manganese and organic material present in the aquifer.

- An increase in the concentration of sulphate (varying around 300mg/l) and hardness (varying around 600 mg/l) in the water has been reported (Horner et al. 2009).

In general, this method is a reasonably simple in-situ remediation measure as it only involves technical oxygen injection and no further chemicals or above-ground treatment steps. On the other hand, appropriate maintenance of the oxygen injection wells is of great importance in order to preserve the efficiency of this long-term measure. Technically, a special flood-proof injection well design might be necessary as flood events can occur at the Delhi field site.

With regard to groundwater chemistry, which is well studied at the Delhi study site (Lorenzen et al. 2010, Lorenzen 2011, Sprenger 2011, Groeschke 2013, Sprenger and Lorenzen 2014), an implementation seems possible. Because of the carbonate contents of the sediments, a calcite buffering system is expected to develop in the aquifer and thus a significant decrease of the pH is not to be expected at the field site. The total time required for nitrification at 10°C (the average groundwater temperature in temperate climate) is 30-40 days which is three times of that required at room temperature (7-10 days; Wise et al. 2000). Therefore, the high groundwater temperatures around 25 °C at the field site would be an advantage for this method as it results in a fast nitrification process. Only the high iron concentrations (up to 19 mg/L) in the water might decrease the efficiency by clogging, which results in a decrease of permeability in the aerobic zone.

Figure 11 shows a schematic set-up of a potential BIOXWAND at the study site based on the Berlin case data and the available information on the Delhi well field. In order to calculate the size of a potential BIOXWAND implementation, it would be essential to fill the knowledge gaps about the groundwater flow regime and to set up a groundwater flow model.

It has also to be noted that water treatment plants in Delhi are not designed to treat hard water, since the major source of raw water is surface water from rivers, which is relatively soft. The implementation at the Delhi study site would thus further require a post treatment operation for increased concentrations of HCO_3^- at the water treatment plants resulting in an increase in cost at a later stage. Mixing of the BIOXWAND-treated groundwater with raw surface water before the water treatment plant inlet may lead to sufficient dilution of the parameters sulphate and hardness, thus ruling out the need of new post treatment strategies.

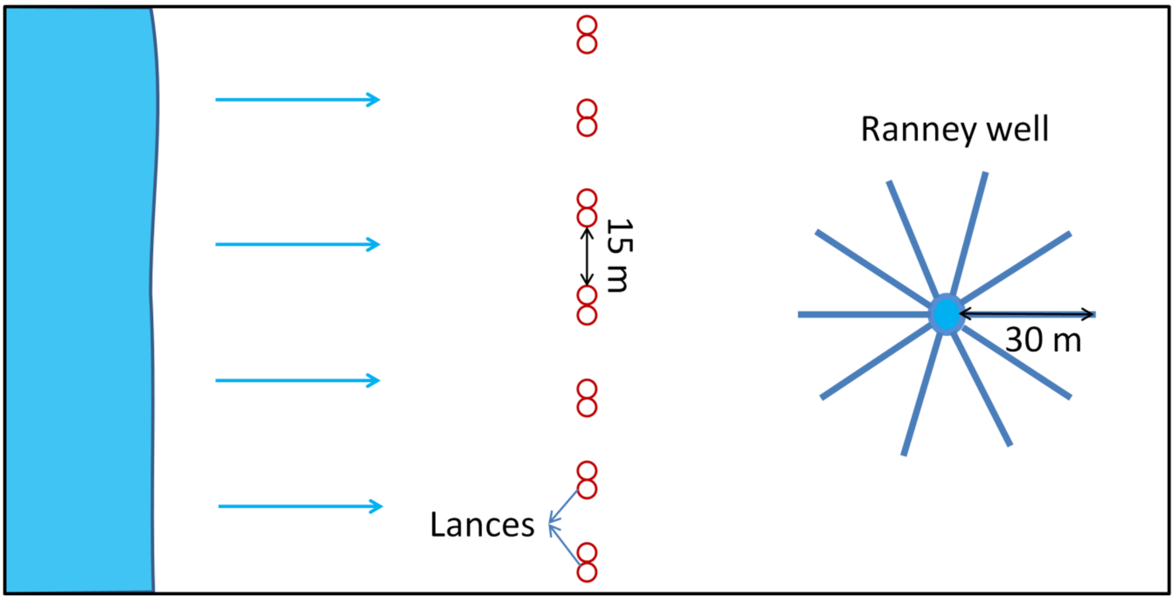

Figure 11Schematic set-up of a possible BIOXWAND as it could look at the field site. Map is not to scale

3.2.2 Sequential reactive barrier remediation using polymer mats (funnel and gate principle)

Example: Sequential reactive barrier remediation of ammonium-contaminated groundwater using polymer mats, Perth, Australia (Patterson et al. 2002, Davidson 2003, Patterson et al. 2004)

A pilot-scale field trial was carried out within an ammonium plume near Perth, Western Australia on the Swan Coastal plain. The site is located 30 m of the shoreline of Cockburn Sound, where leaks of ammonium products from a fertilizer factory about 300 m upgradient of the site caused an ammonium plume in the groundwater. Peak concentrations of 110 mg/L total N were found in about 6 m below ground level. A series of successful large-scale column experiments were performed before establishing the pilot field trial (Patterson et al. 2004).

Permeable reactive barriers (PRBs) are a widely used in-situ method for the treatment of various groundwater contaminations. The concept of PRBs is to place a permeable reactive material in the subsurface across the flow path of contaminated groundwater. As the groundwater moves through the material due to the natural hydraulic gradient, the contaminants are immobilized or transformed to less harmful species (Thiruvenkatachari et al. 2008, Obiri-Nyarko et al. 2014).

For the in-situ treatment of the above-named ammonium plume, a sequential barrier technique using nitrification and denitrification processes was established by installing a sequence of different polymer mats. In this sequence, the first, up-gradient polymer mat was used to deliver oxygen via diffusion. As the groundwater moves past, bacterial nitrification of the ammonium to nitrite and nitrate is induced. The second, down-gradient polymer mat was used to deliver ethanol to induce bacterial denitrification of the nitrate to nitrogen gas. The oxygen and the ethanol delivery polymer mats were installed in a

0.75 m wide and 1 m long flow-through box. 10 m wide impermeable wings were constructed on either side of the reactive zone in order to drive the contaminated groundwater to the flow-through box (funnel and gate principle).

In large-scale column experiments with the polymer mats, ammonium concentration decreased from approximately 120 mg/L to less than 10 mg/L over two weeks. Groundwater oxygen concentration increased from 0.05 to 23 mg/l and then decreased to around 1 mg/L within one week and remained at this level throughout the experiment (Patterson et al. 2002). In the following pilot scale field trial, a 7 m deep treatment wall was constructed. The wall did not reach down to the aquitard, making ammonium removal less efficient with depth as preferential flow paths below the wall could develop near the edges of the wall. Ammonium feed concentrations were an order of magnitude lower than in the column experiments (11-18 mg/L). Nitrification was not only observed on the polymer mat-groundwater interface but also in the groundwater downstream of the polymer mat. This may have been due to the inconsistent groundwater flow (tidal effects) at the field site. A decrease in total N of about 90% was monitored for the field experiments (Patterson et al. 2004).

The advantages are:

- 90% reduction in total N was observed in field experiments
- Provision of in-situ remediation strategies over a short time frame or groundwater flow distance due to the sequential technique
- Use of the natural hydraulic gradient within the groundwater flow
- Relative cost-effective method compared to long-term above-ground treatment systems.

The disadvantages are:

- Measure changes the local groundwater flow regimes.
- Hydraulic conditions have to be well known before installation of PRBs to ensure a groundwater flow through the reactors. A preferential flow around the barrier or below it might develop
- It is important to understand, whether competing chemicals exist in the aquifer

The sequential reactive barriers could be a remediation option for the Delhi site as it seems to be a relatively simple and cost-effective measure. It is thinkable to construct a shield around the existing Ranney wells with particularly high ammonium concentrations (Fig. 12). As the aquifer is not very deep, it might even be possible to prevent undesirable flow below the barrier by constructing it over the full depth of the aquifer down to the clay aquitard.

On the other hand, the polymer mats should not be installed in the drawdown zone of production wells because they seem to be sensitive to changes in redox conditions. This could be a difficulty at the Delhi site because of the existing private wells causing uncontrollable drawdown zones.

Due to limited field scale studies available for this technique of ammonium remediation, the application should be accompanied by a research program for further evaluation of the applicability with its relative advantages and disadvantages.

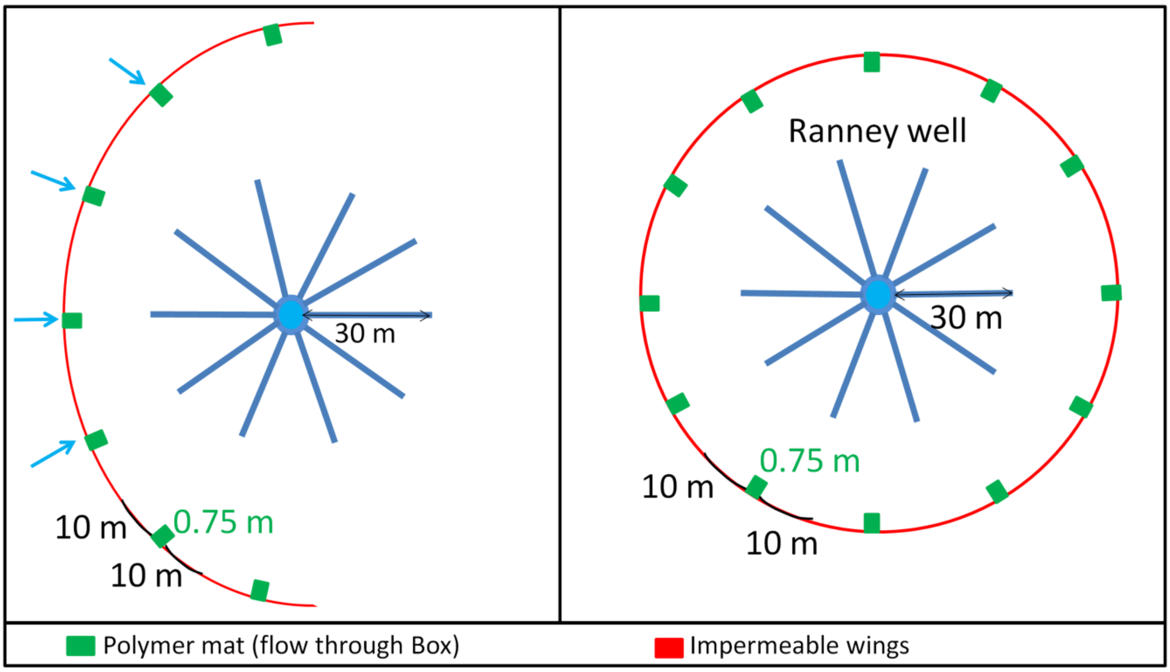

Figure 12Possible set-up of polymer mats at the field site (schematic). Figure is not to scale

3.2.3 Groundwater circulation wells and virtual-permeable reactive barrier*

Example: In-situ remediation of ammonium- and nitrate-impacted groundwater, London Basin, UK (IEG Technology 2008)

No studies or reports about this technology were found. All information is taken from IEG Technology's descriptions of the process. No details were given.

In the chalk aquifer of the London Basin, UK, high concentrations of ammonium were observed due to infiltration of sewage water. On the basis of estimates, an ammonium plume with a width of 105 m and ammonium levels of 160 mg/L was reported. The target ammonium concentration was set to 0.5 mg/L (IEG Technology 2008).

A Groundwater Circulation Well (GCW) induces a circulating flow field that carries clean water and sometimes additives needed for in-situ treatment (like oxygen or nutrients) through the contaminated regions of the aquifer (Borden and Cherry 2000). Two setups have been identified:

Groundwater circulation wells combined with ammonium stripping

To remove most of the ammonium from the aquifer, a GCW was combined with ammonium stripping. This was achieved by a two-step process involving the conversion of ammonium to ammonia by increasing the pH to 9-10 followed by the removal of ammonia by negative pressure air stripping (Fig. 13). The stripped ammonium was recovered ex-situ using a wet scrubbing system. Bioremediation was further accelerated by the addition of nutrient-enhanced reactive zero-valent iron to the groundwater. With this method, ammonium concentrations of 20 mg/L were achieved (IEG Technology 2008).

*The IEG GCW Method is a process patented by IEG

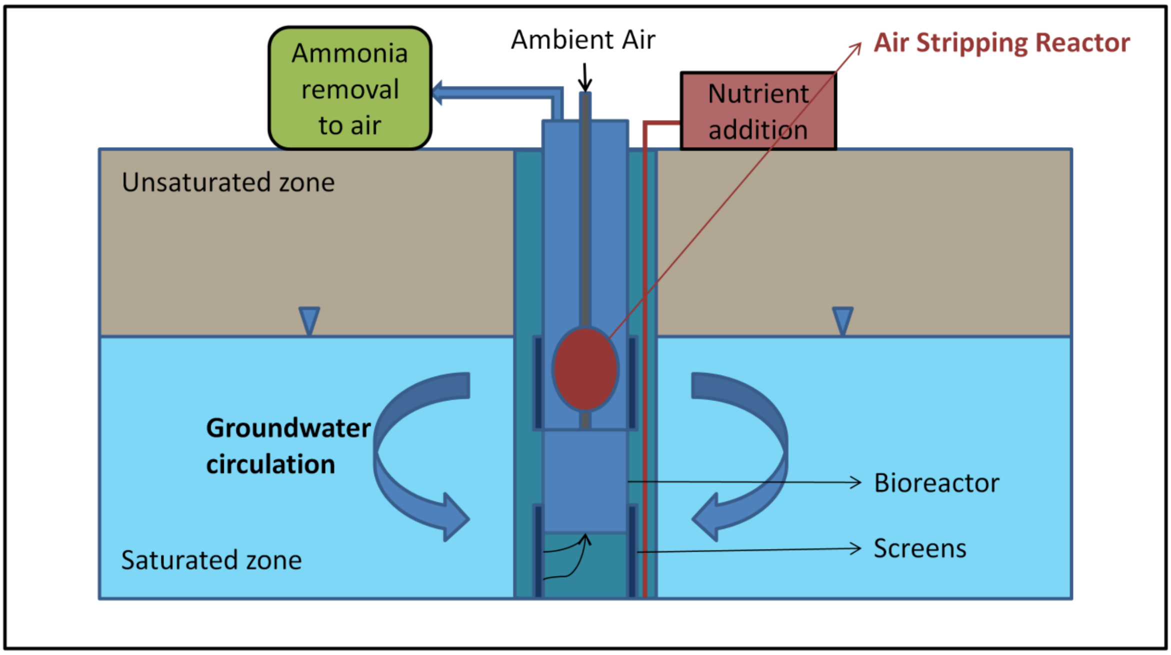

Figure 13Schematic overview of a groundwater circulation well in combination with ammonia air stripping. Modified after IEG Technology 2008

Virtual-permeable reactive barrier

In order to further decrease the ammonium concentrations, a Virtual Permeable Reactive (VPR) barrier consisting of a number of GCWs was installed in two lines perpendicular to the groundwater flow direction. The GCWs in the first line induce an aerobic zone with the nitrification of ammonium to nitrate. The second, down-gradient line of GCWs induces an anaerobic zone for the denitrification of nitrate to nitrogen gas (IEG Technology 2008).

The advantages are:

- Operation with minimal maintenance over an extended period of time
- GCW systems are relatively small and potentially have less negative impact on land use compared with pump and treat methods (Elmore and Graff 2002).
- Groundwater is enriched with oxygen and the circulation increases the potential for natural aerobic degradation processes in the aquifer.
- Possibility of simultaneous treatment of the unsaturated zone and capillary fringe by vapor extraction (IEG Technology 2006, 2011)
- GCWs can be combined with other technologies such as bioremediation, bioventing, soil vapor extraction, surfactant, zero-valent dehalogenation, and oxidation (OST 2002).

The disadvantages are:

- Anisotropy of the target aquifer must be within a range that allows the circulation cell to develop, generally between 3 and 10 (OST 2002).
- GCWs may have limited effectiveness in shallow aquifers because of the limited space for circulation (OST 2002).
- Addition of air can cause clogging in wells.

- If the system is not properly designed or constructed, the contaminant plume may spread beyond the radius of influence or the wells may become clogged (OST 2002).

In Delhi, ammonium concentrations at the field site range between 20 and 35 mg/L in the centre of the plume. Therefore, the air stripping of ammonia through negative pressure is probably not effective, as target concentrations here seem to lie at 20 mg/L. It seems that it would be sufficient to install the two rows of GCWs with the induced aerobic and anaerobic zones for nitrification-denitrification to take place (VPR barrier). However, this method is complex and installation costs could be high.

3.3 Pump and treat methods

In the following section, an overview of pump and treat remediation concepts and cases is given and the applicability in other settings, especially under the conditions met in India is discussed.

3.3.1 Ivey- sol aided SorbitAll filtration method*

Example: Remediation of ammonium-contaminated groundwater plume at a fertilizer facility, Western Canada (Ivey International 2012)

No studies or reports about this technology were found. All information is taken from Ivey International descriptions of the process. No details were given.

For the case study site in Western Canada, several accidental spills from a fertilizer facility were reported over a period of two to three years resulting in an increase of ammonium concentrations in the shallow silty-sand aquifer. The ammonium concentrations ranged between 20 to 700 mg/L and the plume extended over a distance of 2000 m. The remediation was completed within 18 months (Ivey International 2012).

This pump and treat method requires the installation of several 100 mm diameter injection wells, screened across the ammonium plume in an integrated network of 5-spot patterns with nearby extraction wells (Fig. 14). In order to selectively desorb ammonium from soil and fractured bedrock surfaces, the Ivey-sol surfactant comprising several nonionic surfactant formulations is injected through the injection wells. In the case study, this was done bi-weekly. The function of the surfactant is to lower the surface tension of water, improving both its wetting and permeability properties and thus making the desorbed contaminants hydraulically more available for extraction by pumping (Ivey International 2012a). The contaminated water is then extracted and treated ex-situ with the SorbitAll filtration system, an ion exchange technique using clinoptilolite zeolite (Ivey International 2012b). After treatment, the water is recharged through infiltration wells outside the plume boundary. Thus, a hydraulic barrier in the local groundwater table is created with the function to minimize spreading of the ammonium plume and to enhance the induced hydraulic gradient towards the extraction wells. A > 96% reduction in ammonium concentration was achieved at the example site (Ivey International 2012).

*IVEY sol is patented by IVEY International Inc.

The advantages are:

- Injection of Ivey-sol surfactant desorbs the ammonium from the aquifer matrix (Ivey International 2012, 2012a)
- Works well in fine grain soils (silty sand, silt, silty clay, clay) and fractured bedrock

The disadvantages are:

- Operation and maintenance are complex issues with the use of large numbers of pumps in the extraction wells
- Addition of surfactant to the groundwater

This method might not be the first choice for use at the Delhi field site or at similar sites in India. Not only is the installation expensive (installing a complex five spot pattern of injection and extraction wells, which also have to be flood proof at this site), but operational costs are high, too. The injection and extraction well pattern would cover a large area that is currently used for small-scale agriculture and it would be a great disadvantage for the local population, if land use would be changed.

In addition, it is not advised to inject chemical solutions into the groundwater, if a reliable, objective, and close-knit accompanying monitoring cannot be guaranteed. Even though the Ivey solution is said to be non-toxic, biodegradable, and does not persist in the environment after application, the use of those measures might be critical in drinking water protection zones.

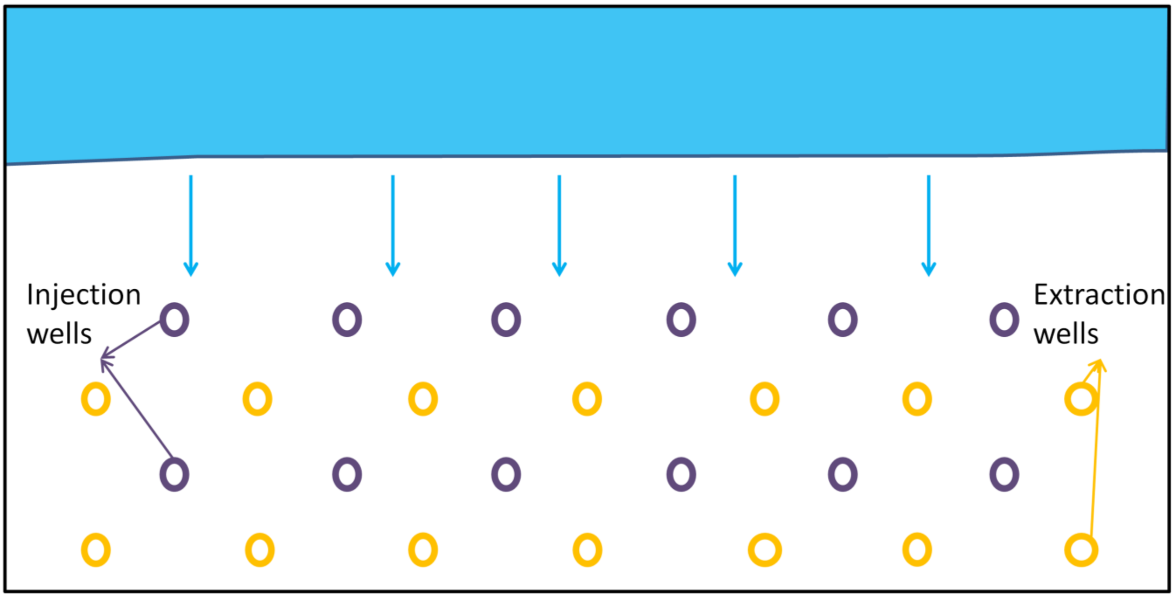

Figure 14Schematic set-up of 5-spot pattern

3.3.2 Nitrification and denitrification remediation*

Example: In-situ groundwater nitrification and de-nitrification remediation processes, Calgary, Canada (Mailath and Chu 2005, Mailath 2008)

In Calgary, close to the Bow River, elevated nitrate and ammonium concentrations were measured in an alluvial sand and gravel aquifer, which is overlain by 2-5 m thick silt and clay glacial deposits (Savage et al. 2006). The main nitrogen source was a former industrial site, which was shut down in 1992. Lanza (2009) reported peak concentrations of 38 mg/L NO_3-N and 75 mg/L NH_4-N for this site, but concentrations in the plume had already decreased for the past decade. Around the year 2000, an in-situ pilot treatment test was implemented to study the nitrification and denitrification remediation method (Mailath and Chu 2005).

Nitrification and denitrification remediation is a pump and treat remediation method only in a wider sense. The treatment itself takes place in the subsoil, but in order to trigger the in-situ reactions, water is pumped from the aquifer, mixed above-ground with certain additives and then re-injected. For a complete nitrification-denitrification treatment, two pairs of wells are needed, each consisting of an extraction and an injection well.

The nitrification step is achieved by extracting groundwater, adding oxygen and nutrients (typically, phosphate is used) and re-injecting it into the aquifer up-gradient from the extraction well. The oxic, nutrient-rich water encourages nitrifying bacteria to develop in the aquifer, who convert ammonia to nitrate. Ammonium is then oxidized in the reaction cell that develops between the injection well and the extraction well (Mailath 2008).

The nutrient is supplied to the groundwater by maintaining a phosphorus concentration of 0.2 - 0.8 mg/L, and the dissolved oxygen concentration is to be maintained above 1 mg/L. In environments where nutrients may already exist (naturally or having been introduced), there may be situations where little or no nutrient addition may be required (Mailath and Chu 2005).

To reduce nitrate concentrations, the same method is applied. In that case, carbon (instead of oxygen) and nutrients are added to the extracted water creating an anaerobic zone around and down-gradient the injection well, where denitrifying bacteria reduce nitrate to nitrogen gas (Mailath 2008). The mass flux of nitrate entering the reaction cell can be modelled to determine the stoichiometric carbon equivalent required to reduce the nitrate, with the groundwater being re-circulated between the extraction wells and the injection wells (Mailath and Chu 2005).

It is important to calculate the reaction cell of the nitrification process in order to install the extraction well at the right distance to the injection well. The reaction cell size increases with increasing hydraulic conductivity and decreasing groundwater gradient. In case that it is possible to add an oxygen-releasing compound as a source for oxygen (e.g. bleaching powder), enough oxygen is added to the system to oxidize all the ammonium in the zone around the well, and no circulation cell is necessary and the extraction well does not have to be installed down-gradient of the injection well (Mailath and Chu 2005).

During in-situ pilot scale testing in the alluvial aquifer named above, ammonium concentrations were reduced from 58 mg-N/L to 6.0 mg-N/L in approximately 120 days and nitrate concentrations were reduced from 66 mg-N/L to 0.2 mg-N/L in approximately 14 days (Mailath 2008).

*This method of Nitrification and De-Nitrification is patented by Mailath and Chu

The advantages are:

- Method is especially applicable under site conditions where the hydraulic conductivity is relatively high, such as in sand and sandy gravel aquifers.
- In soils with relatively high hydraulic conductivities, the reaction cell size of the injection wells can be maximized, and the number of injection wells can be minimized resulting in lower cost systems (Mailath and Chu 2005).
- Method can be well applied in aquifers contaminated with ammonium and nitrate.

The disadvantages are:

- Low hydraulic conductively sediments and bedrock cannot accommodate injection of large volumes of water.
- Four drillings would be required in one line parallel to the groundwater flow direction to complete both, the nitrification and denitrification process.
- High maintenance costs (energy costs for operating pumps, costs of nutrients, oxygen)
- Requires space (two houses/installations for the pumps and the nutrient mixing facilities)

As is common for pump and treat methods, this method requires high maintenance and involves high maintenance costs. It seems unpractical to extract groundwater and re-inject it (nitrification step), extract and re-inject it again for the denitrification step, and then to extract it 50 m further down gradient in a drinking water production well.

For the Delhi site, the installation of one line of four wells would probably not be sufficient to treat all the water in the catchment area of one well. Furthermore, on-site installations (treatment sheds) are required for the pumps and the nutrients mixing facilities.

Table 5 Overview of remediation options for aquifers contaminated with ammonium

Method	Principle	Advantages	Disadvantages	Use in Delhi
Source Control				
Improvement of source water quality	Stopping discharge of untreated sewage into rivers would allow for infiltration of aerated, NH_4^+-free water into the aquifer.	-Might decrease arsenic concentrations in bank filtrate -Decrease in odour pollution, improvement of environment	-Long-term solution: STPs need to be planned and built - NH_4^+ contamination will prevail decades after river quality improves	-Likely to change redox conditions in aquifer (arsenic demobilization) - Indian guideline limits for STP effluents[1] permit high nitrogen discharge into rivers
Aeration basins	Construction of aeration basins along riverbanks would lead to infiltration of NH_4^+-free water.	-Short- term improvement of river water quality -Low-tech	- Sufficient distribution of O_2 is difficult -Might not be suitable for variable flow -Clogging of diffusers might occur	-High COD and BOD in Yamuna in Delhi -Shifting riverbed: adjustment of facilities -Bacterial contamination: safety measures
In-Situ				
Oxygen injection - Bioxwand* (BWB 2007, Horner et al. 2009)	In-situ bioremediation through oxygen gas injections into the aquifer using lances.	-No addition of further chemicals. Suitable for drinking water protection zones.	-O_2 consumed by oxidation of Fe^{2+}, Mn^{2+} and organic material present in the aquifer. -Increase in SO_4 and hardness	-High GW temperatures: fast nitrification process -Post treatment for increased hardness of water at the WTPs might be necessary
Sequential permeable reactive barrier using polymer mats (Patterson et al. 2002, Patterson et al. 2004)	Use of in-situ polymer mats. Up-gradient mat delivers O_2 to induce nitrification as GW moves past. Down-gradient mat delivers ethanol to induce denitrification.	-90% reduction in total N in field experiments -In-situ remediation over short time frame or GW flow distance. - Cost effective	-Measure changes local groundwater flow regimes. -Competing chemicals might exist in the aquifer. -Limited studies available.	-Thinkable to construct shield around wells with high NH_4^+ concentrations -Mats sensitive to fluctuating GW tables (private wells cause drawdown zones)
Groundwater circulation wells (GCW) and virtually permeable reactive barrier* (IEG Technology 2006, 2008, 2011)	GCWs induce circulating flow. Increasing pH in flow cell converts NH_4^+ to NH_3; removal by negative pressure air stripping. Down gradient further GCWs induce aerobic and anaerobic flow cells.	- Less negative impact on land use compared with pump and treat methods[2] -Simultaneous treatment of unsaturated zone and capillary fringe by vapor extraction	-Anisotropy of aquifer must be within range that allows circulation cells to develop[3] -Limited effectiveness in shallow aquifers[3] -Wells may become clogged[3].	-20 mg/L is target concentration for the negative pressure air stripping. Peak NH_4^+ concentrations in plume in Delhi are 20-35 mg/L: Sufficient to install the VPR barrier only. This requires the installations of numerous GCWs.
Pump and Treat				
Ivey-sol aided SorbitAll filtration method* (Ivey International 2012, 2012a)	Installation of injection and abstraction wells across plume. Injection of Ivey-sol surfactant to desorb NH_4^+ from soil. Water is abstracted and treated above ground using ion exchange. Treated water is re-injected.	->96% reduction in the dissolved NH_4^+ at a study site. -Addition of Ivey-sol enhances remediation by desorbing ammonium from sediment -Works well in fine-grained soils	-High operation and maintenance costs -Complex operation and maintenance -Injection of additives into the aquifer	-Land use would have to be changed in order to install 5-spot-pattern of wells across the ammonium plume. - Injection of chemicals problematic in drinking water protection zone
Nitrification and denitrification remediation* (Mailath & Chu 2005)	To trigger in-situ nitrification – denitrification reactions, GW is extracted, mixed with O_2 + nutrients or carbon + nutrients and re-injected. Separate injection and abstraction wells needed for nitrification and denitrification.	- Especially applicable in sand and sandy gravel aquifers -In aquifers with high hydraulic conductivities reaction cell size of injection wells can be maximized and number of injection wells minimized	- Not suitable for low hydraulic conductivities -Four drillings required in one line parallel to GW flow to complete nitrification and denitrification -High maintenance costs and space requirement	-Method is costly and requires much space and thus might conflict with agricultural land use -Method would involve two extraction-injection cycles

* Patented technology [1]Government of India 1986 [2]Elmore & Graff 2002 [3]OST 2002

4 Post treatment options (methods of ammonium removal in raw water)

In the following section, an overview of post treatment options for raw water with elevated ammonium concentrations is given and the applicability in other settings, especially under the conditions met in India is discussed.

4.1 Physico – chemical nitrogen removal

Physico-chemical processes provide a range of methods for the removal of ammonium in drinking water (Health Canada 2013) as well as in wastewater (EPA 1997), although in wastewater treatment most of them have not been widely used because of higher costs, poorer effluent quality and greater sludge volumes compared to biological treatments (EPA 1997). Nevertheless, their applicability in the drinking water context is evaluated.

4.1.1 Air stripping of ammonia

Usually, this method is used in wastewater treatment (Health Canada 2013). Ammonia converts gradually to the gaseous state, if the pH of water increases above a pH of 7 (Huang and Shang 2006). This principle is used for the removal of ammonia via air stripping. The pH of water is increased to about 11 by adding lime or caustic soda. As the pH increases, a greater proportion of ammonium converts from NH_4^+ to NH_3 (Gauntlett 1980). When the liquid is then brought into contact with air in a stripping tower, a gradient exists across the gas/liquid interface and ammonia will be stripped to the air (Huang and Shang 2006). The ammonia in the off-gas can be removed by:

- passing the gas through a filter for biological removal or
- passing it through a scrubber to re-dissolve the ammonia in water, which can be treated separately to the main process stream (EPA 1997).

The advantages are:

- Cost-effective method: Once installed, the system should work for a long time (Huang and Shang 2006)
- The increase in pH to the level of 10-11 yields additional benefits such as the killing of some pathogens and micro-organisms (Jones et al. 2005)
- Widely applied for ammonia removal in the last years, thus sufficient information is available (Huang and Shang 2006)

The disadvantages are:

- Formation of lime scale on the packing materials of air stripping units is a major problem requiring periodical cleaning (EPA 1997).
- Re-carbonation or some other neutralization technique would have to be adopted to make the water suitable for drinking purposes since there is an excessive increase in the pH (Jones et al. 2005). Studies show that re-carbonation is a cost-effective way for maintaining the pH compared to other techniques such as the addition of sulphuric acid (Al-Mutaz and Al-Ghunaimi 2001).

- A volumetric air:water ratio of about 3000:1 is required to achieve effective NH_3 removal (Gauntlett 1980) leading to a large space requirement for the stripping towers.

In all, this method could be recommended for use in water treatment plants in India having ammonium concentration greater than 5 mg/L in the raw water since the process is too expensive for lower ammonium levels. This method seems to be an option discussed in Delhi: “Iron and ammonia is observed in Ranney well waters going to Okhla WTP in Delhi requiring establishment of iron removal plant and ammonia stripping plant” (Government of Delhi 2006 p.8-7).

The climate in India is beneficial for this method as a temperature increase of 10°C increases the efficiency of this method at a particular pH by 30%. 95% ammonia removal is achieved at pH 10 and a temperature of 40°C (Huang and Shang 2006), and temperatures around 40°C are normal during the summer months in India.

On the other hand, it is important to reduce the pH before distributing the water into the network, and to monitor the WTP effluent. Furthermore, the influent water quality has to be checked because other than the target substances may reduce the effectiveness of treatment (Huang and Shang 2006). In addition, effective handling of ammonia gas is recommended though studies show that there is not much release of ammonia gas into the air.

4.1.2 Ion exchange

This method involves the process of displacing ions of one species from an insoluble solid phase by ions of another species in solution. Natural zeolites are used as ion exchange material for ion removal from polluted water (Metcalf & Eddy Inc 2014). The zeolite *Clinoptilolite*, for example, is selective for the ammonium ion in preference to other ions occurring in the water (EPA 1997). The principle is that water is passed through a bed of zeolites. 86-99% ammonium removal can be achieved, depending on concentration and water composition (Health Canada 2013). Removal capacity of ammonium by Clinoptilolite was observed to be 9 mg/g with a 10 mg-N/L solution with no other cations present in the solution (Boulinguiez 2005). The method proved to be successful for up to 200 mg/L NH_4^+ in the inlet water (Weatherley and Miladinovic 2004).

The advantages are:

- Cost-effective method
- Clinoptilolite can be easily and economically re-generated by the use of NaCl (Abd El-Hady et al. 2001) or by biological regeneration (Rahmani and Mahvi 2006)
- Efficiency of ion exchange increases by 20-40%, if the zeolite is pre-heated (Abd El-Hady et al. 2001)
- The ion exchange process has no sensitivity to fluctuation in NH_4^+ influent concentration (Rahmani and Mahvi 2006)

The disadvantages are:

- Potential increase in corrosivity of the treated water (Health Canada 2013 after Schock and Lytle 2010)

- Inconvenient for WTPs with a capacity > 18 MGD because of the space required for ion exchange columns (Kurama et al. 2002)
- Mineral imbalance to be checked after post treatment to ensure meeting the drinking water standards (Health Canada 2013)
- Presence of Ca reduces NH_4^+ adsorption onto the zeolite (Weatherley and Miladinovic 2004)
- High input concentrations require large volumes of zeolites (Li et al. 2011)

WTPs in Delhi have a capacity > 40 MGD (Government of Delhi 2011), therefore, this method is not suitable to retrofit into existing WTPs. It could be used for treating NH_4^+ contaminated GW from Ranney/tube wells by clustering a few wells and installing a small treatment facility for the NH_4^+ treatment prior to conventional treatment. Furthermore, small cities facing an ammonium problem can efficiently use this technique.

4.1.3 Breakpoint chlorination

Breakpoint chlorination refers to the process at which sufficient chloride is added to react with all oxidizable compounds in the solution until additional chloride will occur as free chlorine (Metcalf & Eddy Inc 2014). Typically, chlorine (hypochlorous acid HOCl), a highly active oxidizing substance, is used. When added to water containing nitrogen in form of ammonia or ammonium, the following stepwise reaction occurs (EPA 1997):

$$NH_4^+ + HOCl \rightarrow NH_2Cl + H_2O + H^+$$

$$NH_2Cl + HOCl \rightarrow NHCl_2 + H_2O$$

$$NHCl_2 + HOCl \rightarrow NCl_3 + H_2O$$

Temperature, pH, contact time and the chlorine:ammonia ratio are factors influencing these reactions (Metcalf & Eddy Inc 2014). After the formation of monochloramine (NH_2Cl) the process proceeds by forming dichloramine ($NHCl_2$) and then by decomposing $NHCl_2$. Trichloramine (NCl_3) is formed as an intermediate product throughout the entire decomposition of the chloramines (Health Canada 2013). The deconstruction of the chloramines takes place by oxidizing them to nitrous oxide (N_2O) and nitrogen (N_2) and reducing the chlorine to chloride ion (Metcalf & Eddy Inc 2014).

Theoretically, a stoichiometric ratio of Cl_2:NH_3-N = 7.6:1 will achieve a 95-99% conversion to N_2. Experiments by Takó and Laky (2012) show that a 9-11:1 Cl_2:NH_4-N dose ratio is required in the presence of Fe, Mn and organic matter. Subsequent dechlorination of the water stream may be required (EPA 1997), as the method has the disadvantage of increasing the chloride content of the treated water (Goodall 1979).

The advantages are:

- The methodology is adopted in many current water treatment plants
- The monochloramine and dichloramine formed act as a potential disinfectant (Donnermair and Blatchely III 2003)
- Combination with As removal is possible (Takò and Laky 2012)

The disadvantages are:

- The formed chloramine gives an unpleasant odour to the treated water (Takó and Laky 2012)
- The chlorine in the water reacts with organic material resulting in the formation of undesirable by-products like tri-halo methane (THM) and adsorbable organic halides (AOX). These components are known to be cancerogenic and mutagenic substances (Takó and Laky 2012). As a countermeasure, activated carbon adsorbers have to be installed making the treatment expensive (Janda and Rudovský 1994)
- In order to guarantee that the breakpoint chlorination is always achieved, a tight monitoring of NH_4^+ concentrations as wells as of the several forms of chlorine has to be conducted (Health Canada 2013)
- High ammonia-N:chlorine ratios are required (Griffin & Chamberlin, 1941)
- Method is best suitable for $NH_4^+ << 1mg/L$ (Gauntlett 1980)

Because this method is a disinfection method, the technology is used in many WTPs in India, but with focus on hygienic water parameters. For the effective removal of ammonium a close monitoring and determination of the breakpoint is required (Health Canada 2013). Because of the frequent variations in raw water quality, chlorine dosage has to be constantly adjusted to achieve breakpoint chlorination. In addition, treatment has to be followed by carbon adsorbers if organic compounds are present in raw water, which is generally the case in Indian surface water bodies. This makes the process relatively expensive.

The use of chlorine for ammonium removal can further only be recommended for water sources with less than 1 mg/L of ammonium (Gauntlett 1980). In Delhi it might be difficult to always reach treatable concentrations in the raw water even when mixing ammonium-contaminated groundwater with surface water before treatment. Although the ammonium concentration in the surface water of the Yamuna River at the intake of Wazirabad water works is 0.39 mg/L on average, concentrations of up to 3.55 mg/L have been measured at that location (CPCB 2006). Mixing groundwater with surface water is, therefore, not always sufficient to reduce the high ammonium concentrations in the raw water to treatable limits. Thus, the method of breakpoint chlorination could only be used in India, if the surface water had low and stable ammonium levels. The current problems with breakpoint chlorination of Yamuna river water are described by Kumar (2013).

4.1.4 Reverse osmosis

Reverse osmosis (RO) is a membrane filtration technology which is mainly used for desalination processes (Metcalf & Eddy Inc 2014), but is also applied in water treatment systems (Radjenović et al. 2008). To induce reverse osmosis, a pressure higher than the osmotic pressure occurring between the two differently concentrated solutions has to be imposed across the semi-permeable membrane (Fritzmann et al. 2007). Thus, the water is forced across the membrane whereby the ionic species, such as ammonium, are retained in the waste stream (Health Canada 2013). Reverse osmosis usually requires pre-filtration to remove particulate matter as well as other pre-treatment steps, such as disinfection to limit bacterial activity, and the addition of anti-scaling agents or pH-adjustment to inhibit scale formation (Metcalf & Eddy Inc 2014, Fritzmann et al. 2007).

After RO, water with high dissolved solids/mineral content is converted to water with very low dissolved solids/mineral content. In full scale tests with feed concentrations of 33 mg ammonia-N/L (which includes the ionized and unionized ammonia), 94% ammonia rejection were achieved (Bellona et al. 2008). In other studies even > 98% (Bodalo et al. 2005) and > 96% (Kurama et al. 2002) rejection factors were obtained.

The advantages are:

- Small space requirements
- Few temperature effects
- Low start-up time and continuous and automatic operation (Kurama et al. 2002) as initial water parameters do not have major effect on treatment processes
- Removal efficiency of 96-99%
- Brine with high nitrogen concentration is a useful by-product as fertilizer (Koyuncu et al. 2001)

The disadvantages are:

- Pre-treatment has to be applied: removal of particulate matter and addition of chemicals to prevent scaling and fouling of the membranes (Fritzmann et al. 2007)
- Often, pre-filtration for particle removal and other pre-treatment steps are required
- High investment costs, but comparatively low operating costs (Kurama et al. 2002)
- Post-treatment has to be applied to re-mineralise and re-harden the water in order to adjust it to drinking water standards (Fritzmann et al. 2007).

In spite of its high efficiency and various other advantages, the treatment costs of RO are too high to use this process for water treatment in India. On the other hand, the membrane technique could be an intermediate solution to treat peak ammonium concentrations, if there is a large variation in water quality throughout the year and one technique alone would not be able to fulfil all requirements.

4.2 Biological filters

This method works through the accumulation of nitrifying bacteria through natural attachment to solid particles, forming so called biofilms. Ammonium and biodegradable organic matter (BOM) present in the raw water is removed simultaneously through microbial metabolism in the biofilm (Yu et al. 2007). The ammonium removal occurs in two steps: First, ammonium is oxidized to nitrite by ammonium oxidizing bacteria (mainly *Nitrosomonas*), and then nitrite is oxidized to nitrate by nitrite oxidizing bacteria (mainly *Nitrobacter*) (Andersson et al. 2001). Both oxidation reactions use oxygen as electron acceptor. Thus, sufficient oxygen has to be supplied through an aeration step prior to biological filtration (Lytle et al. 2013).

Depending on the filter material, biological granular activated carbon filters (GAC) and biological rapid sand filters are distinguished. The latter represent a mixture of rapid sand filtration for particle removal and biological treatment (Lee et al. 2014). Both filter material types are widely used in water treatment plants, while for wastewater treatment plants nitrifying trickling filters are preferred (Chaudhary et al. 2003).

To maintain a biofilter facility in an effective way, it is essential to understand the processes occurring within the filter. These are the attachment, growth and detachment of microorganisms (Chaudhary et al. 2003). According to Lee et al. (2014), the efficiency of ammonium removal depends on the ammonium loading rate and is independent of the flow and the influent ammonium concentration. Because of the comparatively low inflow concentration of < 2 mg NH_4-N/ L (Lee et al. 2014) the transferability to study sites with much higher ammonium concentrations is limited. Removal rates < 95% were achieved with feed concentrations below 4.5 mg NH_4-N/L (Health Canada 2013) and Yu et al. (2007) report the successful application of biofilters with feed water containing up to 5.3 mg/l NH_4-N. There are two possible adaptations to increase the efficiency: Either, the loading rate is corrected downwards or the filter area is extremely increased.

Incomplete ammonium removal can be caused by leaving the optimum temperature range (Andersson et al. 2001), insufficient oxygen supply (Lytle et al. 2013), nutrient (phosphorous) limitations (De Vet et al. 2010) and inadequate design and operation (Lopato et al. 2013). Generally, the technique is adopted to successfully treat water having ammonium levels up to 3 – 4 mg/l as was shown in various studies (Health Canada 2013). However, recommendations for an upper limit of ammonium in raw water could not be identified from the Health Canada review.

The advantages are:

- Cost-effective, as it only involves biological treatment
- Low construction and maintenance costs (Yu et al. 2007)
- Simple operation procedure (Yu et al. 2007)
- Simultaneous removal of biodegradable matter (BOM) (Yu et al. 2007)
- Reduction of taste and odor pollution (Chaudhary et al. 2003)

The disadvantages are:

- Based on pilot study results, Lytle et al. (2007) reported that colonization to obtain complete nitrification takes 2-3 months. This was achieved by constantly running aerated raw water through the filters to promote bacterial re-growth.
- The process may increase the level of nitrate and may release bacteria into the finished water. The treated water typically requires polishing (e.g. granular activated carbon [GAC] filtration) and post-treatment such as disinfection to ensure that neither undesirable organisms nor growth products pass into the distribution system (Health Canada 2013 after Wilczak 2006).
- Since nitrite, an intermediate product, is a toxic compound, WTPs need to ensure that treated water undergoes the complete biological process. Incomplete nitrification can occur due to the occurrence of elevated BOM concentrations (Manem and Rittmann 1992), competition for phosphate with other bacteria (De Vet et al. 2010), fluctuations in feed concentration (Rittmann 1990) and if O_2 concentrations are low (Lytle et al. 2013).
- The nitrate level at the outlet needs to be continuously monitored to ensure that NO_3 concentrations remain below guideline values. A de-nitrification unit is recommended to reach permissible nitrate concentration limits.

Although it is one of the least expensive methods for ammonium removal and suitable for Indian climatic conditions the treatment with biological filters can be recommended only with certain restrictions.

Biological filters rely on the microbial oxidation processes (NH_4^+ to NO_2 and NO_2 to NO_3). Therefore, enough oxygen has to be supplied in the feed water. This is difficult to achieve for ammonium concentrations above 1.5 mg/L (Lytle 2013). Janda and Rudovský (1994) propose a two-step nitrification or recirculation for ammonium concentrations around 4 mg/L. The Ranney well P3 already exceeds this limit and it is expected that ammonium concentrations in most of the Ranney wells at the Delhi site will increase in future. This means that the water from the Ranney wells has to be mixed with other water prior to treatment in order to reach a concentration range suitable for the microbiological filters. However, mixing Ranney well water with surface water will result in a variable water composition throughout the year and the microbiology within the biofilter might react very sensitive to these variations. Microbial filters need several days reaction time to adjust to changing inflow conditions (Rittmann 1990) such as fluctuations in nitrogen loading, biodegradable organic matter concentrations, phosphorous availability and oxygen concentrations. To achieve complete nitrification, raw water with relatively constant concentrations should be supplied. Lee et al. (2014) reported stable ammonium removal in biological filters despite of sudden changes in ammonium and loading rates, but in their study ammonium concentrations varied only between 0.04 and 0.2 mg NH_4-N/L, which is an order of magnitude lower than the concentrations found in the wells of the Delhi field site. If the application of this technique should be further enhanced in India, it is strongly recommended to conduct pilot and full scale studies to find optimum operating conditions for the specific local requirements. Malfunctions can only be detected through regular, very accurate sampling of the influents and effluents.

In general, biological treatment may be combined with breakpoint chlorination as this is reported to give good results (Health Canada 2013).

4.3 Emerging technologies

Ammonium is still a major problem in drinking water treatment and therefore research in various removal techniques is ongoing. Some of the emerging technologies are briefly identified in the following list (Health Canada 2013):

Trickling Filters

Tekerlekopoulou and Vayenas (2007, 2008) investigate trickling filters for simultaneous biological removal of ammonium, iron and manganese from potable water under different operating conditions. In a pilot scale study, influent ammonia concentrations in the range of 0.5 - 3.0 mg/L were reduced by 82% in the finished water.

Electrochemical Removal

This process makes use of the adsorption of ions on the surface of two oppositely charged electrodes. A charge barrier capacitive deionization process is reported as effective in removing total dissolved solids, nitrate and ammonia from water. In a pilot scale study, the process realized ammonium removal by 88% at 1000 mg/L NH_4-N as feed concentration (Brosèus et al. 2009).

Submerged Membrane Bioreactors

Although membrane methods have been applied primarily in desalinisation and to some extent in wastewater treatment (see chapter 4.1.4: Reverse osmosis), this technique has recently been considered as a new technology in drinking water treatment. Li and Chu (2003) and Tian et al. (2009) demonstrate with laboratory studies the effectiveness of hollow fiber membrane modules for ammonia removal from sewage influenced surface waters. The reported removal efficiencies obtained by the submerged membrane bioreactors through biological nitrification are in the range of 89 - 98%. The influent NH_3-N concentrations were in the range of 2.00 - 4.24 mg/L.

Table 6 Overview of post treatment options for raw water with elevated ammonium concentrations

	Principle	**Advantages**	**Disadvantages**	**Use in Delhi**
Physico-Chemical				
Air stripping of ammonia [1]Gauntlett 1980 [2]Huang & Shang 2006 [3]Health Canada 2013 [4]Jones et al. 2005	Adding lime increases pH of water to ~11; NH_4^+ converts to NH_3[1]; in contact with air, a gradient exists across the gas/liquid interface and NH_3 will be stripped to the air in a stripping tower[2]. Applied mostly in wastewater treatment[3].	- Increase in pH leads to killing of some pathogens and micro-organisms[4].	-To make water suitable for drinking purposes re-carbonation would have to be adopted[4]. - A volumetric air: water ratio of about 3000:1 is required to achieve effective NH_3 removal[1]. - Space for stripping towers.	-Suitable for Indian climate: high temperatures increase efficiency[2]. - Neutralization before distribution is necessary and WTP effluent has to be tightly monitored.
Ion exchange [5]Abd El-Hady et al. 2001 [6]Rahmani & Mahvi 2006 [7]Kurama et al. 2002 [8]Weatherley & Miladinovic 2004 [9]Li et al. 2011 [10]Govt. of Delhi 2011	Zeolites (e.g. Clinoptilolite and Chabazite) are selective for NH_4^+ ions. Water is passed through a bed of zeolites to achieve 86-99% ammonium removal, depending on concentration and water composition. Method successful for up to 200 mg/L NH_4^+ [3].	-Zeolites can be regenerated using NaCl[5] or biological regeneration[6]. -Method has no sensitivity to fluctuation in NH_4^+ influent concentration[6].	- Inconvenient for WTPs with a capacity >18 MGD because of space required for ion exchange columns[7]. -Presence of Ca reduces NH_4 adsorption onto the zeolite[8]. - High input concentrations require large volumes of zeolites[9].	-WTPs in Delhi have a capacity >40 MGD[10], therefore method not suitable to retrofit into existing WTPs. Could be used for treating NH_4^+ contaminated GW from Ranney/ tube wells by clustering a few wells and installing a small treatment facility for the NH_4^+ treatment prior to conventional treatment.
Breakpoint chlorination [11]Donnermair and Blatchely III 2003 [12]Takó & Laky 2012 [13]Janda & Rudovský 1994 [14]Griffin & Chamberlin 1941	By adding chlorine to water, a stepwise reaction takes place with the NH_4^+-N, first forming mono- and di-chloramines and then, at the breakpoint, N_2, NO_3^- and free residual chlorine. Method best suitable for NH_4^+ <<1mg/L[1].	-The mono-chlor-amine and di-chloramine formed, act as a potential disinfectant[11]. -Can be combined with As removal[12].	-Cl reacts with organic material and by-products are formed. Activated carbon adsorber to be installed too, making treatment expensive[12, 13]. - Requires frequent monitoring of NH_4^+ and chlorine concentrations[3]. - High ammonia-N:chlorine ratios.[14]	-Frequent variations in raw water quality: chlorine dosage has to be continuosly adjusted to reach breakpoint. -Treatment presumably has to be followed by carbon adsorbers because of organic compounds in the raw water. -NH_4^+ in raw water often >1mg/L.
Reverse Osmosis (RO) [15]Bellona et al. 2008 [16]Bodalo et al. 2005	Water is forced across a semi permeable membrane and molecules and ions, (NH_4^+), are retained[3]. 94% NH_3 removal in full scale tests with feed concentrations of 33 mg/L[15]. In other studies >98%[16] and >96%[7].	-Small space required. -Low start-up time and continuous operation[7]. Initial water parameters don't have major effect on treatment process.	-Often pre-filtration for particle removal + other pretreatment steps[3]. -High investment costs, but comparatively low operating costs[7]. -Mineral imbalances can increase corrosive nature of the effluent and post treatment might be necessary[3].	-Membrane technique would be best to use as an intermediate solution to treat peak concentrations. Too expensive for regular use.
Biological				
Biological filters [17]Yu et al. 2007 [18]Andersson et al. 2001 [19] Lytle et al. 2013 [20]Manem & Rittmann 1992 [21]De Vet et al. 2010 [22]Rittmann 1990	Biofilms form through accumulation of nitrifying bacteria on filter material. NH_4^+ is oxidized to NO_2^- and then to NO_3^- by different bacteria[17, 18]. Sufficient O_2 has to be supplied through aeration step prior to filtration[19]. Removal rates <95% with feed concentrations below 4.5 mg NH_4-N/L[3]	-Low construction and maintenance costs[17]. -Simplicity in operation[17]. -Biodegradable matter (BOM) removed simultaneously[17].	-Colonization takes 2-3 months[19]. -Increases nitrate levels, may release bacteria into the treated water[3]. -Incomplete nitrification can occur due to: occurrence of elevated BOM concentrations[20], competition for phosphate with other bacteria[21], fluctuations in feed concentration[22], if O_2 concentrations are low[19]	-Filters react sensitive to changes in raw water concentrations, which are common in Delhi due to mixing of different groundwater sources and surface water. Microbiology reacts sensitive to this and reliable functioning is not guaranteed. -Malfunctions detected only through regular, very accurate sampling of influents and effluents.

5 Conclusion and recommendations

Based on the results of this study, the use of BF in the Yamuna floodplain in Delhi and in similar hydrogeological settings is basically recommended. However, in these locations, bank filtration should not be seen as a natural treatment option, but as an option to adapt and improve water management measures. The two main advantages are (1) temporary water storage in the aquifer and (2) a homogenisation of the raw water composition, improving operating conditions for WTPs. In addition to BF, it is advised to set up an adapted post treatment concept, which is designed specifically for the groundwater parameters at the particular location. Such an adapted site-specific post treatment would have the advantage that it not only reduces elevated ammonium concentrations caused by the infiltration of sewage water, but it would also allow to treat other (geogenic) parameters of concern, for example arsenic and fluoride.

In the long term, it is essential to improve the river water quality by implementing sufficient sewage treatment capacity. As this has been widely recognized several new sewage treatment plants are planned or under construction, e.g. five STPs with a designed capacity of 360 MLD (millions of litres per day) are likely to be commissioned in 2014-15 (Government of India 2014).

As discussed, elevated ammonium concentrations will still prevail long after source water quality improves. According to the laboratory column experiments (section 2.4) and a simplified 1D reactive transport model that was set up for the field site (section 2.5), ammonium desorption in the gravel layer - where the laterals of P3 are presumably located - will take decades. Taking the assumptions described in section 2.5, the period of ammonium desorption to concentrations < 0.5 mg/L will last for about 61 years in the 500 m strip along the river. This result of the simple 1D model is, however, a conservative estimate, as the average linear flow velocity in the gravel layer is probably much higher than the literature value (Sprenger 2011) suggests. In general, due to the continuing accumulation of ammonium on the aquifer matrix, desorption times will increase with prolonged infiltration of contaminated surface water. Thus, a short and medium term solution such as post treatment remains a necessity for the investigated site, even if river water quality improves fast. More detailed 2D models are strongly recommended to make more precise and realistic predictions.

5.1 Recommended remediation

In general, ammonium remediation options are complex and expensive. At BF sites such as Delhi remediation would often be further complicated by the fact that wells are spread along a long stretch of the river. In-situ or pump and treat remediation measures would therefore have to be installed over large areas making remediation extremely costly. If it is decided to develop such a remediation concept, it is of utmost importance to implement the following recommendations:

- Installation of multi-level observation wells at the well field including levelling survey: Regular measurements of the water level when wells are operating and when they are switched off and creation of groundwater contour maps for the different seasons.

- Development of a groundwater flow model based on the water level data: The hydraulic conditions at the well field have to be well known in order to be able to decide on a concept. It is especially important to understand how the wells influence the flow regime.
- Modelling of scenarios of possible remediation concepts.
- Implementation of groundwater monitoring by regularly taking groundwater samples from the observation wells: To be able to evaluate remediation measures, groundwater quality has to be known and documented before the start of remediation measures.
- Implementation of an accompanying groundwater monitoring during remediation.

In case remediation is wanted, it is not recommended to use any option involving the injection of chemicals or additives other than oxygen into the aquifer. Although those methods usually are characterized by a faster removal of ammonium, there is a risk of unwanted secondary reactions and formation of by-products, which might not get degraded on the short flow paths to the production wells. BF sites are always water protection zones and therefore special precaution should be taken.

5.2 Recommended post treatment

Two options were identified as the most applicable for the given context of high ammonium concentrations in raw water: 1. raw water from affected wells can be mixed with raw water from other sources before treatment or 2. raw water from wells can be treated separately in independent WTPs.

In Delhi, the first option is generally chosen. Raw water from the Ranney wells is usually mixed with surface water before treatment. This has the advantage that the groundwater from the Ranney wells is diluted and parameters such as arsenic remain below the guideline values and do not need to be treated. A major disadvantage of this method is that water quality is not constant. Nitrogen concentrations in the Yamuna river upstream Delhi show high variations. As the treatment plants are not designed to cope with peak concentrations, it is not always possible to remain below the guideline values for ammonium and/or nitrate. Furthermore, the mixing of ammonium contaminated groundwater with surface water might increase ammonium concentrations in the WTP influents to a level where chlorination is negatively affected.

The second option is therefore recommended. It is better to treat the raw water from the floodplain aquifer separately, e.g. by further pursuing the concept started with the Common Wealth Games Village WTP, a 4.5 MLD WTP for the water from P4 and nearby bore wells, and the 27 MLD nitrification plant in Okhla.

Although microbial filters (nitrification filters, Table 4.5), as for example used in the Okhla nitrification plant, are a common and cost-efficient option to treat ammonium in raw water, most studies about biological filters for drinking water treatment were not conducted under conditions met in India and the results cannot directly be transferred. Challenges to be met with this technique in locations like Delhi include:

- Supply enough oxygen to cope with the high ammonium concentrations.

- Monitor both inlet and outlet concentrations closely and adapt the hydraulic loading to stabilize nitrogen loading and thus achieve complete nitrification.

Lee et al. (2014) reported stable ammonium removal in new, less concentration sensitive biological filters. However, the reported range of ammonium concentrations is an order of magnitude lower than in the raw water of the Ranney wells in Delhi. Therefore, pilot and full scale studies to find optimum operating conditions for the specific local requirements are recommended if the application of this technique should be further enhanced in India. As an alternative to biological filters, a method with more robustness towards fluctuating input parameters and less downtime in case of failures should be considered, such as ion exchange using zeolites.

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