REPORT

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BANK FILTRATION AND AQUIFER RECHARGE FOR DRINKING WATER PRODUCTION: APPLICATION, EFFICIENCY AND PERSPECTIVES

- AN INTEGRATION OF NASRI OUTCOMES AND INTERNATIONAL EXPERIENCES

by

G. Grützmacher, B. Wiese, I. Hülshoff, D. Orlikowski, E. Hoa
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Title

BANK FILTRATION AND AQUIFER RECHARGE FOR DRINKING WATER PRODUCTION: APPLICATION, EFFICIENCY AND PERSPECTIVES - AN INTEGRATION OF NASRI OUTCOMES AND INTERNATIONAL EXPERIENCES

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Extended Summary

Bank filtration (BF) and aquifer recharge (AR): aquifer storage recharge (ASR), aquifer storage transport recharge (ASTR); are natural and semi-natural methods for drinking water treatment and constitute a major barrier within water supply system. Recent investigations have shown that about 60 % of Berlin's drinking water is produced via BF or AR (Zippel & Hannappel 2008). Most drinking water therefore originates from surface waters within the cities limits and is pumped from wells adjacent to it's many lakes and rivers. Since more than 100 years this system has been supplying safe drinking water so that post-treatment is limited to aeration and subsequent sand filtration. Disinfection is usually not applied (SenStadtUm 2008).

The research project NASRI ("Natural and Artificial Systems for Recharge and Infiltration", KWB 2002 – 2006), funded by the Berliner Wasserbetriebe (BWB) and Veolia (VE) had the aim to characterize the specific hydraulic and hydrochemical conditions at selected BF and AR sites in Berlin and to assess the behaviour of major water constituents, trace organic substances, algal toxins and pathogens during subsurface passage. For this, field investigations at three transsects (Lake Tegel BF-site, Lake Tegel AR-site and Lake Wannsee), laboratory and technical scale experiments were carried out by 7 different working groups. The results of the investigations were documented in 6 extensive research reports and were the basis for nearly 50 scientific publications.

In 2007 the IC-NASRI project (Integration & Consolidation of the NASRI outcomes) was initiated by VE and BWB in order to support the practical implementation and optimization of bank filtration and aquifer recharge for drinking water production with the experience gained during the NASRI project. The aim was to derive practical guidelines for design and operation of BF & AR systems by i) further interpretation of the NASRI data and ii) integrating experience from other BF / AR sites world wide.

Although subsurface passage is characteristic to many systems of managed aquifer recharge (MAR) the investigations within IC-NASRI concentrated on systems where drinking water is produced by infiltration of surface water either from the banks of a lake / river or from infiltration ponds (or similar systems like ditches or irrigation fields). A transfer of the presented results to other MAR systems, which use different recharge methods (e.g. ASR) or different sources (e.g. treated wastewater) therefore needs to be considered carefully, even though many statements may be true for them as well.

This reports aims at providing engineers and scientists involved in drinking water production by BF & AR with up-to-date information on settings of similar systems world wide and on the systems' performance with regard to drinking water treatment. The aim was to give the reader a condensed overview of the topic whereas further details can be taken from the large number of references given in the bibliography.

An analysis of past and current applications of bank filtration (BF) and aquifer recharge (AR) for drinking water production yielded that in Europe, bank filtration (BF) has a long history in safe water production for potable use and was initially implemented in the 19th century with first applications reported from England as early as 1810. Since then bank filtration and, in case of insufficient quantity, aquifer recharge (AR) have been generally applied as a first barrier within the drinking water treatment chain. In many cases additional purification processes became necessary after the 1960's when the quality of the water in the catchment areas was decreasing. A statistical analysis of 170 BF & AR sites for drinking water production showed a high variability of settings: the analyzed sites with capacities ranging from 70 to 780 000 m³/d were situated within aquifers that showed a maximum thickness

between 3 and 300 m. Hydraulic conductivities documented ranged from < $1*10^{-4}$ to $5.7*10^{-01}$ m/s and the distance between bank and well was given as < 10 to 3,500 m. Despite this high variability it is clear that the major objective is to enhance water availability whereas water quality issues seem of minor relevance.

Post-treatment (and pre-treatment for AR-sites) were analyzed at 56 different sites (16 BF sites, 40 AR sites). The compiled data showed that depending on source water quality, travel time and redox conditions a typical treatment chain at an AR site would include particle elimination as pre-treatment for clogging reduction and sometimes iron- and manganese removal (e.g. via aeration and sand filtration). The latter was applied at all analyzed BF sites, showing that this is the usual post-treatment process. Advanced post-treatment (e.g. ozonation, GAC) and final disinfection where also frequently applied – showing that the situation in Berlin, without disinfection and advanced post-treatment is quite exceptional, even for German standards.

International regulations and technical guidelines for BF sites concentrate on i) general source water protection and ii) describing certain pre-requisites for substance elimination (e.g. 50-days-line in Germany or pre-requisites for protozoa removal in the US LT2ESWTR). For AR sites in Europe the quality and quantity of infiltrated water needs to comply with the Water Framework Directive and Groundwater Directive, i.e. no deterioration of the water status may take place. General rules are lacking, but may also not be applicable due to the site specifity. Different approaches are currently discussed in Europe, the US and Australia (e.g. via risk assessment).

Concerning the performance of bank filtration and aquifer recharge performance for substance removal derived from investigations at existing field sites, different classes of substances / substance groups can be distinguished:

1. Substances or substance groups, for which high removal is possible (> 90 %), provided optimal site design:
    - suspended solids,
    - pathogens,
    - cyanobacterial (algal) toxins,
    - nitrate
    - disinfection by-products (THMs).
2. Substance groups, including substances that show highly variable removal (typically 25 to 75 % overall removal):
    - dissolved organic carbon (DOC),
    - pesticides,
    - pharmaceuticals and x-ray contrast media,
    - chlorinated hydrocarbons,
    - mono- and polycyclic aromatic hydrocarbons,
    - endocrine disrupting compounds
3. Substances which are generally poorly or not removed by BF & AR systems or may even become mobilized
    - ammonium
    - major anions & cations (hardness, salinity)
    - inorganic trace elements

Within groups 2 certain individual substances were identified that have the potential to break through BF & AR systems, even for high contact times:

- potentially persistent pesticides
    - triazines: atrazine, desethylatrazine, desisopropylatrazine, cyanazine, propazine, simazine, terbutylazine
    - ureas: chlortoluron, diurone, isoproturone
    - phenoxy-herbicides: mecoprop (under anoxic conditions)
    - organochloride insecticides: hexachlorbenzene (HCB), pentachlorophenole (PCP),
    - anilides / anilines: dimethenamid, metolachlor
    - others: bentazone (plus its transformation product 2-amino-n-isopropylbenzamide), glyphosate, dikegulac
- potentially persistent pharmaceuticals and x-ray contrast media:
    - regardless of redox zone: sulfadimidine, primidone, AMDOPH, carbamazipine (< 20 % under oxic conditions), AOI (< 31 % under oxic conditions)
    - under oxic conditions: sulfamethoxazole
    - under anoxic conditions: phenazone, clindamycine
- 1,1,1-trichloroethane (TCA), trichloroethylene (TCE) and tetrachloroethylene (PCE)
- potentially persistent monocyclic aromatic hydrocarbons: benzene, m-, o-, and p-xylene,
- potentially persistent polycyclic aromatic hydrocarbons (PAH): phenanthrene, flouroanthrene

For all organic micropollutants, the toxicity and persistence of transformation products needs to be taken into account. This is currently a field of research and could be addressed with targeted toxicity testing.

Concluding, it can be stated that the efficiency of BF and AR sites depends strongly on the hydrogeological setting as well as on source water quality. Substance removal and potential mobilization need to be taken into account. In every case, however, it represents an additional barrier for drinking water production from surface water, thus adding substantially to the security and sustainability of the water supply system.

Acknowledgements

Firstly, the authors would like to thank all scientists, technicians and students involved in the NASRI project for contributing to the extensive knowledge that was obtained on the processes in Berlin’s subsurface systems. The following working group leaders were involved in the project;

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- Dr. J. Lopez-Pila, Dr. R. Szewzyk (German Federal Environmental Agency) & Prof. U. Szewzyk (Technical University of Berlin)
- Dr. U. Dünnbier, E. Wittstock & H. Dlubek (Berliner Wasserbetriebe)
- Prof. Dr. M. Jekel (Technical University of Berlin)
- Prof. Dr. M. Jekel & Dr. T. Heberer (Technical University of Berlin)
- Prof. Dr. A. Pekdeger (Free University of Berlin)
- Prof. Dr. G. Nützmann (IGB Leibniz-Institute of Freshwater Ecology and Inland Fisheries)

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Glossary

adsorption	reversible attachment of particles to the aquifer matrix leading e.g. to a retardation of contaminants during subsurface passage
ambient groundwater	groundwater not influenced by surface water or aquifer recharge, typically recharged by precipitation
anoxic zone	combination of anoxic and anaerobic zone according to McMahon and Chapelle (2008), characterized by the absence of oxygen and nitrate and the presence of dissolved iron or manganese (see Table 5, p. 17).
aquifer recharge (AR)	method of MAR typically using surface spreading techniques (e.g. infiltration ponds) to enhance groundwater recharge with stormwater, surface water or treated wastewater (Figure 1)
attenuation	decrease in concentration by mixing and removal (see Figure)
bank filtration (BF)	method of MAR that induces infiltration of water from a river (river-BF) or lake into the subsurface by pumping of wells in the adjacent aquifer
(bio)degradation	These terms relate to a (biologically induced) change of the chemical structure of the respective substance, such that is not detected any more with the used analytical method. Depending on the method, the structural change may be small. For many trace organics, the pathway of (bio-) degradation and the degradation products (-> transformation products) are unknown.
capacity (m^3/d)	maximum volume of water a water works is capable of producing
decay	either die-off of micro-organisms or radioactive decay
elimination	concentration decrease in the subsurface due to decay or (bio-) degradation of substances or filtration of particles
filtration	removal of particles due to straining, electrostatic or other sorptive effects
flow time	travel time of a tracer from the point of infiltration to the well
groundwater	water in the saturated zone of an aquifer
groundwater recharge	comprises recharge by rain or other sources (e.g. MAR)
ion exchange	exchange between different ions adsorbed to the aquifer matrix (in aquifers: mainly cation exchange)
managed aquifer recharge (MAR)	the recharge of stormwater, surface water or treated wastewater into an aquifer by hydraulic means (e.g. pumping of wells in the aquifer adjacent to a river (BF)), via infiltration ponds (AR via pond infiltration) or by injection wells (aquifer storage and recovery – ASR)
mixing	denotes the merging of different waters, e.g. bank filtrate and ambient groundwater or old bank filtrate (see Figure). The mixing concentration C_m [g/m^3] is calculated by

	$C_m = \frac{C_1\,r_1 + C_2\,r_2}{100}$ with $C_{1/2}$ as concentrations [g/m^3] and $r_{1/2}$ as ratios of the two waters [%].
natural groundwater recharge	process of groundwater replenishment by rain or snow melt.
oxic zone	combination of oxic and suboxic zone according to McMahon and Chapelle (2008), characterized by the presence of oxygen, nitrate and sulfate and the absence of dissolved iron and manganese (see Table 5, p. 17).
pond infiltration	aquifer recharge via infiltration ponds
post-treatment	water treatment after subsurface passage
pre-treatment	water treatment prior to subsurface passage
precipitation	formation of a solid substance from dissolved substances, typically in case of pH- or E_H changes (e.g. iron-hydroxide precipitation as a result of aerating iron(II)-rich groundwater)
raw water	water abstracted from a well prior to post-treatment
recharged water	water entering the subsurface by anthropogenic activity, either directly by infiltration (i.e. building up a pressure within the recharge facility) or by abstraction (i.e. creating a negative pressure in the subsurface). Natural gradients may also cause the infiltration of surface water. According to DIN 4049, this water is referred to as “unechtes Grundwasser” (i.e. pseudo-groundwater). Please compare with “groundwater recharge”.
removal	comprises all processes but mixing that contribute to the decrease of contaminant concentrations (compare also: attenuation)
retardation	ratio of a substance’s flow velocity V_s to water velocity V_w ($R=V_w/V_s$ with R as retardation factor) due to ad- and desorption
straining	mechanical component of filtration, the affected particle size may be smaller than pore sizes of the matrix
source water	surface water used for MAR (e.g. stormwater, lake / river water, treated waste water)
transformation products	chemical substances that are formed by -> (bio-)degradation of other substances
travel distance	horizontal distance between the point of infiltration and the production well
travel time	travel time of a substance from the point of infiltration to the well, travel time is calculated: $t_t = t_f\ R$ where t_f is flow time and R the retardation factor (i.e. for tracers the travel time is equal to the flow time) synonymous to residence time or contact time.
water reuse	(usually non-potable) use of treated waste water

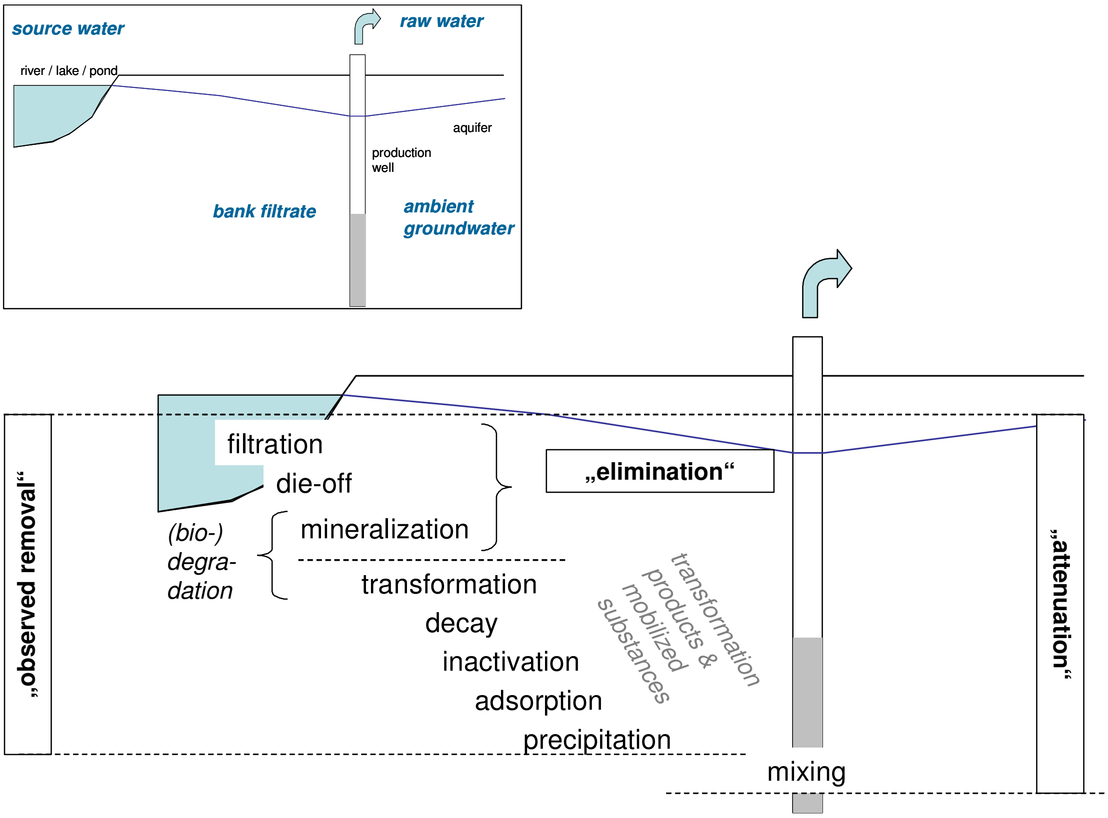

Figure 2: Schematic figure of processes that may reduce substance concentrations during subsurface passage and definition of terms used.

Table of Contents

List of Figures

List of Tables

Chapter 1
Introduction

1.1 Background

Bank filtration (BF) and aquifer recharge (AR) via pond infiltration are natural and semi-natural methods for drinking water treatment and constitute a major barrier within the Berlin water supply system. Recent investigations have shown that about 60 % of Berlin's drinking water is produced via BF or AR (Zippel & Hannappel 2008). Most drinking water therefore originates from surface waters within the cities limits and is pumped from wells adjacent to it's many lakes and rivers. Since more than 100 years this system has been supplying safe drinking water so that post-treatment is limited to aeration and subsequent sand filtration. Disinfection is usually not applied (SenStadtUm 2008).

The research project NASRI ("Natural and Artificial Systems for Recharge and Infiltration", KWB 2002 – 2006), funded by the Berliner Wasserbetriebe (BWB) and Veolia (VE) had the aim to characterize the specific hydraulic and hydrochemical conditions at selected BF and AR sites in Berlin and to assess the behaviour of major water constituents, trace organic substances, algal toxins and pathogens during subsurface passage. For this, field investigations at three transsects (Lake Tegel BF-site, Lake Tegel AR-site and Lake Wannsee), laboratory and technical scale experiments were carried out by 7 different working groups (Table 1).

Table 1: Overview of the working groups involved in the NASRI project.

Working group leaders	**Title of the work package**
Dr. I. Chorus & Dr. H. Bartel (German Federal Environmental Agency, UBA)	*Retention and elimination of cyanobacterial toxins (microcystins) through artificial recharge and bank filtration*
Prof. Dr. J. Lopez-Pila (German Federal Environmental Agency, UBA) & Prof. Dr. U. Szewzyk (Technical University of Berlin)	*Using bacteriophages, indicator bacteria, and viral pathogens for assessing the health risk of drinking water obtained by bank filtration.*
Dr. U. Dünnbier, E. Wittstock & H. Dlubek (Berliner Wasserbetriebe)	*Data management, sampling, analyses and interpretation*
Prof. Dr. M. Jekel (Technical University of Berlin)	*Organic substances in bank filtration and artificial ground water recharge – process studies*
Prof. Dr. M. Jekel & Dr. T. Heberer (Technical University of Berlin)	*Occurrence and fate of drug residues and related polar contaminants during bank filtration and artificial recharge*
Prof. Dr. A. Pekdeger (Free University of Berlin)	*Hydrogeological - hydrogeochemical processes during bank filtration and ground water recharge using a multi tracer approach*
Prof. Dr. G. Nützmann (IGB Leibniz-Institute of Freshwater Ecology and Inland Fisheries)	*Integrated modelling concepts for bank filtration and artificial ground water recharge processes: coupled ground water transport and biogeochemical reactions*

The results of the investigations were documented in 6 extensive research reports (to be published on the KWB homepage) and were the basis for nearly 50 scientific publications (a list of the published research papers is given in Appendix A).

1.2 Scope of the IC-NASRI project and final report

In 2007 the IC-NASRI project (Integration & Consolidation of the NASRI outcomes) was initiated by VE and BWB in order to support the practical implementation and optimization of bank filtration and aquifer recharge for drinking water production with the experience gained during the NASRI project. The aim was to derive practical guidelines for design and operation of BF & AR systems by i) further interpretation of the NASRI data and ii) integrating experience from other BF / AR sites worldwide.

Although subsurface passage is characteristic to many systems of managed aquifer recharge (MAR) the investigations within IC-NASRI concentrated on systems where drinking water is produced by infiltration of surface water either from the banks of a lake / river or from infiltration ponds (or similar systems like ditches or irrigation fields). A transfer of the presented results to other MAR systems, which use different recharge methods (e.g. ASR) or different sources (e.g. treated wastewater) therefore needs to be considered carefully, even though many statements may be true for them as well.

This reports aims at providing engineers and scientists involved in drinking water production by BF & AR with up-to-date information on settings of similar systems worldwide and on the systems' performance with regard to drinking water treatment. The aim was to give the reader a condensed overview of the topic whereas further details can be taken from the large number of references given in the bibliography (Chapter 5).

Chapter 2
Bank Filtration and Aquifer Recharge for Drinking Water Production – Typical Settings, Trends in International Regulations

2.1 Past and Current Applications of BF & AR for Drinking Water Production World Wide

In Europe, bank filtration (BF) has a long history in safe water production for potable use and was initially implemented in the 19th century when direct use of surface water was identified as unsuitable for direct consumption. In England, facilities catching bank-filtered water have been reported as early as 1810 (BMI, 1985). The oldest catchment area in the Netherlands is located in Nijmegen and dates back to 1879 (Hiemstra *et al.*, 2003). In the 1840's, modern central water supplies in Germany implemented extraction wells in the river valleys and used bank filtration unintentionally. The systematic production of bank filtrate started in Germany between 1870 and 1890 (BMI 1975, 1985). Since then bank filtration and, in case of insufficient quantity, aquifer recharge (AR) have been generally applied as a first barrier within the drinking water treatment chain. In many cases additional purification processes became necessary after the 1960's when the quality of the water in the catchment areas was decreasing.

BF currently constitutes an important treatment step - often as part of a multi barrier concept - for drinking water production in many European countries such as France, Finland, Hungary and Switzerland (Tufenkji *et al.*, 2002). For France, the only estimation has been published by Doussan et al. (1997), who stated that the proportion of water pumped from aquifers with direct hydraulic contact to adjacent surface waters accounts for approximately 50% of the total drinking water production (Castany, 1985). In the Netherlands, 13% of drinking water is produced from infiltrating surface water of which 5% derives from bank filtration (Hiemstra *et al.*, 2003). In Germany, BF and AR is applied in the river valleys of the Rhine, Main, Elbe, Neckar, Ruhr while in Berlin, lake water fed by the rivers Havel and Spree is abstracted (Grischek *et al.,* 2002). According to the German Federal Agency for Statistics (2004), bank filtration in Germany accounts for 5.3 % of the drinking water produced but its share is likely higher since former definitions rated bank filtrate as groundwater. Berlin, for instance, for which 60% is derived from bank filtration and artificially recharged groundwater (Zippel & Hannappel 2008) is classified as a federal state abstracting groundwater only. In the United States, BF is usually applied for drinking water pre-treatment with increasing popularity due to its ability to remove chlorine resistant *Cryptosporidium* oocysts (Ray *et al.*, 2002).

In order to compare the situation in Berlin with other sites world-wide, a literature survey was carried out on existing sites at which bank filtration and aquifer recharge *(excluding ASR / ASTR systems)* are carried out for drinking water production. In scan 272 sites were identified and included in the study. The data was compiled in an MS-ACCESS data base (provided on a CD in the appendix) and is evaluated below.

The bulk of the 272 sites is situated in Europe (63%, Figure) followed by North America (19%), Asia/Oceania (13%), Orient (2%), South America and Africa (3%). Most information was found on field sites in: The Netherlands (58 sites, Figure 3), USA (47), Germany (44), India (15), Finland and The United Kingdom (in 10).

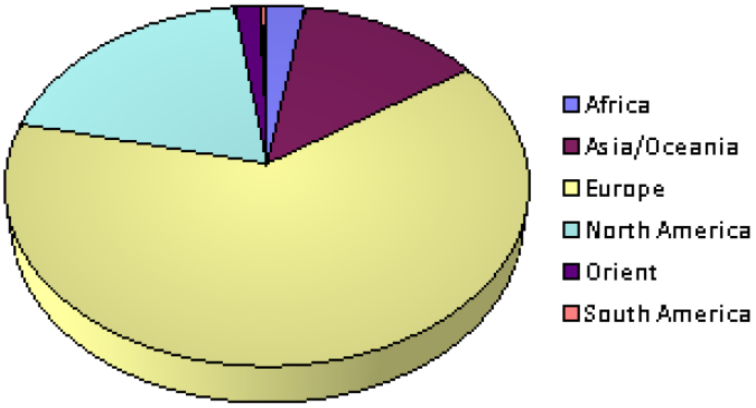

Figure 3: Locations of the identified MAR sites.

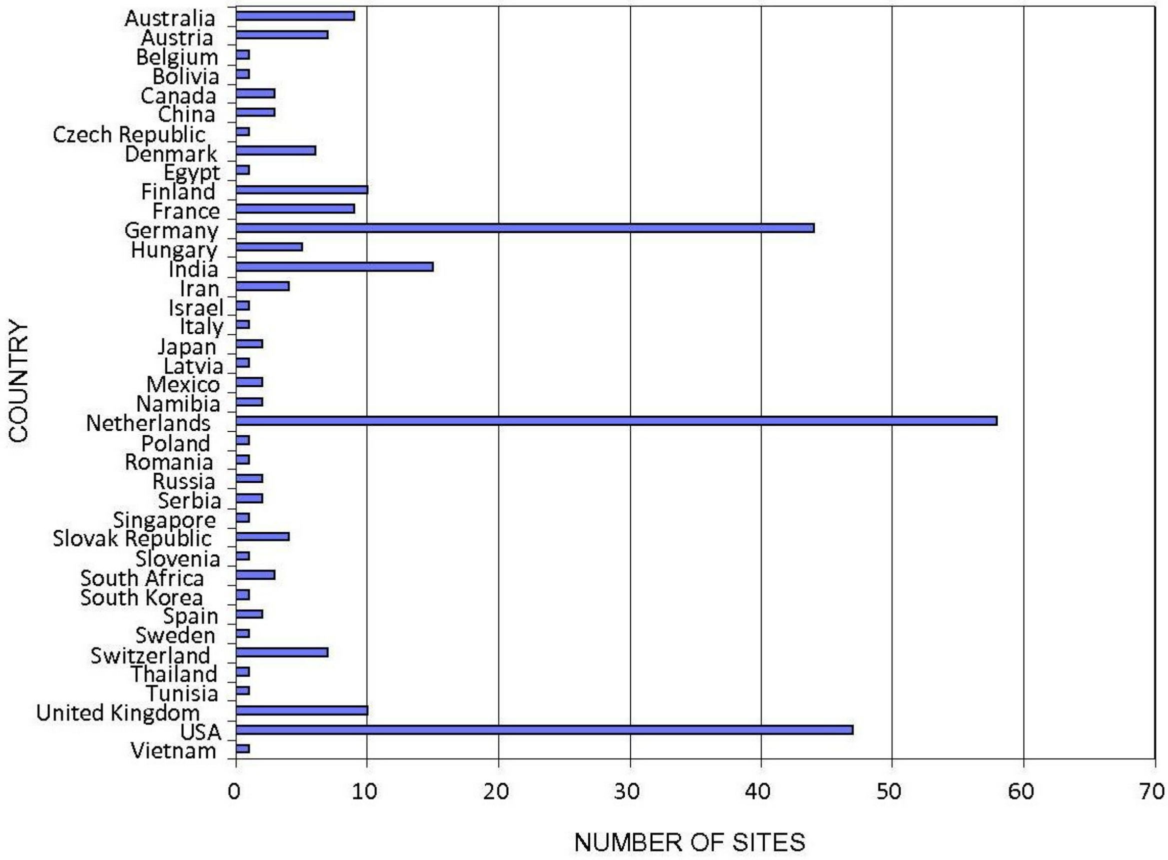

Figure 4: Identified MAR sites per countries.

Bank filtration is the most common type of recharge (187 sites, Figure 5a). Fourteen of the identified bank filtration sites also use other methods of MAR mainly pond infiltration (Figure 5b). Together with these sites the number of facilities which produce drinking water via other methods of aquifer recharge (mainly pond infiltration) amounts to 71 (Figure 5c). *Other techniques like dune infiltration, sprinkling or infiltration ditches are of minor relevance.*

Majority of BF sites is situated in: the Netherlands (51 sites, Figure 5a), but 17 sites was abandoned (Stuyfzand *et al.*, 2004), the USA (38), Germany (26) and India (14). However, on 8 sites, which are located in Germany (Figure 5b), BF processes apply with other methods of MAR, for example Mühlheim process. The most part of AR sites was built in: Germany (10 sites, Figure 5c), the USA (9), Australia (7), the Netherlands (6) and Denmark (6).

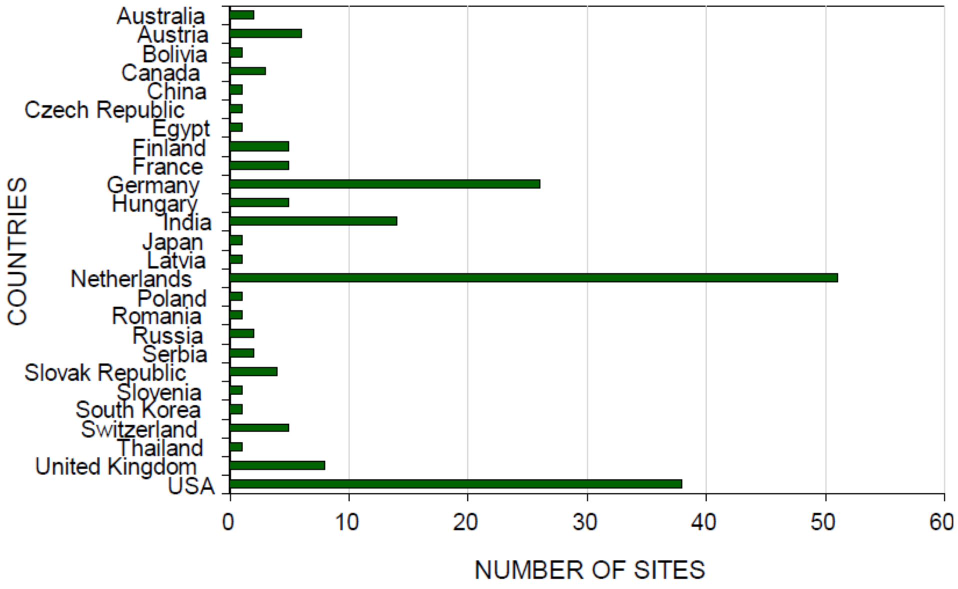

Figure 5a

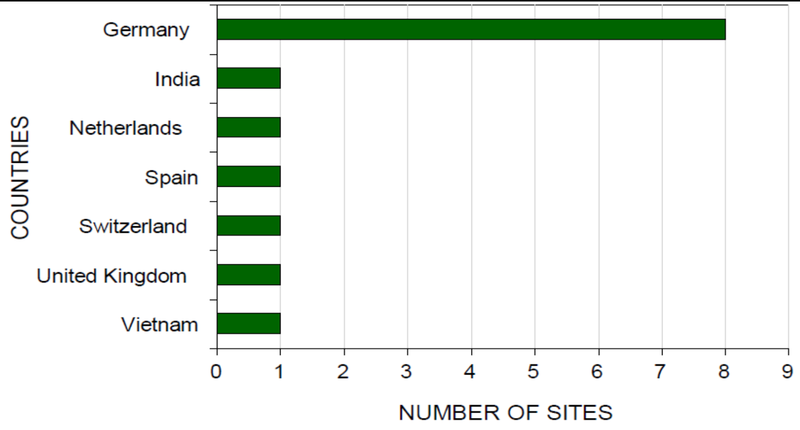

Figure 5b

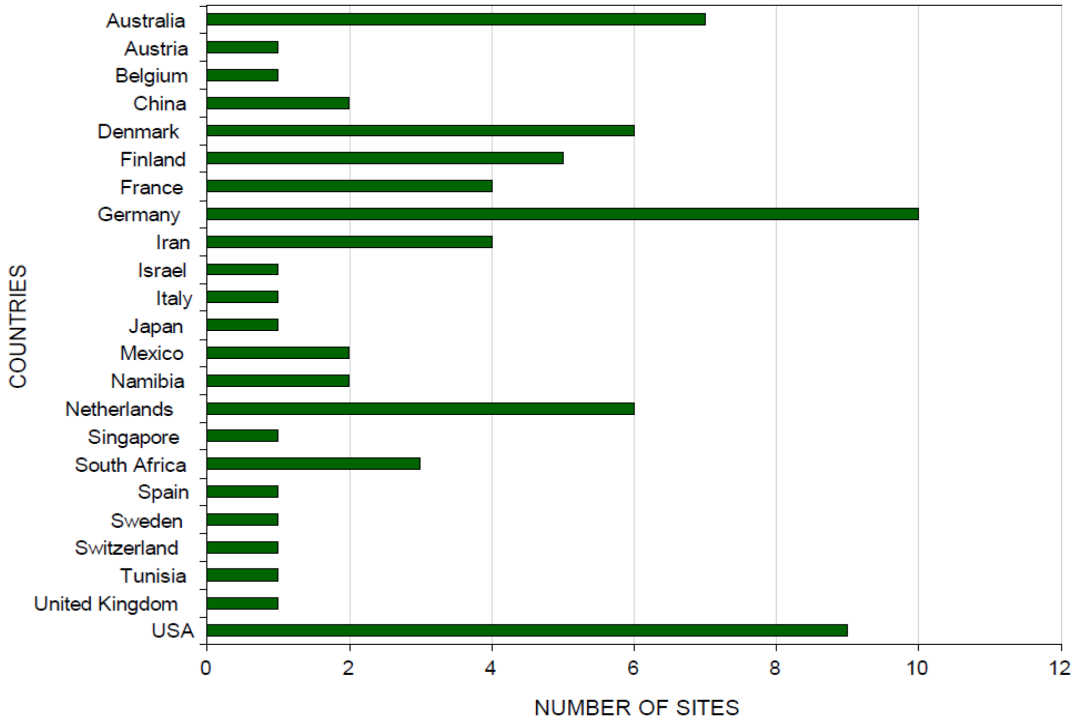

Figure 5c

The majority of source water originates from rivers 181 BF sites and only 19 are derived from lakes. All sites using lake water are - with the exception of one site each in Australia and India - located in Central and Northern Europe.

For the 121 BF/AR sites for which the capacity or currently produced water volume is known, the figures vary widely (Figure 1) between 70 m^3/d (Linsental waterworks, Winterthur, Switzerland) to 780.000 m^3/d (Anaheim Lake, Orange County, USA) with a median value of 25.000 m³/d. Berlin's main water works, namely Spandau, Friedrichshagen, Beelitzhof and Tegel, are all well above this median value with capacities ranging from 160.000 to 250.000 m³/d.

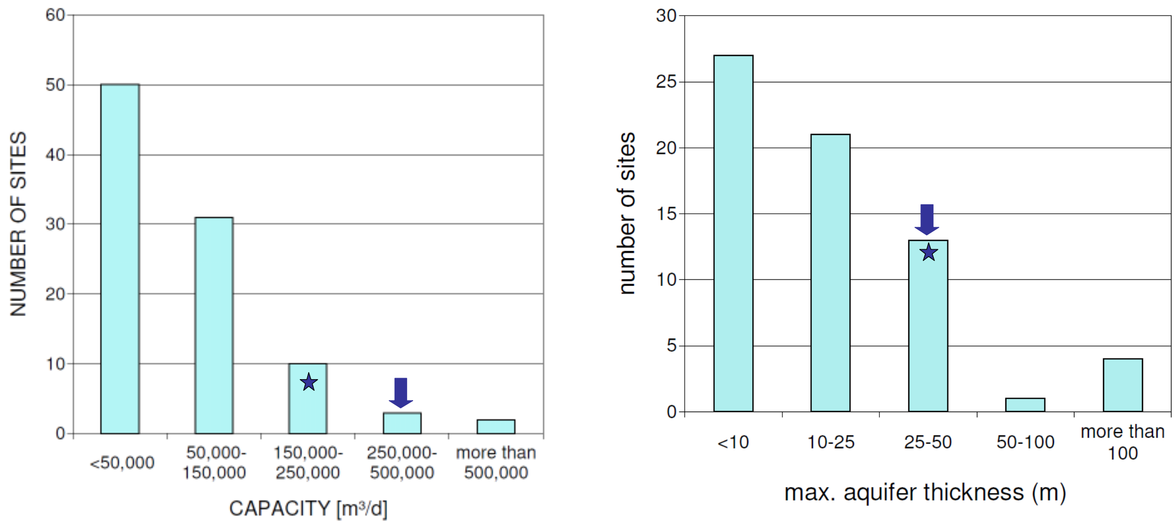

Figure 1: Capacity or currently produced volume of water (left) and max. aquifer thickness (right) of 121 and 64 AR / BF sites worldwide. Berlin's main water works and the Lyon site typically fall into the star- and arrow-labelled categories, respectively.

Of the 78 field sites for which information on the well type was available, 53 (67 %) use vertical wells and 26 (33%) horizontal wells. Of these 12 (15%) sites operate both horizontal and vertical wells. The type of well is closely linked to the thickness of the aquifer with vertical wells being installed in thick aquifers (average: 26 m) and horizontal wells in shallow aquifers (average: 11 m). The maximum aquifer thickness of 69 sites ranges between 5 and 360 m (see Figure 1). For the majority of sites (72%), the maximum aquifer thickness is less than 50 m. It is remarkable that the wells of the site with one of the highest capacities (400,000 m^3/d) at Csespel Island (Budapest, Hungary) are situated in an aquifer of only 12 m maximum thickness.

The maximum hydraulic conductivities from 62 sites are given in Figure 2 (median: $2*10^{-3}$ m/s). In Berlin the values, ranging from $1*10^{-5}$ to $1*10^{-3}$ m/s, are distinctively lower than for the majority of sites.

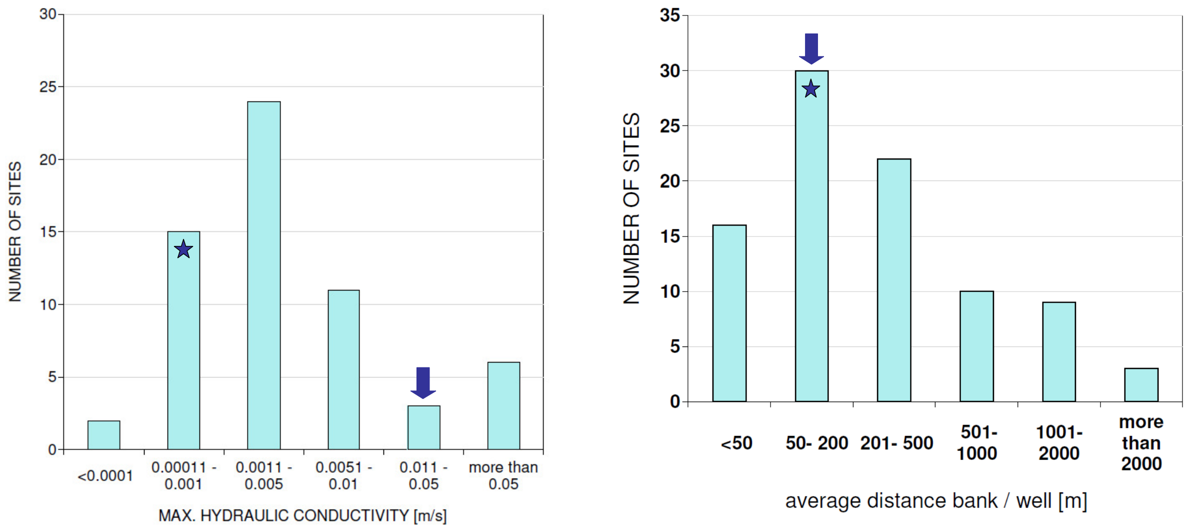

Figure 2: Maximum hydraulic conductivities (left) and average distances between bank and well (right) of 61 and 92 AR / BF sites worldwide. Berlin's main water works and the Lyon site typically fall into the star- and arrow-labelled categories, respectively.

For 92 field sites information on the distance from the bank to the abstraction well was available. Where distances were given as range, the arithmetic mean was calculated. Out of the 92 field sites, 17% abstract water after <50 m subsurface passage, 56% between 50 m and 200 m and 24% kept at least a distance of 1000 m to the bank. Especially those field sites located in the Netherlands, Poland, Slovakia and Finland operate BF systems with a long travel distance. In the Netherlands, 81% of the sites keep at least a distance of 300 m. The Krajkowo site in Poland is located 565 m from the bank and in Slovakia the distances range from 250 m to 1750 m. The lowest travel distances were found for Hungary and the USA: At the Hungarian sites along the river Danube the wells are located between 30 m and 101 m from the bank and the travel distances in the US range between 23 and 131 m. The sites in Berlin lie well below the median of 175 m with minimum travel distances ranging from 30 to 80 m.

Figure 4, shows the relationship between hydraulic conductivity and distance bank / well for those sites for which both parameters are available. Obviously, high hydraulic conductivities are not necessarily linked to large distances between bank and well. This would have been expected, if the sites were designed primarily to serve water quality improvements. On the contrary, other factors like regional geology, maximum bank filtration share and space availability are likely to control the distance between bank and well at existing BF / AR sites.

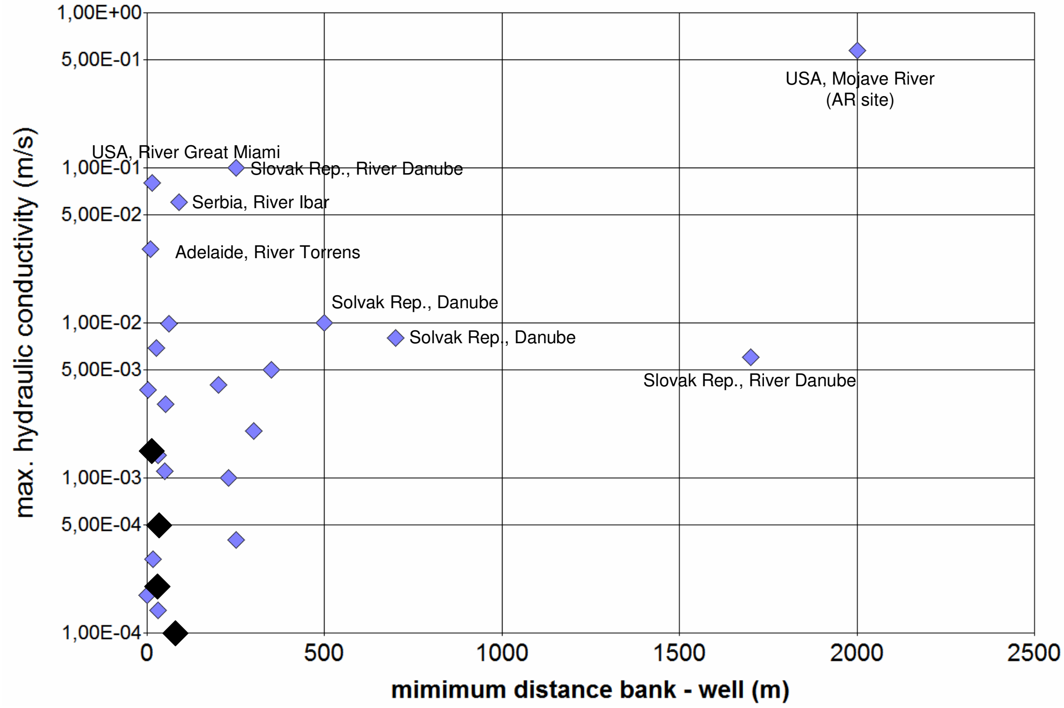

Figure 3: Maximum hydraulic conductivities and minimum distance between bank and well. The Berlin waterworks are marked with large, black diamonds.

Geological information is given for 64 sites of which the majority is situated in unconsolidated aquifers (92 %). Only 4 sites are located in hard rock or limestone aquifers (India – Ayyar basin, US – Texas, US – Pennsylvania and Namibia – Windhoek).

Flow times are available for 39 field sites and range from 1 to 4,000 days, with most flow times ranging from 10 to 50 days. High flow times are especially found at the river Danube with up to 700 days in Slovakia and up to 1000 days in Hungary. No significant difference was observed between lake and river sites.

2.2 Statistical analysis of pre- and post-treatment applied in combination with BF/AR systems

In order to obtain an overview on typical pre- and post-treatment techniques that may be combined with BF & AR 46 water utilities with 56 treatment chains were analysed (see Appendix B). Further information on possible pre- or post-treatment techniques can be obtained from standard literature (e.g. MWH, 2005). The large majority of analyzed sites is located in Germany and Europe due to better data availability (Figure 4). An overview of the treatment steps implemented at the investigated sites is given in Table 2 and Table 3.

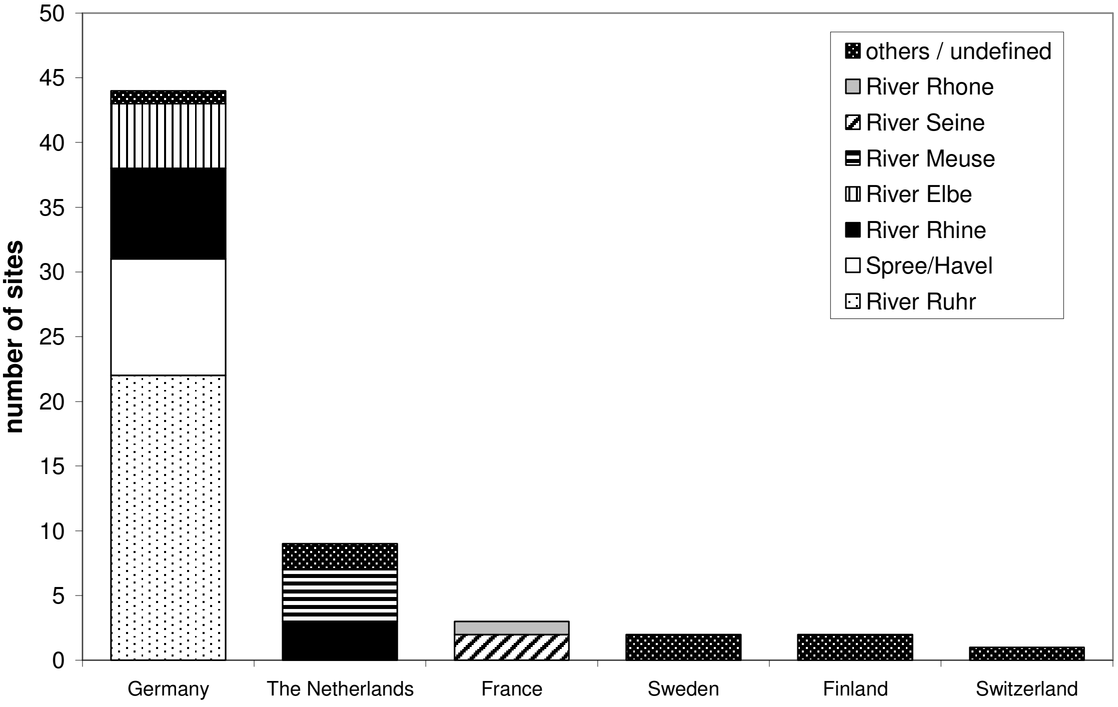

Figure 4 Total number of analyzed treatment chains grouped by countries and surface water used.

Table 4 gives a summary of the occurrence of the identified treatment steps in the investigated BF & AR systems. In order to avoid duplicates, combined BF/AR schemes have been assigned to the group of AR schemes.

For BF systems aeration (or oxidation) combined with filtration are applied in nearly all of the investigated facilities (94 %) as post-treatment. This is due to the fact that, as for Berlin, substances prevalent during anoxic subsurface passage like iron, manganese, ammonium and H_2S need to be removed. In 31 % of the sites advanced post-treatment (granular activated carbon) is also carried out. This is typical for the sites situated along the rivers Elbe, Rhine and Ruhr, which receive their waters from heavily industrialized areas. Disinfection is standard in about half of the facilities investigated (56 %). In six of the other seven facilities (all located in Berlin) it is only applied on demand. This result is certainly biased by the fact that the majority of treatment chains investigated is located in Central Europe and would look different in the United States, where chlorination is mandatory.

Table 2: Post-treatment of 16 water works using BF as treatment step (the order of columns does not necessarily represent the order of treatment steps).

site		**bank filtration**	**aeration**	**ozonation**	**deacidification**	**flocculation**	**sedimentation**	**KMnO4**	**sand & gravel filter**	**GAC**	**neutralisation**	**disinfection**	
												UV	**ClO_2 /Cl_2**
Dresden - Tolkewitz	D	X	X		X	X		X	X	X			X
Meißen	D	X	X		X				X	X			X
Torgau	D	X	X		X	X	X	X	X				X
Elsnig	D	X	X		X				X	X			X
Essen-Überruhr	D	X	X	X		X			X	X		X	
Düsseldorf	D	X		X	X					X			X
Waterwork Stiepel	D	X			X	X			X			(X)	X
Berlin-Tegel	D	X	X						X				(X)
Berlin Friedrichshagen	D	X	X						X				(X)
Berlin Stolpe	D	X	X						X				(X)
Berlin Tiefwerder	D	X	X						X				(X)
Berlin Beelitzhof	D	X	X						X				(X)
Berlin Kladow	D	X	X						X				(X)
Kuopio Vesi - Itkonniemi	FIN	X	X		X	X	X	X	X		X		X
Kuopio Vesi - Jänneniemi	FIN	X	X		X				X		X	X	X
River Rhine, # 3	NL	X	X						X				

(x): on demand

A typical pre-treatment for AR systems is the removal of particulate matter either with flocculation / sedimentation or sand filtration. No pre-treatment was only found at two sites, one of which operates infiltration meadows (Berlin-Stolpe). The reason for the removal of particulate matter prior to infiltration is clearly the enhancement of the infiltration capacity which will in addition benefit the generation of oxic conditions in the subsurface. In nearly half of the investigated cases, additionally activated carbon is applied prior to infiltration in order to enhance the removal of organic trace compounds.

The relative frequency of aeration / filtration as post-treatment is lower for AR schemes (35 %) than for BF schemes (94 %). This indicates that the redox conditions in the aquifer may generally be less anoxic (as also shown for Berlin Tegel during the NASRI project). Advanced post-treatment is carried out with activated carbon and ozonation, but usually only at those sites which had not applied it as pre-treatment.

Table 3: Pre-and post-treatment of 40 water works using AR or AR & BF as treatment step (the order of columns does not necessarily represent the order of treatment steps).

		pre-treatment											**post-treatment**									
site		**microsieve / screening**	**flocculation**	**sedimentation**	**aeration**	**ozonation**	**sand / gravel filtration**	**KMnO$_4$**	**chlorination**	**disinfection**	**GAC / PAC**	**subsurface passage**	**aeration**	**deacidification**	**flocculation**	**ozonation**	**sand & gravel filter**	**GAC / PAC**	**neutralisation**	**softening**	**disinfection**	
																					UV	**ClO$_2$ / Cl$_2$**
Dresden-Hosterwitz	D		X				X					X	X	X				X				X
Mülheim Styum-Ost	D											X		X		X	X	X				X
Mülheim Styrum West	D		X			X	X				X	X		X								X
Mülheim Dohne	D		X			X	X				X	X		X								X
Mülheim Essen Kettwig	D		X			X	X				X	X		X								X
Halingen/Frödenberg	D	X		X			X				X*	2X										
Hengsen	D			X			X					X		X								X
Hengsen	D			X			X					X		X								X
Villingst	D			X			X				(X)	X		X								X
Ergste	D			X			X				(X)	X		X								X
Ergste	D			X			X				(X)	X		X								X
Westhofen1	D			X	X		X				(X)	2x	X	X								X
Westhofen2	D			X	X		X				(X)	2x		X								X
Witten	D		X	X							(X)	X		X	(X)	X	X					X
Hagen-Hengstey	D		X		X		X					X										X
Essen-Überruhr	D		X			X	X	(X)			X	X							X		X	
Essen-Horst	D			X	X		X	(X)			(X)	X							X		X	
Essen-Horst	D				X							X							X		X	
Water work Essen	D	X			(X)		X	(X)			(X)	X		X							(X)	
Water work Essen	D				X							X		X							(X)	X
Wasserwerk Haltern	D		X	X							X	X		X			X				(X)	
Berlin-Tegel	D	X										X	X				X				(X)	
Berlin-Stolpe	D											X	X				X				(X)	
Berlin-Spandau	D		X									X	X				X				(X)	
Leiduin Water plant	NL		X				X					X	X			X	X	X		X		
Monster	NL	X	X				X					X	X				X	X		X		
Scheveningen	NL	X	X				X					X	X				X	X		X		
Katwijk	NL	X	X				X					X	X				X	X		X		
Flins Aubergenville	F				X						(X)	X						(X)				
Flins Aubergenville	F				X						(X)	X						(X)				
Lyon Crèpieux-Charmy	F			X								X										X
Basel Langen-Erlen	CH						X					X										X
River Rhine, #1	NL		X	X			X		X	X		X	X				X	X				X
Westland, #4	NL	X										X	X				X					
River Meuse, #6	NL		X	X			X		X	X		X	X				X	X				
Lake Haringvliet, #8	NL											X				X	X	X				X
Wiesbaden-Schierstein	D		X	X	X		X				X	X	X				X	X				X
Wiesbaden-Schierstein	D		X	X	X		X				X	X	X				X	X				X
Dösebacka	S			X								X									X	
Dösebacka	S			X								X			X		X				X	

(x): on demand

Table 4 below displays the pre- and post treatment steps for BF and AR schemes as published by the investigated waterworks. It has further to be noted that several treatment methods fulfil more than one purpose (e.g. ozonation breaks down complex organic matter and acts as potent disinfectant). Methods are herein grouped according to their main purpose but they may have more than one benefit. Minor treatment steps such as sand traps or mixing chambers were not included in the analysis.

Table 4: Observed occurrence of different pre- and post-treatment steps in BF and AR systems.

treatment	bank filtration (BF) (n = 16)	aquifer recharge (AR) (n = 40)		total (n = 56)
	post-treatment	pre-treatment	post-treatment	
removal of particulate matter				
screening	-	2	-	2
microsieving	-	4	-	4
coagulation/flocculation	5	17	2	23**
sedimentation	3	18	-	21
sand filtration (SSF, RSF, multi-layer, dry filtration)	15	28*	17	49***
removal of organic micropollutants				
activated carbon (PAC / GAC)	5 (-/5)	16 (10/6)	13 (8/5)	32****
ozonation	2	4	4	10
$KMnO_4$ (algicide) on demand	2	3	-	5
removal of inorganic trace elements				
aeration/stripping (Mn, Fe, NH_4, H_2S, CH_4, CO_2)	15	11	14	34*****
disinfection				
chlorination	7	2	23	32
UV treatment	2	-	3	5
not specified	-	-	2	2
on demand	6	-	6	13
no disinfection	1	-	7	8
pH/hardness adjustment (preservation of the network)				
deacidification/neutralisation	8	-	19	27
softening (ion exchange)	-	-	4	4

* 3 of these treatment chains use subsurface passage (AR) as “pre-treatment”
** 1 AR site gives flocculation in pre-and post-treatment
*** 11 AR sites give filtration steps as pre- and post-treatment
**** 2 AR sites give AC treatment as pre- and post-treatment
***** 3 AR sites give aeration as pre- and post-treatment

2.3 Overview of the Legal Framework Regulating Managed Aquifer Recharge and Existing Technical Guidelines

2.3.1 Introduction

Aquifer recharge facilities are subject to a wide range of regulations, including groundwater protection, water rights or water use permits and drinking water regulations. Since the injection of water into an aquifer can affect the quality of the groundwater, its preservation is the focus of most of the available regulation. Legal requirements generally rely on existing non-specific regulation, except for the USA, where artificial groundwater recharge via well injection is regulated by the *Underground Injection Control Program* (UIC). With the Water Framework Directive to be operational by 2012, EU countries will have to implement regulations for the authorisation of artificial recharge (WFD).

Professional associations in the USA, Australia and Germany provide several technical guidelines and recommendations. An overview of their content is presented in the following chapters. Today, the source water quality that must be achieved prior to injection is disputed among experts. A summary of the current discussions is presented in chapter 2.3.3.

2.3.2 Overview of the different existing approaches on aquifer recharge regulation

The USA is the only country identified in this study, which applies specific regulations for the injection of water into an aquifer. The regulation for groundwater recharge is given to the EPA through the *Underground Injection Control Program* (UIC) of the safe drinking water act. The primary purpose of the program is to protect aquifers that are used or could be used for drinking water supply. The UIC program regulates the quantity and quality of injected water as well as geological constraints for the injected fluids. In addition, regulations for reclaimed water, indirect potable reuse and wellhead protection affect groundwater injection programs (Bloetscher *et al.*, 2004). The number of groundwater recharge wells included in the UIC program was estimated to reach 1.185 in the different US states (U.S. EPA, 1999). Some states implement the program under their own authorities. Aside from the UIC program, additional local state laws may govern groundwater protection. Authorisation of aquifer recharge wells are either given by rule (e.g. Oklahoma, Texas) or by individual permits (e.g. Arizona, Florida, Nevada, Oregon). Source water for aquifer recharge and ASR wells is required by most regulatory agencies to meet primary[1] and secondary[2] drinking water standards in order to prevent degradation of ambient ground water quality (USEPA, 2009). In Florida, for instance, source water concentration of a particular parameter may be as high as natural background levels in groundwater if the concentration is greater than the maximum contaminant level (Maliva *et al.*, 2006). In the case of indirect potable reuse, where treated wastewater is injected into subsurface, other unregulated compounds may require specific attention. In the “Draft Groundwater Recharge Reuse Regulations[3]” the California Department of Health Services’ includes monitoring requirements for pharmaceuticals, endocrine disrupting chemicals and personal care products.

For the specific case of bank filtration, the U.S. regulations focus on the removal of *Cryptosporidium* and other microorganisms (EPA’s Long-Term 2 Enhanced Surface Water Treatment rule, LT2ESWTR). The design criteria specified in LT2ESWTR are based on conservative estimates drawn from colloid filtration theory and an analysis of microbial monitoring data from existing bank filtration sites (Tufenkji *et al.*, 2002). The EPA proposes that horizontal and vertical wells drilled into unconsolidated, granular aquifers would be suitable for 0.5 log (68%) removal credit or 1.0 log (90%) removal credit when located at least 7.6 or 15.2 m from the river/lake respectively (Tufenkji *et al.,* 2002).

In Europe, the Water Framework Directive obliges member states to establish by the end of 2009 a programme of measures to achieve environmental objectives (e.g. abstraction control, measures to prevent or control pollution) that would be operational by the end of 2012. One measure mentioned is aquifer recharge. Directive 2000/60/EC sets out the

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[1] EPA's Primary Drinking Water Standards quantify levels of contaminants that affect health. These standards or Maximum Contaminant Levels (MCL) are enforced by the EPA on public drinking water systems. The list is available at http://www.epa.gov/safewater/contaminants/index.html#listmcl

[2] Secondary drinking water standards deal with aesthetic qualities of water like taste, color, and smell while health advisories levels are guidelines for contaminants that may be regulated in the future by the EPA. The list is available at http://www.epa.gov/safewater/contaminants/index.html#listsec

[3] Available at http://www.cdph.ca.gov/healthinfo/environhealth/water/Pages/Waterrecycling.aspx

requirement of controls including a requirement for prior authorisation of artificial recharge or augmentation of bodies of groundwater, provided that the use of the source does not compromise the achievement of the environmental objectives established for the source or the recharged body of groundwater.

Among European countries, the Netherlands, where artificial recharge accounts for 22% percent of the water demand (Murray *et al.,* 2007), have a specific regulation for groundwater recharge. The *Infiltration Act* (IB, 1993) specifies the quality and quantity that can be infiltrated (Kooistra *et al.*, 2003). The permit required for the infiltration of water shall minimise groundwater pollution.

The state of South Australia has established in 2004 the Code of Practice for Aquifer Storage and Recovery (ASR Code). The ASR Code outlines the requirements of the Environment Protection Authority for the storage and recovery of water in aquifers. By following these requirements, the operator should be able to comply with the regulations of South Australia. The ASR Code aims at protecting the quality of groundwater quality and dependent ecosystems. Artificial recharge should improve or at least maintain groundwater quality.

2.3.3 List of existing technical guidelines

The American Society of Civil Engineers (ASCE) published *Standard Guidelines for Artificial Recharge of Ground Water*[4] (ASCE, 2001). They specify the steps necessary of planning a *MAR* project including design, construction, operation, maintenance, field investigations and testing procedures. Additionally, they list the economic, environmental and legal considerations (Dillon, 2003).

In Germany, recommendations for drinking water production via bank filtration and artificial groundwater recharge is given in the Technical Rules and Guidelines published by the German Technical and Scientific Association for Gas and Water (DVGW) and the German Institute for Standardization (DIN). In detail, these are the *Technical Rule W251 of the Applicability of River Water as Raw Material for Drinking Water Supply* (DVGW, 1996) and the *W126 for Planning, Construction and Operation of Groundwater Recharge Systems for Drinking Water Abstraction* (DVGW, 2007). The *DIN2000* describes the standard for drinking water quality requirements and for the planning, construction and operation of drinking water facilities. For bank filtration, surface water contamination is to be minimised to ensure that bank filtrate meets drinking water requirements after subsurface passage (DIN, 2000). The minimum quality standards are listed and commented in the *Technical Rule W251* (DVGW, 1996). There are no threshold values for the source water used in artificial recharge, however, groundwater quantity and quality shall not deteriorate (DVGW, 2007).

Australia has recently drafted detailed *Guidelines for Managed Aquifer Recharge (EPHC, 2008)* and a Technical Guidance for ASR (Dillon & Molloy, 2006). The aim of the *Guidelines for Managed Aquifer Recharge* is to define a consistent basis for Australia to protect human health and the environment when implementing managed aquifer recharge. This is achieved by assessing potential hazards/risks and by identifying preventive measures (Dillon *et al.*, 2009). The hazards addressed in the guidelines include pathogens, inorganic chemicals, salinity, nutrients, organic chemicals, turbidity/particulates, radionuclides as well as pressure or flow-related hazards. For each hazard, the guidelines outline sources or causes, the effect on public health and environment, management approaches including preventive measures, the proposed validation, verification and operational monitoring. They further list the acceptance criteria for the various stages of risk assessment (Dillon *et al.*, 2009). Since the recharged water may disturb the natural geochemical condition of the aquifer and thereby mobilise unwanted substances (e.g. arsenic), the MAR guidelines propose a scientific

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[4] Available at http://www.asce.org/bookstore/book.cfm?book=4056

approach taking into account the interaction of recharged water and aquifer. Thus, Dillon *et al.* (2009) disagree that treating water to drinking standards prior to recharge will protect the aquifer and recovered water.

The Technical Guidance for ASR (Dillon & Molloy, 2006) provides guidance for projects planning subsurface water storage meant for potable reuse or ecological benefit (e.g. wetland maintenance) as well as agricultural or industrial use.

Chapter 3
Bank Filtration and Aquifer Recharge Performance for Substance Removal

3.1 Introduction

Bank filtration has been in use since the 19th century for its capacity to remove pathogens from surface water. Today, the city of Berlin provides drinking water via bank filtration to 3.4 million inhabitants without any further disinfection (SenStadtUm 2008). Pathogens, suspended solids and algal toxins are undesired substances, for which subsurface passage can achieve high removal rates. However, there are other substances of growing concern (e.g. EDCs and specific pharmaceuticals), which may show low removal rates or may even be persistent.

This chapter presents a synthesis of existing information on the performance of bank filtration and aquifer recharge (via pond infiltration) to remove substances from surface water. This analysis is based on published data and the data acquired during the NASRI project.

At this stage, an important advantage of BF & AR needs to be mentioned, compared to surface water supply only: During subsurface passage the temperature of the surface water is equilibrated to reach annual the mean after sufficient travel time. The necessary travel time is determined by the dispersion coefficient and the maximal amplitude of seasonal temperature variations. In Berlin, during the NASRI project, surface water temperatures varied between 0°C in winter and 26°C in summer. As given in Figure 5 the standard deviations of the water temperatures measured in the observation wells decreased from more than 8°C after only a few days of travel time down to 1-3 °C after 100 d travel time.

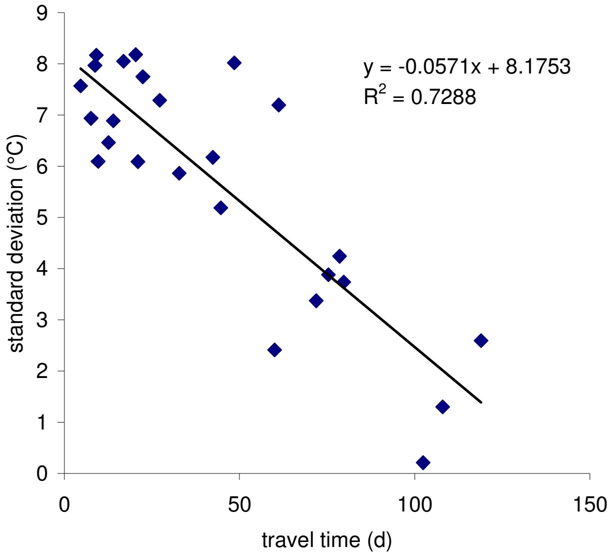

Figure 5: Standard deviations of water temperature vs. travel times for observation wells with > 90 % bank filtrate at Lake Tegel and Lake Wannsee transsects, investigated during the NASRI project.

Concentrations of dissolved or suspended substances decrease or are “attenuated” during infiltration by filtration (or straining), die-off, (bio-)degradation (mineralization or transformation), inactivation, decay, adsorption, precipitation and mixing with ambient groundwater (Figure). In theory, mixing with ambient groundwater should not be regarded as a removal process. For the NASRI data we will therefore use the term “removal” only for the those processes that decrease the substances’ concentrations during infiltration excluding dilution e.g. by ambient groundwater.

In some publications “removal” is referred to as overall reduction of concentration and the share of ambient groundwater not given. On the other hand, an analysis of the literature compiled by Schmidt (2007) for the database on trace organics’ removal in BF & AR systems showed that for 73 % of the removal rates given the share of bank filtrate or recharged groundwater was 80 % or higher (n=699). A lower share was only given in 6 % of the cases, for 21 % no information was available.

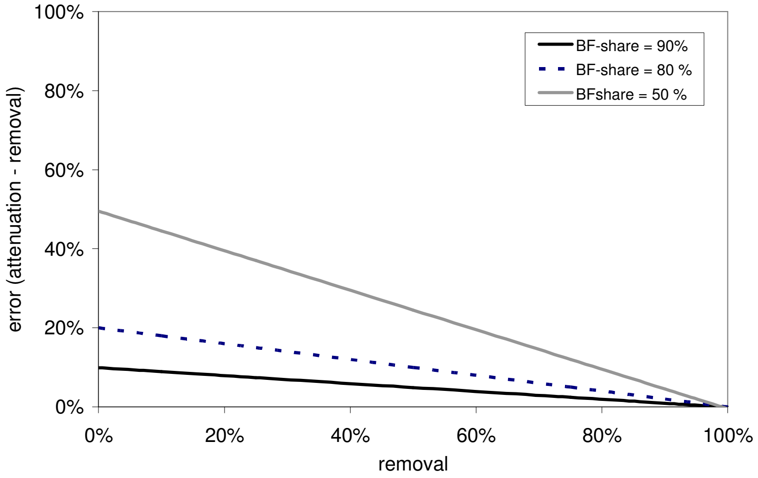

Figure 6: Difference between attenuation and removal for different shares of bank filtrate and removal rates (concentration in surface water = 100 * concentration in ambient groundwater or more).

Figure 6 shows the impact of the share of bank filtrate and removal rate on the overall attenuation rate on the basis of simple mixing calculations. These considerations show that as long as the share of bank filtrate is 80 % or higher, the difference between removal and attenuation will not exceed 20 %, which is in the order of magnitude of analytical and sampling errors. Especially for high removal rates the difference is negligible. If on the basis of available data a differentiation is not possible, the terms removal and attenuation will therefore be used synonymously in the following chapters.

As in all aquifer systems the water quality of bank filtrate is significantly affected by reduction/oxidation (redox) processes (McMahon and Chapelle 2008). The exact definitions for the different redox zones are often not consistent between the different publications. The analysis of the NASRI data followed the definition given by McMahon and Chapelle (2008), in which the redox state is determined using the species oxygen, nitrate, manganese (II), iron (II) and sulfate (Table 5). As most publications do not follow this detailed differentiation, the oxic and sub-oxic zone are in the following combined as “oxic” conditions, whereas the anoxic and anaerobic zone merge to form the “anoxic” zone.

Table 5: Applied classification of redox zones for the interpretation of NASRI data (modified from Wiese et al. 2009).

Redox zone	**Dominant redox process**	**Redox species**					**Simplified classification**
		O_2 [mg/l]	NO_3^- [mg/l]	Mn^{++} [mg/l]	Fe^{++} [mg/l]	SO_4^{2-} [mg/L]	
Oxic zone	O_2-reduction	≥ 0.5	any	< 0.05	< 0.1	any	**“oxic”**
Suboxic zone		< 0.5	any	< 0.05	< 0.1	any	
Anoxic zone	Mn- and Fe-reduction	< 0.5	< 0.5	Mn ≥ 0.05 or Fe ≥ 0.1		any	**“anoxic”**
Anaerobic zone	Methano-genesis	< 0.5	< 0.5	any	≥ 0.1	< 0.5	

Despite these possible artefacts in the data interpretation, it is proposed to classify the substances in three categories:

1. Substance groups / substances for which a high removal (> 90%) is achieved, provided optimal conditions are met
2. Substance groups, whose substances show highly variable removal in BF&AR systems (typically 25 to 75 % overall removal)
3. Substance groups / substances which are persistent or may become (re-)mobilised and to which special attention should be paid

The classification will provide an update on the common understanding of the performance of subsurface passage for major water pollutants and undesired compounds integrating current knowledge on emerging contaminants. The following chapters will discuss each of these categories.

3.2 Substances or substance groups, for which high removal (> 90 %) is possible

The primary purpose for the use of subsurface passage in water treatment was to obtain hygienically safe drinking water at a time when chemical disinfection (e.g. chlorination) had not been in use. It is broadly accepted that the benefits of bank filtration include the elimination of suspended fines and pathogens (Weiss *et al.,* 2002, Schubert, 2006; Partinoudi & Collins, 2007). The current study confirms that high removal efficiency can be achieved for the above-mentioned parameters. Although bank filtration is usually reliable in removing these substances, occasional system failures may occur. Therefore, the achievable maximum removal rate should be examined site-specifically.

In addition to pathogens and suspended solid removal, recent studies demonstrated the capacity of subsurface passage to effectively remove some algal toxins (microcystins), provided certain requirements are met as well as nitrate and disinfection by-products under sub-oxic, anoxic or anaerobic conditions.

3.2.1 Suspended solids

The public health and environmental risks associated with turbidity in relation to managed aquifer recharge include (EPHC, 2008):

i) exceeding drinking water guidelines for turbidity

ii) reduced disinfection performance leading to increased risk from microbial pathogens

iii) increased risk of transporting a range of contaminants that can adsorb to particles such as heavy metals, phosphorus, various organics and microbial pathogens

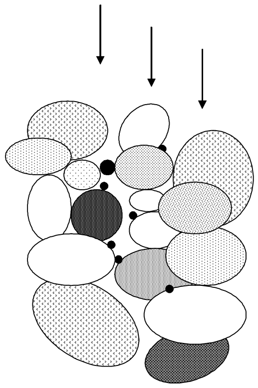

Figure 7: Schematic illustration of particles straining during subsurface passage.

Subsurface passage removes suspended solids from source water by physical retention when water infiltrates into the subsurface (Figure 7). This is mediated by straining of larger particles but also by physical processes such as Van-der-Waals forces retaining smaller particles.

High loads of suspended solids in source water may enhance clogging, especially in infiltration ponds, rendering pre-treatment (e.g. micro-sieve, coagulation/filtration) necessary to maintain sufficient infiltration rates (see chapter 2.2).

Despite the importance of this parameter, few studies have specifically addressed the removal of suspended solids during bank filtration. The collected information from the literature is compiled in Table 6 comparing turbidity in source water and at the abstraction well. In all studied cases, the minimum removal rate observed remains high with 92% (Kalama, Washington, USA). The highest removal rate was observed for Terre Haute IN, USA with 99.9%. In all above-mentioned studies, drinking water wells were situated at least 27 m from the surface water.

Table 6: Observed removal of suspended solids at selected bank filtration sites.

	source water (average NTU)	**bank filtrate (average NTU)**	**calculated mean removal**	**comments**
Jeffersonville, IN, USA (Weiss *et al.*, 2005)	60.1	0.1	99.8%	Well #2 located 30 m from the river
Terre Haute, IN (Weiss *et al,* 2005)	190.3	0.1	99.9%	Collector well located 27 m from the Wabash River
Parkville, MO, USA (Weiss *et al.*, 2005)	78.6	0.5	99.4%	Well located 37 m from the river
Ohio River, USA (Wang, 2002)	45	0.1	99.8%	RBF 30 meters from the river
Kalama, Washington, USA (Mikels, 1992)	5	0.4	92.0%	Max values
Nainital Lake, India (Dash, 2008)	7.1	0.25	96.5%	Value during monsoon; wells up to 84 m from the lake
Sidfa RBF site, Egypt (Shamrukh & Abdel-Wahab, 2008)	6.7	0.3	95.5%	Well located 30 m from the Nile river
Lyon, France (Veolia Eau)	19	0.22	98.8 %	data from 2007, personal communication Veolia Eau

The US Environmental Protection Agency (EPA) recommends turbidity to be in average below 1 NTU and always to be less than 5 NTU (US-EPA 1998). This threshold value is met in all mentioned studies. Since high turbidity is an indicator for low filtration quality, this parameter can be used to identify the efficiency and performance of the filtration process.

Summary, suspended solids

Observed removal of suspended solids at BF & AR sites ranges from 92 % to 99.9 % with an average of 97.6 %. In all documented cases the threshold values for drinking water of 1 NTU in average is met. Monitoring turbidity can provide valuable information on the performance of the filtration process over time (e.g. regarding removal of algae or pathogens).

3.2.2 Pathogens

The presence of pathogenic bacteria, viruses or protozoa in drinking water poses a major threat to human health as they are the primary cause for water-borne diseases world wide (WHO, 2006). Due to potentially high concentrations in source water and low infectious doses, removal rates of several orders of magnitude are postulated necessary in drinking water treatment (e.g. US-EPA, 1998).

During the subsurface passage of pathogens, their numbers are reduced by a combination of processes including straining, inactivation and adsorption to the aquifer matrix (Schijven *et al*; 2002).

Table 7 provides the removal efficiencies observed at studied bank filtration sites. The data shows a broad range of possible values:

- 3.2 to 6-$\log_{10}$ for non-spore-forming bacteria,
- 0.4 to >4.9-$\log_{10}$ removal for spore-forming bacteria,
- 2.1 to 8-$\log_{10}$ for viruses and
- 0.9 to 2.5- $\log_{10}$ for protozoan (oo)cysts.

Since field conditions differ and published data is often fragmentary, it is not possible with the currently available data to correlate removal rates directly with travel distances or flow time. It is expected that other factors would also strongly influence the removal of pathogens. These factors comprise variations in pore water velocity, the degree of groundwater dilution, solution chemistry, surface characteristics of aquifer media and inherent heterogeneities in the microbial population (Gollnitz *et al.*, 1997). Temperature may additionally be of high importance for virus inactivation with higher inactivation correlating to higher temperatures (Schijven *et al.,* 2002).

Table 7: Removal of pathogens or indicator organisms by bank filtration.

pathogen or indicator	**travel time in d (distance in m to bank)**	**site**	**removal efficiency** **(measured or estimated)**	**reference**
non-spore-forming BACTERIA				
Total coliforms	11-19 d (84 m)	RBF (Lake Naini)	5-$\log_{10}$	Dash *et al.* (2008)
Total coliforms	n.a. (37 m)	RBF (Missouri River)	5.5 to 6.1-$\log_{10}$	Weiss *et al.* (2005)
Total coliforms	15 d (30 m) 63 d (25 m)	2 RBF sites (River Meuse and River Rhine)	>5- $\log_{10}$	Schijven *et al.* (2002)
Faecal coliforms	11-19 d (84 m)	RBF (Lake Naini)	4-$\log_{10}$	Dash *et al.* (2008)
Thermotolerant (faecal) coliforms	n.a. (<15m)	RBF River Meuse, sandy gravel aquifer	4-$\log_{10}$	Medema *et al.* (2000)
Faecal streptocooci	15 d (30 m)	RBF River Rhine	>3.2- $\log_{10}$	Schijven *et al.* (2002)
Faecal streptocooci	63 d (25 m)	RBF River Meuse	>3.5- $\log_{10}$	Schijven *et al.* (2002)
spore-forming BACTERIA				
Bacillus sp. (aerobic sporeformer)	n.a. (27-177 m)	3 RBF sites (Ohio, Wabash & Missouri River)	0.8-$\log_{10}$ to >3.1-$\log_{10}$	Weiss *et al.* (2005)
Clostridium sp. (anaerobic sporeformer)	n.a. (27-177 m)	3 RBF sites (Ohio, Wabash & Missouri River)	0.4- $\log_{10}$ to >4.9- $\log_{10}$	Weiss *et al.* (2005)
Spores of sulphite-reducing clostridia	n.a. (13 m) 18 d (25 m)	RBF River Meuse,	3.3- $\log_{10}$ 3.9- $\log_{10}$	Medema *et al.* (2000)

pathogen or indicator	**travel time in d (distance in m to bank)**	**site**	**removal efficiency** **(measured or estimated)**	**reference**
(SSRC)	43 d (150 m)	sandy gravel aquifer	5.0- $\log_{10}$	
SSRC	63 d (25 m)	RBF River Rhine	3.1- $\log_{10}$	Schijven *et al.* (2002)
SSRC	15 d (30 m)	RBF River Meuse	3.6- $\log_{10}$	Schijven *et al.* (2002)
VIRUSES				
bacteriophage (male-specific)	n.a. (13 m) 18 d (25 m)	RBF River Meuse, sandy gravel aquifer	3.9- $\log_{10}$ 6.0- $\log_{10}$	Medema *et al.* (2000)
bacteriophage (male-specific)	n.a. (37 to 177 m)	3 RBF sites (Ohio, Wabash & Missouri River)	2.1- $\log_{10}$to >2.6- $\log_{10}$	Weiss *et al.* (2005)
bacteriophage (male-specific)	15 d (30 m)	RBF River Rhine	6.2- $\log_{10}$	Schijven *et al.* (2002)
bacteriophage (somatic)	n.a. (13 m) 18 d (25 m) 43 d (150 m)	RBF River Meuse, sandy gravel aquifer	3.8- $\log_{10}$ 5.1- $\log_{10}$ 7.8- $\log_{10}$	Medema *et al.* (2000)
bacteriophage (somatic)	n.a. (37 to 177 m)	3 RBF sites (Ohio, Wabash & Missouri River)	4.0- $\log_{10}$ to >4.4- $\log_{10}$	Weiss *et al.* (2005)
bacteriophage (somatic)	n.a. (1 m) n.a. (4 m)	RBF (Yamuna River)	3.3- $\log_{10}$ 4.6- $\log_{10}$	Sprenger *et al.,* 2009
bacteriophages MS2 & PRD1 (male-specific)	n.a. (2.4 m), n.a. (30 m)	dune infiltration	3-$\log_{10}$ 8- $\log_{10}$	Schijven *et al.* (1999)
adenovirus	n.a. (50 m)	RBF (Yamuna river)	>4- $\log_{10}$	Sprenger *et al.* (2009)
norovirus	n.a. (50 m)	RBF (Yamuna river)	>4- $\log_{10}$	Sprenger *et al.* (2009)
reovirus	n.a. (13 m)	RBF Meuse River, sandy gravel aquifer	3.7- $\log_{10}$	Medema *et al.* (2000)
PROTOZOA				
Giardia cysts	n.a.	RBF City of Kearney	2- $\log_{10}$	Schijven *et al.* (2002)
Giardia cysts	n.a.	RBF Sonoma County	2.5 - $\log_{10}$	Schijven *et al.* (2002)
Giardia cysts	n.a. (27-177 m)	3 RBF sites (Ohio, Wabash & Missouri River)	1.3 - $\log_{10}$ to >1.9- $\log_{10}$	Weiss *et al.* (2005)
Cryptosporidium oocysts	n.a. (27-177 m)	3 RBF sites (Ohio, Wabash &	0.9 - $\log_{10}$to >1.5- $\log_{10}$	Weiss *et al.* (2005)

pathogen or indicator	travel time in d (distance in m to bank)	site	removal efficiency (measured or estimated)	reference
		Missouri River)		

n.a.: no information available

The highest removal rates were observed during canal infiltration into fine dune sand yielding an 8-log removal for viruses within 30 m (25 d flow time). The highest removal rates for spore- and non-spore-forming bacteria were >4.9-log and 6.1-log respectively and have been observed at BF sites in the US characterised by fine to coarse sand and gravel after 37 m (Weiss *et al*., 2002, Weiss *et al*., 2005). In many cases higher removal may have been possible but could not be detected due to limited source water concentrations.

Schijven *et al.* (2002) suggest that short path lengths, high heterogeneity, coarse matrices, high gradients and accompanying high flow velocities decrease the efficiency of pathogens removal. This is confirmed by a reported case of *E. coli* breakthrough at a BF site near the river Rhine in Germany as described in Eckert *et al*., (2008). In this particular case, breakthrough occurred during a flood event, where travel time and distances were shortened by an increased hydraulic gradient (without flooding the well head). The removal of the biologically-active layer by streambed scouring was another reason suggested to have caused the breakthrough.

Finally it can be stated that subsurface passage as used in BF and AR may represent an important – if not the main – barrier for pathogens during drinking water treatment. In West Berlin, for example, drinking water has been produced for decades mainly by BF and AR without further disinfection.

A sufficient removal capacity for pathogens is, however, connected to certain requirements that may not be possible to achieve at every site and at every time (e.g. during flooding). Thorough initial assessment and continuous monitoring of indicator parameters like turbidity are therefore recommended. Finally, one crucial factor is the contamination level of the infiltrating water and therefore the protection of surface water is relevant when using BF or AR (Dimkic, 2008).

Summary pathogens

Subsurface passage can present an important or even main barrier towards pathogens for drinking water production. Observed removal at field sites ranges from

- from 2.1 to 8-$\log_{10}$ for viruses
- from 3.2 to 6-$\log_{10}$ for non-spore forming bacteria
- from 0.4 to > 4.9-$\log_{10}$ for spore-forming bacteria
- from 0.9 to 2.5-$\log_{10}$ for protozoan (oo)cysts

Crucial factors for the functioning of this barrier towards pathogens are source water protection, sufficient travel times (> 50d), a fine and homogeneous aquifer matrix and low flow velocities.

3.2.3 Cyanobacterial (algal) toxins

Toxins produced by cyanobacteria (blue-green-algae) occur in surface waters worldwide (especially during so-called algal blooms) and need to be controlled in drinking water due to their liver- and neurotoxic effects. These cyanotoxins comprise a wide variety of different substances; however, most information on subsurface removal is available for microcystins

(MCYST), a hepatotoxic, cyclic peptide. This toxin occurs mainly within the cyanobacterial cells and may be released in significant amounts only by cell-lysis (death).

For mainly intra-cellular toxins, straining is the most important and efficient removal process (Chow *et al.*, 1999). Extra-cellular MCYST is usually well bio-degradable (half-lives lie in the range of hours, Grützmacher *et al.,* 2005), though in environments without adapted microbial community, lag phases of up to one week may occur before degradation commences (Welker *et al.,* 2001).

Field studies on cyanotoxin removal during subsurface passage are scarce and difficult to interpret due to the fact that source water concentrations vary by several orders of magnitude and due to limited information on the ratio between intra- and extra-cellular toxins. Table 8 gives an overview of observed removal at different field sites in Europe.

Table 8: Removal of cyanobacterial toxins (microcystins) by bank filtration.

analysed parameter	**flow time (d) (distance (m) to well)**	**site**	**removal efficiency** **(& remarks)**	**reference**
total MCYST, cyanobacterial cells	(100 m)	Jokionen water works, Lake Rehtijärvi, Finland	MCYST: > 83%, cyanob. cells: detected in BF, ($k_f < 10^{-1}$ m/s, 60 – 80 % BF share)	Lahti et al. (1998)
total MCYST, cyanobacterial cells	(400 m)	Forssa water works, Lake Kaukjärvi, Finland	MCYST: > 84%, cyanob. cells: detected in BF, ($k_f < 10^{-1}$ m/s, 70 % BF share)	Lahti et al. (1998)
total MCYST, cyanobacterial cells	n.a.	four Finnish waterworks using BF	MCYST: usually 90%, max. 99% (incl. post-treatment) cyanobacterial cells: mostly absent in wells	Lahti et al. (2001)
extracell. MCYST	n.a.	Riga, infiltration ponds	(1.5 µg/L MCYST-LR equivalent in raw water, initial total source water concen-tration and travel time not determined)	Eynard et al. (2000)
total MCYST	9 - 33 d (< 10 m)	Berlin, BF site Lake Wannsee	> 95.8%	NASRI (Chorus & Bartel, 2006)
extracell. Adda-containing substances (including MCYST)	14 d (4 m) 33 d (8 m)	Berlin, BF site Lake Wannsee	97% 99%	NASRI (Chorus & Bartel, 2006)

Though literature states limited elimination of cyanobacterial cells and total toxins, detailed investigations in Berlin during the NASRI project (Chorus & Bartel, 2006) showed high removal rates of > 95 % even for travel distances smaller than 10 m (flow times 0.5 – 1 month). The reported breakthrough of cells in Finland (Lahti *et al.*, 1998) has likely been caused by insufficient particle removal in coarse esker material (kf up to 10^{-1} m/s). In these cases, the cells will protect toxins from degradation thus enabling long persistence in the subsurface. In the other reported case of breakthrough (Riga, Eynard *et al.*, 2000) the authors themselves question the specificity of the analytical method. Due to this and the fact that total source water concentrations and travel times were not determined, these results were not taken further into account.

Given the high biodegradation rates (>1 d^{-1}) obtained from different laboratory and technical scale experiments during the NASRI project (Chorus & Bartel, 2006), Grützmacher *et al.* (2007) postulated the following pre-requisites for sufficient removal of MCYST to < 1 µg/L (WHO drinking water guideline) by bank filtration at source waters with frequent cyanobacterial blooms (i.e. adapted microbial population):

- total MCYST < 500 µg/L
- middle to fine grained sandy aquifer,
- aerobic conditions
- temperatures > 15 °C,
- residence times > 9 d.

For suboptimal conditions, residence times need to be much higher (> 70 d, see Figure 8).

Recent investigations have shown that for another frequently occurring cyanotoxin, cylindrospermopsin, biodegradation rates are similar to those determined for MCYST though their extra-cellular toxin share might be generally higher and the occurring lag phases remarkably longer (Klitzke *et al.*, 2009).

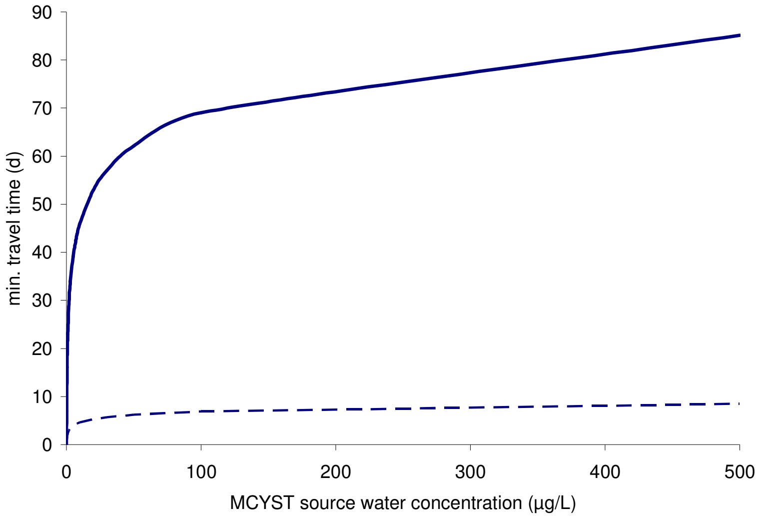

Figure 8: Minimum subsurface travel time for sufficient removal of MCYST depending on source water concentration of total MCYST for a) worst-case conditions (solid line), i.e. anoxic/anaerobic conditions, temperature < 15°C, and b) optimal conditions (dashed line). Adapted from Grützmacher et al. 2007)

Detailed monitoring for cyanobacterial toxins in raw water is usually not necessary. For some countries so-called alert level frameworks have been developed (e.g. South Africa, Du Preez & Van Baalen, 2006) recommending a stepwise approach in case of known or supposed cyanobacterial blooms, starting with simple methods for source water monitoring (e.g. fluorescent probe, Izydorczyka *et al.*, 2009).

Summary algal toxins
Predominantly cell-bound cyanobacterial toxins (microcystins) show up to 99 % removal during subsurface passage. Limitations can occur in case of high extra-cellular concentrations, coarse aquifer material, anaerobic conditions, low temperatures and short travel times.

3.2.4 Nitrate

Under optimal conditions subsurface passage can attenuate nitrate (NO_3) concentrations to a high degree (up to 99 %).

During infiltration, nitrate is stable in the oxic zone. However, after oxygen has been consumed, nitrate can be used as electron acceptors by bacteria. It may then be either converted into gaseous N_2 and N_2O or become reduced to NH_4^+. The different oxidation states of nitrogen and the corresponding standard redox potential are presented in Figure 9.

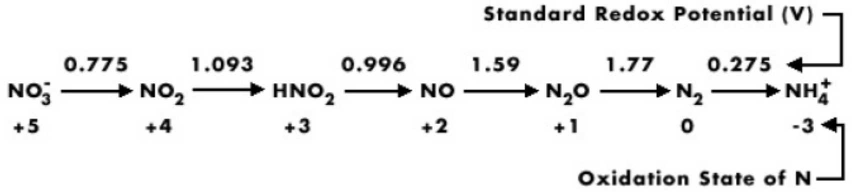

Figure 9: The range in oxidation states of nitrogen. Denitrification involves the transfer of electrons during the reductive transformation of nitrate-N (V) to harmless dinitrogen (0) gas. Ammonium-N (-III) is the most reduced form of nitrogen and is the end product of dissimilatory nitrate reduction (Tiedje, 1988).

According to Herbert & Kovar (1998), different nitrate reduction processes can occur in the same aquifer depending on the availability and reactivity of electron donor material. Nitrate removal would be initiated by denitrification by organic matter and be followed by nitrate reduction by pyrite oxidation.

Published site observation data confirm the role of the organic content, which is either present in surface water or in the aquifer matrix. For the Torgau aquifer (Elbe River, Germany), Grischek *et al.* (1998) identified denitrification in infiltrating river water. In this case, the oxidisable organic C required for the denitrification was originating from both the infiltrating river water and solid matter in the river bed sediments and aquifer material. This hypothesis was further studied by Kedzoriek *et al.* (2008) in a well field of the Lot River (France). The results suggested that 30 to 80% of the total organic carbon consumed by redox reactions during river bank filtration is particulate organic carbon from the aquifer as opposed to dissolved organic carbon from the infiltrating water. In a field study, near the Seine river (France), Doussan *et al.* (1997) concluded that rapid denitrification could occur in the sediment during the infiltration, provided it contains > 1% of particulate organic carbon.

In addition to the importance of the organic content in the sediment or aquifer matrix, Doussan *et al.* (1997) showed that the flow rate of water infiltrating riverbed sediments influences the depletion of nitrate. According to Doussan *et al.* (1997), limiting the flux to a few meters per day while having 1-2% organic content in the sediment should promote denitrification and prevent the further production of ammonia.

In Berlin, during the NASRI project (Pekdeger *et al.,* 2006) surface water at Lake Tegel showed high seasonal variations of NO_3 (2 to 15 mg/L) with an average of 8 mg/L. In the bank filtrate nitrate concentrations were clearly not a function of flow time but of prevailing redox conditions influenced by geochemical variations of the aquifer (<1% org. C), shore (<4% org. C) and lake bottom (>20% org. C) sediment and by the clogging status of the AR pond. During phases of high clogging in the AR pond (e.g. during June 2003) 2 d of travel time were sufficient to reduce nitrate concentrations to below detection limit (< 0.2 mg/L corresponding to > 97 % removal). On the other hand 84 % of the samples with travel times below 50 d still showed detectable amounts of nitrate. Only after a travel time of 130 days nitrate was no longer detectable in any of the samples.

To conclude, high removal (> 97 %) can be achieved provided riverbank filtrate reaches anoxic conditions while benefiting from the presence of electron donors in the aquifer (mainly sedimentary organic carbon, detritus from the clogging layer or pyrite). Both, operation and aquifer properties influence the denitrification capacity of a BF system. It is important to notice that while organic content or clogging is seen here as beneficial for the denitrification process, its role as electron donor could also be detrimental by promoting ammonia production and iron/manganese dissolution.

Summary nitrate

During absence of oxygen and presence of sufficient carbon sources nitrate is rapidly denitrified during BF & AR and can no longer be detected (> 97 % removal) even after contact times of only 2 days.

Factors that limit denitrification are:

- oxic conditions (promoted by short travel times, high flow velocities and low DOC),
- insufficient organic carbon sources.

3.2.5 Disinfection By-Products

Drinking water chlorination may form harmful, potentially carcinogenic disinfection by-products (DBPs) where organic matter is present in source water (Rook, 1974). Trihalogenmethanes (THMs) account for 90% of the by-products produced, the major component (>90%) being chloroform (trichloromethane). Other groups of disinfection by-products are chloral hydrates, cyanogen hydrates, cyanogen chlorides, chlorophenols, bromates, haloacetonitriles (HANs) and halogenated acetic acids (HAAs).

There are few studies on by-product removal during subsurface passage since disinfection is commonly a post-treatment step to BF and AR systems. Therefore, the capacity of BF/AR to remove precursors rather than actual by-products has been studied and been advocated (Weiss *et al.*, 2003; Weiss *et al.* 2004; Drewes & Summers, 2002; Partinoudi & Collins, 2007). Since chlorination is not in use in the area of Berlin, it was not subject to the NASRI trials. The efficiency of BF/AR systems to remove dissolved organic carbon (DOC) as precursors is discussed in more detail in chapter 3.3.1.

Disinfection by-products may be present in the effluent from STP when treated sewage receives final disinfection or in source water of AR systems when an AR technique (e.g. ponded infiltration) follows an oxidation step (e.g. ozonation). The primary removal process for THMs and HAAs in the subsurface is biodegradation whereas other disinfection by-products (e.g. HANs) are mainly removed by hydrolysis (Pavelic, 2004; Pavelic *et al.* 2005).

Schmidt *et al.* (2003) reviewed literature on THM removal by BF/AR systems where the share of ambient groundwater accounted for less than 20% (see Table 9).

Table 9: Removal of THM's during oxic and anoxic BF & AR passage[1)]

initial conc. (µg/l)	removal	conditions[2)]
chloroform (oxic)		
0.4	28%	25-40 d, BF Rhine
1.0	20-45%	10-60 d, BF Rhine
0.45	53%	40-60 d, BF Rhine
2.0	12-85%	30-300 d, BF Rhine
0.82	46-75%	30-300 d, BF Rhine
2.0	21-71%	60-400 d, BF Rhine
0.72	28%	5-10 d, AR Ruhr
0.9 – 8	0-58%	5-15 h
5.5	27%	15 d

initial conc. (µg/l)	removal	conditions[2)]
chloroform (anoxic)		
15	0%	14 d, BF Rhine
12-33	92-94%	2-30 d, BF Rhine
6.5	33%	20-40 d, BF Rhine
<1.9	85%	100-150 d, BF Elbe
20-100	>95%	50-80 d, AR Netherlands
20-100	>95%	40-100 d, AR Netherlands
5.5	95%	15 d
Bromoform		
5.2	87%	15 d, oxic
5.2	84%	15 d, anoxic
5.2	99%	15 d, suboxic

[1)] adapted from Schmidt *et al.* (2003), [2)] flow time in days (d) or hours (h)

The data compiled in Table 9 suggests that brominated THMs are faster attenuated than chlorinated THM's irrespective of redox conditions. For chlorinated THMs, large variations are observed (aerobic: 0-85%, anoxic: 0-95%) but a trend for better removal under anoxic conditions is recognisable. In more than two-third of the anoxic sites, a good removal efficiency (85 to >95%) was found.

Pyne (2006) confirmed the fast and redox-independent removal for brominated THM species (as well as for HAAs). The study by Pyne (2006) further proposed the complete attenuation of 9 days and 9 weeks for HAAs and THMs, respectively, to be representative for ASR wells in the US.

For THM - the major group of disinfection by-products - redox conditions are crucial for attenuation and more important than retention time. Since for all other DBPs, the removal efficiency seems to be redox-independent, an anoxic to anaerobic passage is considered best suitable for the removal of DBPs. The presence of organic carbon in the aquifer has further been suggested to be favourable by dissipating residual chlorine and enabling microbial degradation.

Summary Disinfection By-Products (DBPs)
For the major group of DBPs, trihalogenmethanes (THMs) anoxic to anaerobic conditions are favourable for removal. Under these conditions a removal of 85 % to more than 95 % can typically be observed.

3.3 Substance groups, including substances that show highly variable removal (typically 25 to 75 % overall removal)

Several substance groups are only partially removed by BF & AR systems due to the fact that some substances are well, others are only partially removed or even persistent. The degree of removal is therefore strongly dependent on the actual presence of the individual substances in the source water and therefore difficult to predict. Based on available literature and NASRI results, the characteristics of these substances during subsurface passage are identified in the following with special emphasis on those individual substances that tend to persist. Substance groups, for which even under optimal conditions only a partial removal (25 to 75%) is achievable, are:

- bulk DOC
- pesticides
- pharmaceuticals and x-ray contrast media
- chlorinated hydrocarbons
- monocyclic and polycyclic aromatic hydrocarbons
- endocrine disruptors

3.3.1 Bulk Dissolved Organic Carbon (DOC)

High concentrations of dissolved organic carbon in drinking water are to be avoided in order to limit nutrient availability to microorganisms and prevent microbial re-growth. The removal of DOC further aims at preventing shifts in redox and pH conditions as well as ensuring efficacy of water treatment processes without any impairments such as the formation of disinfection by-products or membrane fouling. Last, not least, organic carbon can impair colour, taste and odour of water if present in excess.

Bank filtration may remove DOC by adsorption and biodegradation. Though adsorption has been observed for DOC of higher molecular weight (Gerlach *et al.* 1998), biodegradation is regarded the predominant process for removal (Sontheimer, 1991; Gerlach *et al.*, 1998). Some fractions of DOC are readily biodegradable (e.g. polysaccharides), others are more persistent. A known and rather persistent fraction is the fraction of humic substances (HS), which are complex carbon structures often accounting for 50 to 80% of the DOC (Abbt-Braun, 1997).

Removal documented for different bank filtration sites is compiled in Table 10. The average removal covers a wide range from 14 % to 84 % but usually lies between 39 % and 50 % (25th and 75th percentile).

Table 10: Removal of dissolved organic carbon (DOC) by BF.

site	**surface water DOC in mg/L**	**well water DOC in mg/L**[2)]	**removal in % min to max or average**	**comment**
BF River Rhine (Brauch *et al.*, 2000)[1)]	2.5 to 2.7	*1.3*	50	attenuation
BF River Rhine (Denecke, 1998)[1)]	3.1 to 3.4	*1.7 to 1.9*	44	
BF Tegel + Wannsee, Berlin (Jekel, 2006, Wiese *et al.,* 2009)	7.1 to 7.2	4 to 5	25 to 44	weeks to months
BF Tegel + AR Tegel, Berlin (Grünheid *et al.*, 2005)	7.3	4.2 to 4.7	35 to 42	BF: up to 4 months (anoxic) AR: up to 50 days (aerobic)
BF River Elbe (Kühn & Müller, 2000)	6.2	4.5	27	
BF River Rhine (Kühn & Müller, 2000)	2.9 to 6.0	1.1 to 2.7	50	DOC decrease in surface water from 1975 to 1997
33 BF sites across Europe (Lenk *et al.*, 2005)	1.4 to 9	0.2 to 5.1	14 to 84	attenuation with 80-90% bank filtrate share in wells,

site	**surface water** **DOC in mg/L**	**well water DOC in mg/L 2)**	**removal** **in %** **min to max or average**	**comment**
	(4.4)	(2.1)	(46)	average concentrations in brackets
BF River Elbe (Ludwig *et al.*, 1997) 1)	6	*3.9*	35	
BF River Neckar (Simon & Kußmaul, 1996) 1)	3 to 5 (*4*)	*2.4*	40	after 1 m (50% in total) average in brackets
BF Tegeler See (Ziegler, 2001)	8.5	5.4 / 5.2	37 / 39	after 32 m and 80 m
BF Müggelsee (Ziegler, 2001)	6.9	4.3 to 4.9	29 to 35	
BF River Ohio (Weiss *et al.,* 2002)	2.7	*0.7 to 1.1*	58 to 74 (66)	61 to 177 m, 3 to 19 d average in brackets
BF River Wabash Weiss *et al.,* 2002)	4.1	*0.5 to 1.5*	64 to 88 (76)	27 to 122 m average in brackets
BF River Missouri Weiss *et al.,* 2002)	3.6	*2.3*	36	37 m

1) adapted from Ziegler (2001) 2) calculated values in italic

A recent study by Lenk *et al.* (2005) investigated the DOC attenuation at 33 BF sites across Europe with BF shares of 80 to 90 %. They derived a non-linear regression model formula (R^2= 0.74) as follows:

$$\mathbf{y = -0.503 + 0.811 \ln(x_1) + 0.236\, x_2^{0.437} + 7.428\, x_4}$$

where:

y = DOC attenuation (mg/L)

x_1 = DOC concentration in surface water (mg/L)

x_2 = flow time in the subsurface (d)

x_4 = transmissivity (m^2/s)

This suggests the highest attenuation rates for low source water concentrations, low transmissivities and high flow times. High attenuation rates (82 to 84%), for instance, were observed for a BF site at the river Rhine near Cologne in Germany where the travel distance was about 300 m and flow times between 95 and 134 days (transmissivity: 5.3×10^{-3} m^2/s). The lowest attenuation (14%) was observed in Austria nearby the river Enns, however, travel distance and flow time were only 0.2 m and 0.01 d respectively (transmissivity: 3.8×10^{-3} m^2/s). The DOC attenuation (%) against flow time according to this equation for conditions found in Berlin (average source water concentration: 7.1 mg/l, transmissivity: 2×10^{-2} m^2/s) is given in Figure 10 (red curve).

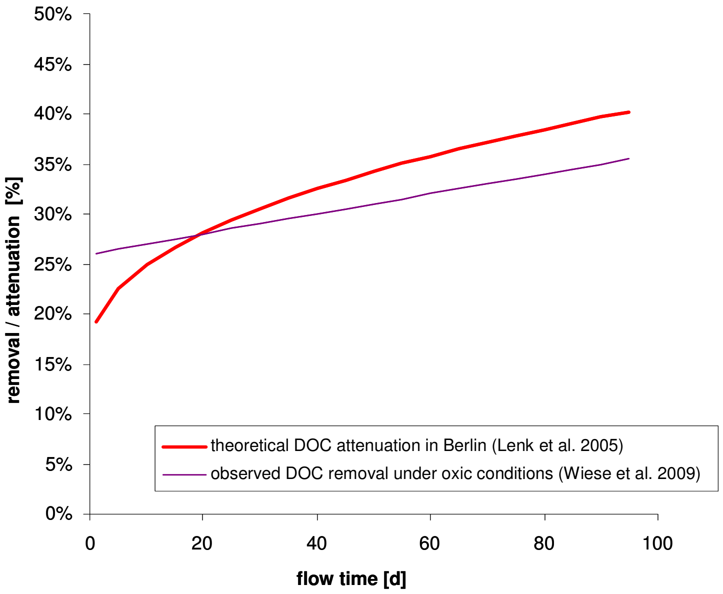

Figure 10: Theoretical DOC attenuation in Berlin according to the formula by Lenk et al. (2005) and regression curve of removal observed at the field sites during the NASRI project (Wiese et al. 2009).

Removal rates derived from NASRI-data (Wiese et al. 2009) for oxic conditions are also given in Figure 10. For other redox zones, no time dependency was determined. The NASRI field data suggested an only slightly higher mean removal rate during oxic subsurface passage than under nitrate (28%), iron (26%) or manganese (29%) reducing conditions. However, removal rates derived from technical scale experiments (soil columns, 30 m) yielded a more pronounced difference showing 47% removal under oxic and 31% under anoxic conditions (Jekel, 2006). The reason for this discrepancy might lie in the different aquifer matrix with higher shares of sedimentary bound organic carbon in the Berlin aquifers (up to 1 %, Pekdeger et al., 2006) than in the columns (0.04 %, Jekel 2006).

In general, the NASRI data follows the empirical equation by Lenk *et al.* (2005) quite well, representing a site with low DOC removal. This is due to the high background of DOC including substantial shares of humic substances in source and groundwater as well as in the aquifer matrix itself.

A residual of DOC typically remains in bank filtrate (>50%). However, it is subject to variation, e.g. due to changing input concentrations as well as seasonal and redox changes (Jekel, 2006, Ziegler, 2001). Taking the concomitant degradation of an unknown share of sedimentary-bound organic carbon into account, the DOC degradation rates in the subsurface are likely higher than what is observed.

A complete DOC removal is not achieved by bank filtration but the readily biodegradable fraction is consumed and bank filtrate will exhibit low re-growth potential (Jekel, 2006). Depending on the pre-treatment applied, carbon may be rendered bio-available again by reduction of complex carbon compounds (e.g. by ozonation) or produce undesired disinfection by-products. It is therefore necessary to adapt the post-treatment to the DOC residual in the bank filtrate.

Summary DOC

Dissolved organic carbon is typically removed by 39 to 50 % during subsurface passage in BF and AR systems. Biodegradable DOC (e.g. polysaccharides) is removed during the first few meters of infiltration so that bank filtrate exhibits a low re-growth potential.

Factors counteracting maximum removal are:

- short travel times,

- anoxic or anaerobic conditions,
- high background DOC in source or ambient ground water with substantial shares of humic substances
- high amounts of organic carbon in the aquifer matrix.

3.3.2 Pesticides

The "Pesticide Manual" lists some 1,524 different pesticides including insecticides, herbicides and fungicides (BCPC 2008). They are utilised in many applications in households and agriculture whereby they enter the water cycle (Miller, 2004). Their occurrence exhibits high seasonal variations due to the mode of application. Therefore dispersion plays an important role for attenuation of peak concentrations in addition to adsorption and biodegradation. The latter removal processes are highly substance-specific, however, due to international regulations the key parameters characterizing adsorption (K_{oc} or K_d) and biodegradation (DT_{50}) are available for the majority of pesticides (http://sitem.herts.ac.uk/aeru/iupac). On this basis pesticides can be classified with respect to their mobility in the subsurface (e.g. as "CA Groundwater Contaminant" according to Kegley et al. 2008).

However, pesticides persistent during BF / AR are not necessarily limited to those frequently found in other groundwater systems: Mathys (1994) examined the degree of pesticide pollution in an agricultural area based on data collected from surface and groundwater bodies between 1987 and 1992. The observation wells influenced by bank filtrate and aquifer recharge accounted for significantly higher pesticide levels than ambient groundwater wells. This highlights the importance of the upper soil and unsaturated zone for attenuation of pesticides (Stieber et al. 2007) which are usually by-passed during BF and AR.

Reported removal efficiencies for pesticides by means of BF / AR range between 0 % and 100 % (see Table 11). Generally high removal (usually 50 % and more) can be observed for organochlor insecticides and anilides whereas the removal of triazines, ureas and phenoxy herbicides is redox-dependant: Triazines are usually not well removed (median removal rates usually < 30 %), however for anoxic and anaerobic sites removal can reach more than 90 %. On the other hand, removal observed for ureas and phenox herbicides is usually higher under oxic / suboxic conditions.

In Berlin, during the NASRI project, the fate of the DDT-transformation products p,p'-DDA and o,p'-DDA as well as of bentazone and mecoprop was investigated at three field sites. For the DDT transformation products no removal was observed, whereas bentazone showed removal between 20 % and 90 %. For mecoprop a redox-dependent removal could be quantified: with average source water concentrations of 0.014 µg/L samples from the oxic and Mn-reducing zone showed 55 % to 90 % removal whereas samples obtained from the suboxic and Fe-reducing zones showed a maximum of 43 % removal (Wiese et al. 2009).

Table 11: Median removal rates for pesticides given in the TZW database (Schmidt 2007), substances of high relevance in the EU according to Tedesco et al. (2009) marked with shaded areas.

pesticide	median removal	number of sites	comments
Triazines			
Ametryn	> 25 %	1	
Atrazine	20 %	30	oxic / suboxic: 14 % anoxic / anaerobic: 57 %
Desethylatrazine	7 %	10	indications for higher removal under anaerobic conditions
Desisopropylatrazine	8 %	2	

pesticide	**median removal**	**number of sites**	**comments**
Cyanazine	8 %	2	
Propazine	18 %	2	
Simazine	27 %	14	oxic / suboxic: 24 % anoxic / anaerobic: 65 %
Terbuthylazine	30 %	6	
Terbutryne	84 %	1	
Triazines (incl. Desethyltriazine, Atrazine, Simazine)	49 %	1	
Ureas			
Chlortoluron	0 %	5	100 % removal at oxic AR sites
Diurone (DCMU)	12 %	7	mainly anoxic BF sites
Isoproturone	0 %	9	> 50 % removal at oxic BF / AR sites
Methabenzthiazurone	85 %	2	
Metoxuron	> 70 %	1	
Ureas (incl. Chlortoluron, Isoproturone, Methabenzthiazurone)	86 %	1	
Phenoxy herbicides			
2,4-D (2,4-Dichlorophenoxyacetic acid)	92 %	2	
Dichlorprop	70 %	2	indications for lower removal under anoxic conditions
MCPA (2-methyl-4-chlorophenoxyacetic acid)	87 %		
Mecoprop (MCPP)	40 %	6	oxic / suboxic: 85 % anoxic: 0 %
2,4,5-T (2,4,5-Trichlorophenoxyacetic acid)	90 %	1	
Organochloride insecticides			
Endosulfan	85 %	1	
Hexachlorobenzene (HCB)	33 %	9	suboxic: 0 % anoxic: 92 %
HCH (total)	71 %	4	
- alpha HCH	55 %	3	
- beta HCH	72 %	1	
- gamma HCH (lindane)	80 %	7	indications for lower removal under oxic conditions
- delta HCH	57 %	1	
PCP (Pentachlorophenol)	47 %	3	
Anilides / anilines			
Acetochlor	41 %	2	
Alachlor	76 %	1	
Dimethenamid	9 %	1	
Metazachlor	89 %	6	
Metolachlor	49 %	4	
Others			
paraoxon-eq.	94 %	2	
parthion-eq.	75 %	2	
bentazone	0 %	6	
2-amino-n-isopropylbenzamide	0 %	3	bentazone metabolite
carbaryl	90 %	1	
iso-chloridazon	80 %	4	

pesticide	median removal	number of sites	comments
dikegulac	0 %	5	
flufenacet	63 %	1	
glyphosate	24 %	2	
AMPA	90 %	3	glyphosate metabolite
Metalaxyl	> 75 %	1	
trifluralin	51	1	

To conclude, of those pesticides that were classified of high relevance for drinking water production in the EU (Tedesco et al. 2009)

- 5 show good removal of > 70 % (2,4-D, Dichlorprop, MCPA, Metazachlor, AMPA),
- 4 show medium removal between 30 % and 70 % (terbuthylazine, mecoprop, metolachlor and flufenacet),
- 7 show low removal of < 30 % (atrazine, desethylatrazine, simazine, diurone, isoproturone, bentazone and glyphosate)

(for 4 pesticides no field studies from BF / AR sites are available). Depending on the substance type and source water concentrations conventional or advanced post-treatment might therefore be necessary. However, the effort for post treatment is reduced by BF / AR, especially at sites with travel times of several weeks due to the dispersion of peak concentrations typical for pesticide occurrence in surface waters.

Summary Pesticides

For pesticide removal no general recommendation can be given, due to the fact that this group comprises substances of highly variable chemical structure. In addition, redox conditions play an important role for the removal of some types of pesticides. However, even for persistent pesticides a smoothing of seasonal peaks can be expected due to dispersion within the aquifer.

Pesticides of which field studies have shown **limited removal (< 50 %)** in BF & AR systems are:

- triazines: atrazine, desethylatrazine, desisopropylatrazine, cyanazine, propazine, simazine, terbutylazine
- ureas: chlortoluron, diurone, isoproturone
- phenoxy-herbicides: mecoprop (under anoxic conditions)
- organochloride insecticides: hexachlorbenzene (HCB), pentachlorophenole (PCP),
- anilides / anilines: dimethenamid, metolachlor
- others: bentazone (plus its transformation product 2-amino-n-isopropylbenzamide), glyphosate, dikegulac

3.3.3 Pharmaceuticals and x-ray contrast media

Pharmaceuticals used in human and veterinary medicine may enter the environment via excretion, inadequate disposal of medications or via municipal effluent. Although concentrations in source water are far below medical doses, the possibility of adverse, synergistic effects on human health and potential emergence of antibiotic-resistant bacteria have raised concerns (Kleiner et al. 2007).

Most pharmaceuticals observed in surface waters show little tendency to adsorb to particulate matter (for a compilation of retardation factors see Wiese et al. 2009), therefore (bio-)degradation it the most prominent removal process during BF & AR. Investigations in The Netherlands and in Berlin have shown that the redox potential in the aquifer is decisive for the degree of elimination (Stuyfzand 1998, Massmann et al. 2007).

During the NASRI project in Berlin investigations on 17 relevant (i.e. potentially mobile or so far little investigated) pharmaceuticals and x-ray-contrast media (incl. the sum parameter AOI) as well as on 6 related transformation products were carried out at 3 different field sites (see Massmann et al. 2007 and Heberer et al. 2008 for details). The resulting comprehensive interpretation documented by Wiese et al. 2009 is the basis for the removal values given in Table 12.

Table 12: Observed removal rates for pharmaceuticals and x-ray contrast media.

substance	**observed removal (%)**		**reference**
	oxic	**anoxic**	
Antibiotics			
Clarithromycin	> 96		Wiese et al. 2009
Clindamycin	> 97	27 - 98	Wiese et al. 2009
Dehydroerythromycin	> 97		Wiese et al. 2009
Roxithromycin	> 99		Wiese et al. 2009
Sulfamethoxazole	31 - 47	74 - 89	Wiese et al. 2009
Sulfadimidine	no removal observed**		Wiese et al. 2009
Sulfadiazin	n.a.	50 – 70	Hartig 2000
Trimethoprim	> 95		Wiese et al. 2009
Lipid regulators			
bezafibrate	removal observed**		Wiese et al. 2009
clofibric acid	66 - > 75	51 - 90	Wiese et al. 2009, Ternes et al. 2000
Anticonvulsants			
carbamazipine	8 - 16	20 - 51	Wiese et al. 2009
primidone	< 4		Wiese et al. 2009
Anti inflammatory drugs			
phenazone	73 - 91	6 - 46	Wiese et al. 2009
propyphenazone	56 - 95	0 * - 79	Wiese et al. 2009
Analgesics			
diclofenac	> 80 - > 95	61	Brauch et al. 2000, Wiese et al. 2009
indometacine	removal observed**		Wiese et al. 2009
X-ray contrast media			
Iopromide	94 - 95	70 - 99	Wiese et al. 2009
Diatrizoic acid	65 – 95		Putschew & Jekel 2001
AOI (sum parameter)	4 – 31	38 – 58	Wiese et al. 2009
Transformation products			
AAA	98 – 99	56 – 90	Wiese et al. 2009
AMDOPH	1 – 26	0*	Wiese et al. 2009
AMPH	removal observed**	0*	Wiese et al. 2009
DP	removal observed**	0*	Wiese et al. 2009

substance	observed removal (%)		reference
	oxic	anoxic	
FAA	93 – 94	removal observed**	Wiese et al. 2009
PDP	n.a.	0*	Wiese et al. 2009

* increase observed due to formerly higher concentrations in surface water or metabolite formation

** no quantification possible due to high number of values < LOD.

Of the 14 substances (+ AOI) for which a quantification of removal rates in different redox zones is possible, the majority (71 %) is either well (> 95 %) to moderately well (> 50 %) removed regardless of redox conditions or tends to be better removed under oxic conditions. These are the antibiotics clarithromycin, clindamycin, dehydroerythromycin, roxithromycin and trimehoprim, the lipid regulator clofibric acid, the anti inflammatory drugs phenazone and propyphenazone, the analgesic diclofenac and the x-ray contrast media iopromide. One investigated pharmaceutical was shown to be persistent (the anti-convulsant primidone) and three parameters were preferably (but not rapidly) removed under anoxic conditions (the antibiotic sulfamethoxazole, the anticonvulsant carbamazipine and AOI as sum parameter for iodinated x-ray contrast media).

Concerning the removal kinetics only five of the substances investigated in Berlin showed a time-dependency. Table 13 gives a summary of the derived half-lives (DT_{50}) supplemented with data from different AR and ASR sites in The Netherlands (Stuyfzand et al. 2007). The data shows that residence times in the range of a few months will enable the removal of clofibric acid, diclofenac and sulfamethoxazole whereas for carbamazipine the necessary contact times lie in the range of decades.

Table 13: Half lives for pharmaceuticals and x-ray contrast media in BF, AR and ASR systems.

substance	DT_{50} (d)		reference
	oxic / (sub)oxic	anoxic	
Carbamazipine	- / 9999	> 7300	Stuyfzand et al. 2007
Clindamycin	3.4 / -	-	Wiese et al. 2009
Clofibric acid	23 / -	-	Wiese et al. 2009
Dehydroerythromycin	2.3 / -	-	Wiese et al. 2009
Diclofenac	23 / -	-	Wiese et al. 2009
Iopromide	- / 0.7 to < 7	-	Stuyfzand et al. 2007
Phenazone	- / 2	>> 730	Stuyfzand et al. 2007
Sulfamethoxazole	39 / 9999	30	Wiese et al. 2009, Stuyfzand et al. 2007

To conclude, the available data on pharmaceuticals and x-ray contrast media in BF and AR systems shows that some may be removed well (> 90 %) from affected surface waters. For optimal performance oxic conditions and sufficient contact time needs to be ensured (for 95 % removal of most pharmaceuticals and x-ray contrast media: > 15 d, for 95 % removal of additionally clofibric acid, diclofenac and sulfamethoxazole > 73 d). Under these pre-requisites those of the investigated parameters to be expected in the raw water would limit themselves to carbamazipine, primidone and AOI.

However, it has to be taken into account that biodegradation of the pharmaceuticals and x-ray contrast media will lead to the formation of transformation products. As seen for Berlin, these might be more stable than the parent compound and even persist in the subsurface for decades (Massmann et al. 2008).

Summary Pharmaceuticals and X-ray Contrast Media

For pharmaceuticals and X-ray contrast media many substances show good removal during BF & AR. Some PhACs (e.g. carbamazipine) show redox-dependant removal.

Pharmaceuticals and X-ray contrast media of which field studies have shown limited removal (< 50 %) in BF & AR systems are:

- regardless of redox zone: sulfadimidine, primidone, AMDOPH, carbamazipine (< 20 % under oxic conditions), AOI (< 31 % under oxic conditions)
- under oxic conditions: sulfamethoxazole
- under anoxic conditions: phenazone, clindamycine

3.3.4 Chlorinated hydrocarbons

Chlorinated hydrocarbons comprise many water pollutants of high toxicity and persistence among which are the already mentioned organochlorine pesticides (e.g. DDT) and disinfection by-products. Other chlorinated hydrocarbons include typical organic groundwater contaminants as e.g. carbon tetrachloride (CTC), trichloroethylene (TCE), tetrachloroethylene (PCE) and 1,2-dichloroethane (1,2-DCA). These are usually released into the environment in the form of solvents, cleaners and degreasers via spills and leakages. Relevant concentrations in surface waters would therefore typically be limited to peak values subsequent to an accident or spill. On the other hand chlorinated hydrocarbons are the most frequently detected groundwater contaminants at hazardous waste sites (Rivett et al. 2006) therefore ambient groundwater may be affected by chlorinated hydrocarbons, especially in urban areas.

Chlorinated hydrocarbons are attenuated in the subsurface by adsorption and biodegradation – though the efficiency of both is limited. Table 14 gives the range retardation coefficients calculated from published K_{ow} values and parameters typical for the Berlin bank filtration sites. The retardation of highly soluble CHCs would therefore usually lie below 5, meaning only moderate (and reversible) attenuation by sorption whereas less soluble CHCs can be retarded significantly.

Table 14: Theoretically determined retardation of chlorinated hydrocarbons by adsorption at a Berlin BF site.

Parameter	**Range for highly soluble chlorinated hydrocarbons** (DCM, TCM, VC, 1,2-DCA, 1,1-DCE, cDCE, tDCE)	**Range for less soluble chlorinated hydrocarbons** (CTC, TCE, PCE, 1,2-DCB, 1,4-DCB)
K_{OC} (mL/g)*	8.8 – 65	94 – 616
K_d (mL/g) (= f_{OC} * K_{OC} with f_{OC} = 0.01**)	0.088 – 0.65	0.94 – 6.16
R (= 1+(ρ/η) * K_d, with ρ = 1.8 g/cm³ and η = 0.3)***	1.5 – 4.9	6.6 - 38

* according to Rivett et al. 2006
** according to Pekdeger et al. 2006.
***standard values according to Hölting & Coldewey 2009

In theory, chlorinated hydrocarbons with a low number of substituents are predominantly biodegraded under oxic conditions while higher substituted hydrocarbons (e.g. TCE, PCE, DCE) are biodegraded better in the absence of oxygen. Field data studies from Noble & Morgan (2002) found average half-lives for DCE at groundwater temperatures (10 °C) to be 39 days under aerobic and 4,060 days under anaerobic conditions.

Efficient shock load mitigation for the highly soluble chlorinated hydrocarbon 1,2-DCA by bank filtration has been observed by Kühn & Müller (2000) where a peak concentration of 35 µg/l in surface water reached a monitoring well after 3 weeks of subsurface passage. In the bank filtrate, the 1,2-DCA was clearly removed and residuals showed a plateau concentration of 1 µg/l for 2-3 weeks (Figure 11).

Data from BF and AR sites confirm the redox dependent removal for some chlorinated hydrocarbons: CTC and PCE show significantly higher removal under anoxic / anaerobic conditions. The values given for TCE on the other hand do not show differences between the redox zones as this would be expected (see above).

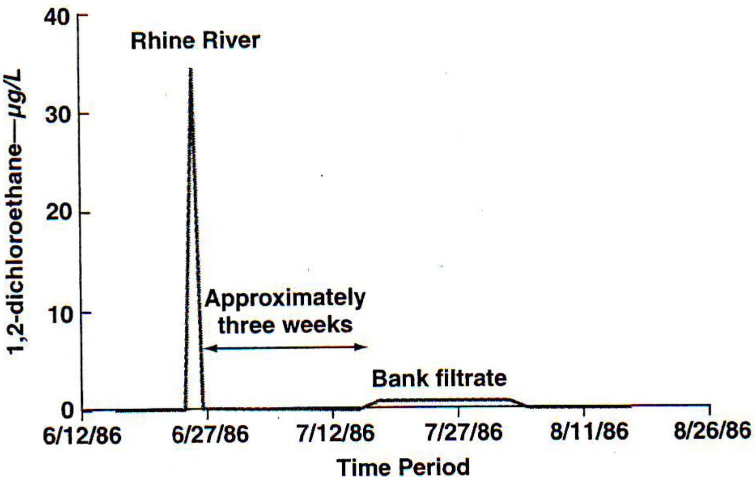

Figure 11 1,2-DCA concentrations in source water and bank filtrate at a BF site along the river Rhine (from Kühn & Müller 2000).

Table 15: Observed removal for chlorinated hydrocarbons at BF & AR sites (compiled from Schmidt 2007).

Parameter	median observed removal (%)	number of studies	Comments
Highly soluble			
1,2 dichloroethane (1,2-DCA)	70	1	suboxic conditions
1,1,1-trichloroethane (TCA)	0	4	oxic conditions only
Trichloroethylene (TCE)	5	18	
tetrachloroethylene (PCE)	15	22	- oxic / suboxic: 0 % - anoxic / anaerobic: 75 %
1,1,2,2-tetrachloroethane	67	1	oxic conditions only
Less soluble			
Carbon tetrachloride (CTC)	64	13	- oxic: 58 % - suboxic / anoxic: 77 %
Hexachloroethane	>99	1	anoxic conditions
hexachlorobutadiene (HCBD)	95	5	80 % anoxic / anaerobic conditions

To conclude, chlorinated hydrocarbons are generally quite mobile and persistent in the subsurface suggesting BF and AR to be only of limited benefit for the removal of these compounds. On the other hand, shock loads by which these substances typically occur in surface waters are dampened by dispersion and retardation. A site optimized with respect to this function would need to show average travel times of a few weeks so that peak concentrations lasting some hours to a few days can be efficiently reduced.

Summary Chlorinated Hydrocarbons (CHCs)

Whereas less soluble CHCs can be removed by up to > 99 % at BF & AR sites, highly soluble CHCs tend to show little removal, especially under oxic conditions (0 – 67 %). There are indications that anoxic conditions are favourable for the removal of highly soluble CHCs like tetrachloroethylene (PCE).

CHCs that have shown to break through in existing BF & AR systems are:

- 1,1,1-trichloroethane (TCA), trichloroethylene (TCE) and tetrachloroethylene (PCE)

3.3.5 Monocyclic and polycyclic aromatic hydrocarbons

The most common monocyclic aromatic compounds are benzene, toluene, ethylbenzene and xylene (BTEX group). BTEX find application as fuel additives and solvents in many industrial processes.

BTEX are moderately soluble in water and thus, mobile in water systems. Mean half-lives for BTEX compiled by the Rivett *et al.* (2006) from Noble & Morgan (2002) were <220 days under aerobic conditions and <519 days under anaerobic conditions. Degradation of BTEX members was observed to be best in the order: toluene >ethylbenzene >m-xylene >o-xylene >benzene >p-xylene (Dou *et al*. 2008). This is supported by the data from BF and AR sites compiled from Schmidt (2007) given in Table 11.

Figure 12: Median removal rates observed for aromatic hydrocarbons during BF/AR passage compiled from Schmidt 2007.

Substance	**median observed removal (%)**	**number of studies**	**Comments**
Monocyclic aromatic hydrocarbons			
Benzene	48	8	
Ethylbenzene	72	6	
Toluene	81	10	
Polycyclic aromatic hydrocarbons (PAH)			
Naphtalene	94	4	
Phenanthrene	28	1	
Fluoranthrene	40	4	
Benzo(a)pyrene	77	4	

Polycyclic aromatic hydrocarbons (PAH) are generated as by-products from incomplete combustion processes of organic matter as well as during wood and metal treatment and therefore ubiquitously found in the environment. Though less biodegradable than the BTEX group due to a more complex structure PAHs are generally better absorbable to the aquifer matrix (Rivett at al. 2006). E.g. under conditions found in Berlin Benzo(a)pyrene (K_{OC} = 969,000 mL/g) would be retarded by a factor of 58,141 compared to a tracer (see Table 14 for the method of calculation) thus leaving time for degradation and dispersion (more than 14.000 years if the flow time is 3 months).

In case of surface water contamination with aromatic hydrocarbons, bank filtration systems have a certain capacity to mitigate shock loads and degrade the pollutants. However, degradation may take months to years and the concomitant changes in redox zonation are likely to impair the overall cleaning capacity of bank filtration.

Summary Mono- and Polycyclic Aromatic Hydrocarbons

Median values of observed removal of monocyclic aromatic hydrocarbons ranges from 48 % (benzene) to 81 % (toluene). Polycyclic aromatic hydrocarbons (PAHs) are generally less mobile in aquatic systems due to their affinity to organic aquifer matter.

Substances likely to be poorly removed (< 50 %) in BF & AR systems are:

- Monocyclic aromatic hydrocarbons: benzene, m-, o-, and p-xylene,
- Polycyclic aromatic hydrocarbons (PAH): phenanthrene, flouroanthrene.

3.3.6 Endocrine-disrupting chemicals

Endocrine disrupting chemicals (EDCs) encompass a large group of chemicals. The important group of pesticides (many of which are endocrine disruptors) is discussed separately (Chapter 3.3.2 pesticides). Non-pesticides that interfere with the endocrine system and are commonly found as pollutants in the environment are estradioles, bisphenol A (BPA), n-nonylphenoles (NPs), octylphenoles (OPs) and phtalates. Field studies on their fate in BF / AR systems are scarce and usually do not cover different settings regarding travel times and redox conditions. Table 16 gives an overview of the available data compiled from Schmidt (2007).

Table 16: Observed removal rates compiled from Schmidt (2007) and NASRI data.

Substance	range of observed removal (%)	number of studies in Schmidt (2007)	additional references
alkylphenolpolyethoxccarboxylate (APnEC)	94	1	
Estron (E1)	89	1	
	90		Zühlke 2004
Nonylphenols			
- NP1EO	75 to 99	3	
- NP2EO	> 50 to > 99	3	
- NP1EC	34 to 96	2	
- NP2EC	38 to 79	1	
- Nonylphenol	85 to 95	3	
Phtalates			
- DMP	> 93	1	
- DEP	50 to 97	2	
- DBP	40 to 90	2	
- DEHP	25 to 75	2	
Bisphenol-A	> 95	1	

In Berlin, during the NASRI project, Estron (E1), 17ß-estradiole (E2) and 17α-ethinylestradiole (EE2) were measured at the bank filtration and aquifer recharge site at Lake Tegel (Zühlke 2004). For E2 and EE2 singular values were detected in the source water, not exceeding 0.2 ng/L (LOQ). Both substances were not detectable (< 0.1 ng/L) in the bank filtrate (oxic and anoxic conditions, travel times > 3 d). E1 was present in the source water at concentrations of 1 ng/L in average. At the AR site the observation well closest to the pond (oxic conditions, travel times 3 – 21 d) showed average concentrations of close to the detection limit (0.1 ng/L) whereas in all other observation wells the substance was not detectable (< 0.1 ng/L).

Summary Endocrine Disruptors

Though there is little published data from field sites, it appears that many endocrine-disrupting chemicals (e.g. bisphenol-A, estron) are removed by more than 90 % by BF/ AR systems. For phtalates and some nonylphenolic compounds, however, removal might be limited (< 50 %). As for all organic micropollutants, the toxicity (here: endocrine-disrupting effect) and persistence of transformation products is likewise to be taken into account.

3.3.7 Other organic trace compounds

In addition to the organic trace substances mentioned, a large number of organic trace compounds is found in the environment and in BF & AR systems. Table 17 gives information on additional substances investigated during the NASRI project. While 1,5-NDSA and EDTA were shown to be persistent, some removal was observed for 1,7-NDSA, 2,7-NDSA, MTBE and NPS.

Table 17: Observed removal rates for selected industrial chemicals and complexing agents.

substance	observed removal (%)		reference
	oxic / **suboxic**	**anoxic** / **anaerobic**	
1,5-NDSA	< 3		Wiese et al. 2009
1,7-NDSA	47 – 57	20 - 29	Wiese et al. 2009
2,7-NDSA	47 – 54	22 - 28	Wiese et al. 2009
EDTA	0*		Wiese et al. 2009
	0 – 46		Schmidt 2007
MTBE	20 – 26		Wiese et al. 2009
	34 – 66	no data	Schmidt 2007
NPS	removal observed**		Wiese et al. 2009

* increase observed

** no quantification possible due to concentrations below LOQ

A review based on published values (Stuyfzand et al. 2007, Schmidt 2003) and NASRI data by Wiese & Grützmacher (2009) with the aim to identify substances persistent in BF & AR systems revealed that of 340 scanned organic trace substances 45 (13 %) showed little or no removal (< 30 %) under field conditions. In general, BF and AR can therefore be classified as suitable for the removal of trace organics.

Summary Other Organic Trace Substances

Of the other organic trace substances investigated during the NASRI project the following have shown to be only poorly (< 30 %) removed by BF & AR, regardless of redox zone:

- 1,5-NDSA, EDTA, MTBE

Limited removal (< 50 %) was also observed for

- 1,7-NDSA and 2,7 NDSA (with slightly enhanced removal under oxic conditions).

3.4 Substances which are generally poorly or not removed by BF & AR systems or may even become mobilized

As demonstrated in the chapters above, BF & AR systems support the removal of many substances problematic for drinking water from surface waters. However, several substance groups of concern are not or only poorly removed by subsurface passage or even may become (re-)mobilized. Among these substances are:

- Ammonium
- Major anions & cations
- Inorganic trace elements

3.4.1 Ammonium

Ammonium attenuation in the subsurface is due to adsorption and oxidation (nitrification), however, it can also be produced by ammonification of organic bound nitrogen (Canter 1996). While ammonium adsorption will only be of relevance in fine-grained, org.-C-rich aquifers, the oxidation of ammonium (nitrification) is limited by the maximum amount of oxygen dissolvable in water (11 mg/L at 10 °C, with lower concentrations at higher temperatures). Therefore the nitrification capacity in theory could amount to 6.2 mg/L maximum (> 10 °C), but only in absence of other oxygen reducing substances in the aquifer. As soon as other electron acceptors (e.g. biodegradable organic carbon) are present in the infiltrating water or the aquifer matrix the capacity to oxidize ammonium will be clearly lower.

In Berlin during the NASRI project (Pekdeger *et al.* 2006) average ammonium concentrations in the surface water of 0.21 mg/L (40 % < LOD) were completely removed during the oxic subsurface passage at the AR site Tegel (< 0.04 mg/L). No removal or even increasing concentrations (up to 6.5 mg/L) were observed for anoxic / anaerobic conditions in deeper observations wells of the Lake Tegel and Lake Wannsee bank filtration sites. As a result, ammonium concentrations in the production wells of the BF sites lie around 1 mg/L, whereas the production well adjacent to the AR pond shows median ammonium levels of 0.2 mg/L (due to mixing with elevated ammonium levels in ambient groundwater of up to 1 mg/L).

Increasing ammonium concentrations during subsurface passage have also been reported from a field site in Delhi, India with high shares of untreated waste water in the source river Yamuna (Pekdeger et al. 2008): median surface water concentrations of 8 mg/L rose up to 16 mg/L and 24 mg/L in shallow and medium deep groundwater, respectively.

Summary Ammonium
High removal (> 99 %) of ammonium from surface water by subsurface passage is limited to strictly aerobic conditions and concentrations well below 6 mg/L under saturated flow conditions. On the contrary, for scenarios with high shares of organic N either in source water (e.g. through high shares of untreated sewage) or within the aquifer matrix an increase of ammonium levels has to be expected.

3.4.2 Major Anions & Cations (total dissolved solids, TDS)

The chemical water composition is constituted by a range of major cations: Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}; and anions: SO_4^{2-}, Cl^{-}, HCO_3^{-} which usually occur in concentrations in the range of mg/L thus comprising the major constituents of the parameter “total dissolved solids” (TDS). Though not health relevant, high levels of TDS (> 1000 mg/L, WHO 2003) may be objectionable to consumers (taste considerations) and may also be of concern for the production of industrial process water (especially Ca^{2+} and Mg^{2+} affecting water hardness).

When water infiltrates into the subsurface, it interacts with the surrounding sediment texture and minerals containing ions such as calcium, magnesium and bicarbonate can become dissolved. This is promoted by the production of protons during biodegradation and may buffer the pH. However, during the NASRI project, Greskowiak et al. (2006) observed a significant drop in pH (- 0.5 pH units) during summer in the recharged water of the AR pond Tegel that was not buffered by the calcite of the aquifer due to its limited solubility at higher temperatures.

Some anions (e.g. chloride) are conservative and show little interaction with the surroundings. During the NASRI project chloride, that showed seasonal variations, was investigated as tracer to determine the travel times between source water and observation wells. Other anions (e.g. sulfate) can be involved in microbial redox processes. In Berlin, during the NASRI project, Pekdeger et al. (2006) modelled sulfate reduction in a mud core drilled in the bottom of Lake Tegel and obtained half lives for sulfate between 50 d and 350 d. However, even after more than 100 d travel time no indications for sulfate reduction were observed in young bank filtrate that infiltrated along the lake shore. This shows that next to travel time aquifer composition is decisive for the occurrence of sulfate reduction.

Summary Major Anions & Cations (total dissolved solids, TDS)
Major anions and cations are usually not retained during BF & AR. On the contrary, due to calcite dissolution increasing concentrations of Ca^{2+}, Mg^{2+} and HCO_3^- are possible. Relevant sulfate reduction can only be expected after travel times of > 50 d in aquifers with high shares of organic material under strictly anaerobic conditions. The composition of the raw water in BF & AR systems concerning major anions and cations will be primarily determined by the mixing ratio of bank filtrate and ambient groundwater.

3.4.3 Inorganic trace elements

Though strictly speaking all inorganic trace elements found in the environment are of geogenic origin, they will herein be distinguished into those predominantly entering the water cycle due to anthropogenic activities (Pb, Cu, Zn and Cd) and those originating from the subsurface (As, F, Fe, Mn, Se, Cr, U).

Inorganic trace elements predominantly originating from anthropogenic activities enter the water cycle through point sources (e.g. industrial effluents) or, with increasing relevance in Europe, from diffuse sources (e.g. run-off from metal coated roofs). For those inorganic trace elements that originate predominantly from the subsurface a change in redox conditions during groundwater abstraction can trigger remobilization. Table 18 gives an overview of the removal observed at BF & AR field sites.

The complex interplay of precipitation/dissolution, microbial uptake and release, sorption and desorption processes does not remove these substances permanently from the water phase and results in large “removal” variations. “Removal rates” are to be interpreted with care since removal is temporary and soil may serve as sink and source. Therefore, they are provided in Table 18 as maximum decrease (attenuation) and maximum increase indicating an actual gain.

Table 18: Decrease/increase percentage rates observed for inorganic substances during BF/AR passage.

inorganic substance	surface water conc.	max. decrease (-) and max. increase (+)	comment
of predominantly anthropogenic origin			
Lead (Pb)			
Schmidt *et al.* (2003)	13 µg/l	Max. decrease:-75%	
Stuyfzand *et al.* (2006)	0.3 µg/l	Max. increase: +333%	100 m, anoxic

inorganic substance	surface water conc.	max. decrease (-) and max. increase (+)	comment
Cadmium (Cd)			
Schmidt *et al.* (2003)	2 µg/l	Max. decrease:-99%	
Stuyfzand *et al.* (2006)	0.03 µg/l	Max. increase: +33%	10 m, anoxic
Copper (Cu)			
Stuyfzand *et al.* (2006)	5 µg/l	Max. decrease:-90%	10 m, anoxic
		Max. increase: n.a.	
Zinc (Zn)			
Schmidt *et al.* (2003)	0.18 mg/l	Max. decrease:-82%	
		Max. increase: n.a.	
Nickel (Ni)			
Stuyfzand *et al.* (2006)	3.3 µg/l	Max. decrease:-88%	100 m, anoxic
Stuyfzand *et al.* (2006)	1.5 µg/l	Max. increase: +13%	24 m, oxic, shallow aquifer
predominantly of geogenic origin			
Iron (Fe)			
Stuyfzand *et al.* (2006)	80 µg/l	Max. decrease:-75%	24 m, oxic
Stuyfzand *et al.* (2006)	0.98 mg/l	Max. increase: +170%	220 m, anoxic
Manganese (Mn)			
Stuyfzand *et al.* (2006)	0.4 µg/l	Max. decrease:-75%	24 m, oxic
Stuyfzand *et al.* (2006)	0.13 µg/l	Max. increase: +1107%	100 m, anoxic
Arsenic (As)			
Schmidt *et al.* (2003)	4.2 µg/l	Max. decrease:-93%	
		Max. increase: n.a.	
Fluoride (F)			
Stuyfzand *et al.* (2006)	0.2 mg/l	Max. decrease: -65%	84 m, oxic passage
Stuyfzand *et al.* (2006)	0.15 mg/l	Max. increase: +100%	100 m, anoxic
Selenium (Se)			
Stuyfzand *et al.* (2006)	0.2 µg/l	Max. decrease:-95%	100 m, anoxic
		Max. increase: n.a.	

n.a. = no information available

The interplay between water phase and solid phase is complex so that predictions on removal rates are often difficult and reasons for limitations are not obvious. The drilling of groundwater wells in Bangladesh, for instance, and the arsenic epidemic is to date not fully explained. It has been hypothesized that the introduction of oxygen by extensive water abstraction had mobilised pyrite-bound arsenic by oxidation (Höll, 2002). Others proposed that under the anoxic conditions that followed aerobic degradation, iron(hydroxy-)oxides were microbially reduced to siderite ($FeCO_3$) in conjunction with reduction of arsenic (V), whereby the toxic arsenic (III) became remobilised (Jönsson & Sherman, 2007). In both scenarios, a shift in redox conditions would be the driving force behind arsenic mobilisation into the water phase.

Aside from unfavourable redox conditions, colloidal transport does affect the retention of adsorbed trace elements (e.g. Pb, Cd) (Bourg *et al.,* 1989; Christensen *et al.*, 1999). Thus, coarse aquifers unsuitable to retain colloids are therefore also likely not apt to retain inorganic trace elements.

Probably the most suitable conditions for the avoidance of undesired trace elements are found in oxic aquifers and in the presence of organic matter (Schmidt *et al.,* 2003), both due to sorption and precipitation processes. Under oxic conditions, iron and manganese are stable as hydroxy(oxides) to which other inorganic trace elements (e.g. Cd, Cu, Zn, As, F, Ni) may adsorb or co-precipitate.

Summary Inorganic Trace Elements
For inorganic trace elements there is a potential for removal during subsurface passage. More important, however, is their ability to be mobilized from the subsurface due to changing redox conditions. Oxic subsurface passage is generally deemed more suitable for the immobilization of inorganic trace elements due to the co-precipitation with iron and manganese hydroxy(-oxides). Given that there is potential for removal, it is best evaluated substance and site-specifically. Nevertheless, it needs to be taken into account that the soil matrix serves as sink and source.

Chapter 4
Summary and Conclusions

Experiences gained from the detailed investigation of BF and AR sites in Berlin during the NASRI project together with international experiences in the field of bank filtration and aquifer recharge have given rise to the following conclusions regarding their utilization for drinking water production:

- in Germany, as in other countries in Central Europe, BF has been implemented for drinking water production as the “second best solution”, following protected groundwater sources. This general principle is followed in order to achieve the highest possible number of safety barriers. If possible, direct surface water use is therefore avoided. This has led to a high number of BF sites – and in case of limited availability AR sites – in these regions.
- the 170 sites compiled show a high degree of variation:
  - capacity: 70 to 780 000 m^3/d
  - aquifer thickness: 3 to 300 m
  - aquifer hydraulic conductivity: $< 1*10^{-4}$ to $5.7*10^{-01}$ m/s
  - distance bank – well:< 10 to 3,500 m
- post-treatment (and pre-treatment for AR-sites) needs to be taken into account. Depending on source water quality, travel time and redox conditions a typical treatment chain would include:
  - for AR sites: particle elimination as pre-treatment for clogging reduction,
  - for BF and possibly also AR sites - after recovery: iron- and manganese removal (e.g. via aeration and sand filtration)
  - for BF and AR sites - depending on source water quality and protection:
    - advanced post-treatment (e.g. ozonation, GAC)
    - final disinfection
- International regulations and technical guidelines for BF sites concentrate on i) general source water protection and ii) describing certain pre-requisites for substance elimination (e.g. 50-days-line in Germany or pre-requisites for protozoa removal in the US LT2ESWTR). For AR sites in Europe the quality and quantity of infiltrated water needs to comply with the Water Framework Directive and Groundwater Directive, i.e. no deterioration of the water status may take place. General rules are lacking, but may also not be applicable due to the site specifity. Different approaches are currently discussed in Europe, the US and Australia (e.g. via risk assessment).
- Concerning the performance of bank filtration and aquifer recharge performance for substance removal derived from investigations at existing field sites, different classes of substances / substance groups can be distinguished:
  - Substances or substance groups, for which high removal is possible (> 90 %):
    - Observed removal of **suspended solids** at BF & AR sites ranges from 92 % to 99.9 % with an average of 97.6 %. In all documented cases the threshold values for drinking water of 1 NTU in average is met. Monitoring turbidity can provide valuable information on the performance of the filtration process over time (e.g. regarding removal of algae or pathogens).

  - Subsurface passage can present an important or even main barrier towards **pathogens** for drinking water production. Crucial factors for the functioning of this barrier towards pathogens are source water protection, sufficient travel times (> 50 d), a fine and homogeneous aquifer matrix and low flow velocities. Observed removal at field sites ranges from
    - from 2.1 to 8-$\log_{10}$ for viruses
    - from 3.2 to 6-$\log_{10}$ for non-spore forming bacteria
    - from 0.4 to > 4.9-$\log_{10}$ for spore-forming bacteria
    - from 0.9 to 2.5-$\log_{10}$ for protozoan (oo)cysts
  - Predominantly cell-bound **cyanobacterial toxins** (e.g. microcystins) show up to 99 % removal during subsurface passage. Limitations can occur in case of high extra-cellular concentrations, coarse and unsorted aquifer material, anaerobic conditions, low temperatures and short travel times.
  - During absence of oxygen and presence of sufficient carbon sources **nitrate** is rapidly denitrified during BF & AR and can no longer be detected (> 97 % removal) even after contact times of only 2 days. Factors that limit denitrification are:
    - oxic conditions (promoted by short travel times, high flow velocities and low DOC),
    - insufficient organic carbon sources.
  - For the major group of DBPs, **trihalogenmethanes** (THMs) anoxic to anaerobic conditions are favourable for removal. Under these conditions a removal of 85 % to more than 95 % can typically be observed.
- Substance groups, including substances that show highly variable removal (typically 25 to 75 % overall removal)
  - **Dissolved organic carbon** (DOC) is typically removed by 39 to 50 % during subsurface passage in BF and AR systems. Biodegradable DOC (e.g. polysaccharides) is removed during the first few meters of infiltration so that bank filtrate exhibits a low re-growth potential. Factors counteracting maximum removal are:
    - short travel times,
    - anoxic or anaerobic conditions,
    - high background DOC in source or ambient ground water with substantial shares of humic substances
    - high amounts of organic carbon in the aquifer matrix.
  - For **pesticide** removal no general recommendation can be given, due to the fact that this group comprises substances of highly variable chemical structure. In addition, redox conditions play an important role for the removal of some types of pesticides. However, even for persistent pesticides a smoothing of seasonal peaks can be expected due to dispersion within the aquifer. Pesticides of which field studies have shown limited removal (< 50 %) in BF & AR systems are:
    - triazines: atrazine, desethylatrazine, desisopropylatrazine, cyanazine, propazine, simazine, terbutylazine
    - ureas: chlortoluron, diurone, isoproturone
    - phenoxy-herbicides: mecoprop (under anoxic conditions)

        - organochloride insecticides: hexachlorbenzene (HCB), pentachlorophenole (PCP),
        - anilides / anilines: dimethenamid, metolachlor
        - others: bentazone (plus its transformation product 2-amino-n-isopropylbenzamide), glyphosate, dikegulac
    - For **pharmaceuticals and X-ray contrast media** many substances show good removal during BF & AR. Some PhACs (e.g. carbamazipine) show redox-dependant removal. Pharmaceuticals and X-ray contrast media of which field studies have shown limited removal (< 50 %) in BF & AR systems are:
        - regardless of redox zone: sulfadimidine, primidone, AMDOPH, carbamazipine (< 20 % under oxic conditions), AOI (< 31 % under oxic conditions)
        - under oxic conditions: sulfamethoxazole
        - under anoxic conditions: phenazone, clindamycine
    - Whereas less soluble **CHC**s can be removed by up to > 99 % at BF & AR sites, highly soluble CHCs tend to show little removal, especially under oxic conditions (0 – 67 %). There are indications that anoxic conditions are favourable for the removal of highly soluble CHCs like tetrachloroethylene (PCE). CHCs that have shown to break through in existing BF & AR systems are:
        - 1,1,1-trichloroethane (TCA), trichloroethylene (TCE) and tetrachloroethylene (PCE)
    - Median values of observed removal of **monocyclic aromatic hydrocarbons** ranges from 48 % (benzene) to 81 % (toluene). Polycyclic aromatic hydrocarbons (**PAH**s) are generally less mobile in aquatic systems due to their affinity to organic aquifer matter. Substances likely to be poorly removed (< 50 %) in BF & AR systems are:
        - Monocyclic aromatic hydrocarbons: benzene, m-, o-, and p-xylene,
        - Polycyclic aromatic hydrocarbons (PAH): phenanthrene, flouroanthrene.
    - Though there is little published data from field sites, it appears that many **endocrine-disrupting compounds** (e.g. bisphenol-A, estron) are removed by more than 90 % by BF/ AR systems. For phtalates and some nonylphenolic compounds, however, removal might be limited (< 50 %). As for all organic micropollutants, the toxicity (here: endocrine-disrupting effect) and persistence of transformation products is likewise to be taken into account.
    - Of the **other organic trace substances** investigated during the NASRI project the following have shown to be only poorly (< 30 %) removed by BF & AR, regardless of redox zone:
        - 1,5-NDSA, EDTA, MTBE
    - Limited removal (< 50 %) was also observed for
        - 1,7-NDSA and 2,7 NDSA (with slightly enhanced removal under oxic conditions).
- Substances which are generally poorly or not removed by BF & AR systems or may even become mobilized
    - High removal (> 99 %) of **ammonium** from surface water by subsurface passage is limited to strictly aerobic conditions and concentrations well below 6 mg/L under saturated flow conditions. On the contrary, for scenarios with high shares

of organic N either in source water (e.g. through high shares of untreated sewage) or within the aquifer matrix an increase of ammonium levels has to be expected.

- **Major anions and cations** are usually not retained during BF & AR. On the contrary, due to calcite dissolution increasing concentrations of Ca^{2+}, Mg^{2+} and HCO_3^- are possible. Relevant sulfate reduction can only be expected after travel times of > 50 d in aquifers with high shares of organic material under strictly anaerobic conditions. The composition of the raw water in BF & AR systems concerning major anions and cations will be primarily determined by the mixing ratio of bank filtrate and ambient groundwater.
- For **inorganic trace elements** there is a potential for removal during subsurface passage. More important, however, is their ability to be mobilized from the subsurface due to changing redox conditions. Oxic subsurface passage is generally deemed more suitable for the immobilization of inorganic trace elements due to the co-precipitation with iron and manganese hydroxy(-oxides). Given that there is potential for removal, it is best evaluated substance and site-specifically. Nevertheless, it needs to be taken into account that the soil matrix serves as sink and source.

The efficiency of BF and AR sites depends strongly on the hydrogeological setting as well as on source water quality. Substance removal and potential mobilization need to be taken into account. In every case, however, it represents an additional barrier for drinking water production from surface water, thus adding substantially to the security and sustainability of the water supply system.

Chapter 5
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Appendix B

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