Water Resource Quality (WRQ)

Geogenic Contamination Handbook

Addressing arsenic and fluoride in drinking water

WHO Collaborating Center
for Sanitation and Water
in Developing Countries

eawag
aquatic research

7 Mitigation options

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It can be very difficult to determine the best technological approach for providing water free of arsenic and fluoride. Often people think first of contaminant removal technologies, but it may be more cost-effective and sustainable to exploit alternative water resources. In either case, some sort of water treatment is likely to be necessary to ensure both chemical and microbial water safety. A wide range of technological options are available at different scales: in professionally managed centralised plants, in small community-scale systems or at the household level. Each of these scales has advantages and disadvantages, and the most suitable solution is determined by the local context (Table 7.1).

Introducing a new technology is a complex process, which should be participatory, involving all stakeholders from the outset. The institutional framework, legislation, funding, support and long-term financing needs to be determined (Chapters 5 and 6), as is promoting safe water use among the affected population and facilitating behaviour change (Chapter 8). Insufficient operation and maintenance (O&M) can quickly lead to technological failure, so these aspects need to also be planned and considered before the technology is installed. The Operation and Maintenance Network gives useful tools and information on this issue. Detailed information on the whole process of supporting sectors in scaling up WASH technology is presented in the Technology Applicability Framework (TAF) of the WASHTech project.

Water Safety Plans

Water Safety Plans (WSP) can provide a systematic means to address and manage health-related water risks. They provide a practical framework to implement a systematic, risk-based approach to most effectively ensure consistent supplies of safe drinking water. The WSP approach requires that hazards and associated risks be identified in the entire water supply chain, from catchment to point of use, and it gives a framework for the prioritisation and management of those hazards and risks (Bartram et al., 2009; WHO, 2012; WHO/IWA, 2013). WHO and its partner organisations, including the International Water Association (IWA), actively support the WSP approach. Several tools exist to assist in the development and implementation of WSPs (WHO 2012; WHO/IWA 2013).

WHO (2012) Water safety planning for small community water supplies: step-by-step risk management guidance for drinking-water supplies in small communities. World Health Organization, Geneva, Switzerland.

WHO/IWA (2013) Water safety plan quality assurance tool. World Health Organization, Geneva, Switzerland.

Table 7.1 *Drinking-water treatment at different scales*

Scale	Advantages	Disadvantages
Centralised 	• Process parameters can be controlled and optimised. • There may be economies of scale, but these are counterbalanced by increasing costs of large distribution systems.	• Requires large capital investments and incurs significant recurring costs. • Requires trained personnel and constant operation and maintenance. • Difficult to extend to areas of low population density. • Risk of low community inputs and support. • Potential of microbial contamination during distribution and collection.
Community scale 	• Processes can be regulated and optimised better than at household scale. • Relatively inexpensive. • Demand-responsive: can be designed for local needs. • With community leadership and support, sustainability may be greater.	• Processes cannot be regulated and optimised to the same extent as in centralised schemes. • Limited capacity for operation and maintenance. • Potential of microbial contamination during distribution and collection.
Household 	• Takes advantage of existing water supply infrastructure (e.g. boreholes). • Allows targeting of people most at need. • Relatively easy and inexpensive to implement.	• Systems may not be operated correctly. • Lifetime of chemical removal filters is difficult to predict, so it is hard to know when replacement is needed. • Effective replacement requires supply chain and motivation. • Routine monitoring is a challenge. • Some populations can easily be excluded due to lack of information or financial resources.

Difficult questions to answer:

- Which water resource should be developed? Is it better to remove the chemical or to find a chemically safe resource?
- Which technology is best suited for water treatment in this particular setting?
- On which scale can this technology best be applied?

Answers should be based on the combined understanding of available water resources, institutional setting (Chapter 5), financing strategies (Chapter 6) and acceptability (Chapter 8). Those responsible for water supply often have to make choices between these different approaches without a solid evidence base and sometimes without a clear method for taking decisions. A list of factors for the comparative evaluation of technologies is given below (Fig. 7.1).

Robustness
- Frequency of major interventions
- Maturity of technology

Affordability
- Capital costs
- Capital maintenance costs
- Operational costs

Availability
- Raw materials
- Spare parts
- Qualified personnel
- Organisational support

Back-up
- Training
- Monitoring
- Maintenance

Ease of use
- Simplicity of operation
- Requirement of electricity

Acceptability
- Cultural aspects
- Water aesthetics
- Water standards
- Waste management

Fig. 7.1   Selected criteria for technology evaluation

Note: The choice of technology heavily depends on local conditions. A filtration technology may be suitable for water with low contamination, whereas the same technology may be too expensive for highly contaminated water. In another region, salinity or industrial contamination may require the use of alternative water resources etc.

Water treatment: A fundamental difference between arsenic and fluoride

In geogenically contaminated water, arsenic concentrations can range from >10 to around 500 mg/L, while fluoride concentrations can be orders of magnitude higher, generally ranging from >1.5 to 20 mg/L.

Filtration is a frequently used water-treatment technology. Since fluoride concentrations are so much higher than arsenic concentrations, more frequent regeneration and replacement of filter material is necessary, and the water treatment costs are subsequently higher.

7.1 Exploiting alternative water resources

The provision of drinking water from alternative sources that are not contaminated with arsenic and fluoride has proven to be a popular mitigation option. In Bangladesh, for example, "well switching" is most commonly used for mitigation of arsenic contamination. The underlying reason for this is the difficulty, in terms of acceptance, supply, monitoring, maintenance and overall cost, in establishing technologies to remove contaminants. Therefore, before efforts are made to treat contaminated water, it is worthwhile to determine whether alternative water resources are available.

Resource availability is a question of scale and thus of institutional engagement:

Regional-scale solutions may be sought by government agencies that need to provide water not only for drinking, but also for agriculture and industry. This may include the provision of piped drinking water derived from surface water or groundwater.

Many water resource tools of differing degrees of sophistication have been developed to support planning and implementation. One central theme is Integrated Water Resources Management (IWRM), a planning and implementation tool for managing water resources for different uses, including agriculture, industry, personal use, recreation and ecosystem protection. See the website of the Global Water Partnership (GWP and UN Water) for more information and downloadable resources.

Local-scale solutions may include rainwater harvesting, making use of uncontaminated groundwater from different locations in the aquifer by "well switching" or the treatment of local surface-water resources, such as rivers, lakes or ponds.

Here the focus is on ensuring that microbial contamination does not replace geogenic contamination as a health problem, since groundwater is often selected as a replacement for microbially contaminated surface waters. Water storage is another important issue. Infrastructure is required to collect, treat and deliver drinking water to consumers. "Household Water Treatment and Safe Storage" is a strategy for making surface-water sources safe in resource-poor settings (see section below). Numerous texts provide guidance on the exploitation of surface water, groundwater and rainwater for drinking; see the References section for a small selection.

Surface water

Surface water is the water found in rivers and lakes. Surface water is replenished naturally by precipitation and is "lost" naturally through discharge to the seas and oceans, by evapotranspiration, by evaporation and by sub-surface seepage. Although the only natural input to any surface-water system is precipitation within its watershed, the total quantity of water in that system at any given time is also dependent on many other factors. These factors include storage capacity in lakes, wetlands and artificial reservoirs, the permeability of the soil beneath these storage bodies, the runoff characteristics of the land in the watershed, the timing of the precipitation and its interaction with groundwater, and local evaporation rates. All of these factors also affect the proportions of water lost.

Although surface water is seldom contaminated by arsenic and fluoride, it nearly always requires treatment to improve the microbial water quality. Pathogens differ in their susceptibility to various treatments. For example, Cryptosporidium cysts may be retained by filters but are resistant to chlorination; the opposite is true of many viruses. Furthermore, all treatment systems are subject to occasional failures which may not be recognised by the operators. The key to developing a robust and reliable system for providing safe water is to implement multiple barriers for pathogen control. Different pathogens can be removed in different stages, according to their particular weaknesses, resulting in water of progressively higher quality. The multiple-barrier approach protects against the transmission of pathogens in the event that one barrier should fail. A typical multiple-barrier system for treating surface water might include sedimentation, some type of filtration (multi-stage filtration, slow sand filtration or coagulation followed by rapid filtration) and disinfection.

Numerous texts provide guidance on the design of treatment plants that can be used for conventional drinking-water treatment. An excellent starting point, available for free download on the internet, is the "Small community water supplies" (IRC, 2002). The IRC in 2006 also produced a detailed report on multi-stage filtration (IRC, 2006).

Groundwater

Groundwater is water that fills the cracks and spaces between underground rocks and sediments. Underground rocks and sediments that hold substantial amounts of water are called aquifers – these can gain water from, or lose water to, surface water bodies. Sometimes it is useful to make a distinction between shallow aquifers that are closely associated with surface water and deep aquifers that are isolated from the surface, containing what is sometimes called "fossil water".

A critical factor in the use of groundwater is that abstraction rates need to be lower than replenishment rates. In arid climates, replenishment rates may be very low. This results in a lowering of the groundwater table.

Because of natural filtration through sediments, groundwater is typically of a much higher microbial quality than surface water. However, groundwater is not necessarily free from pathogens: especially where aquifers are near the surface and water tables are high, sediments contain little silt, and clay and on-site sanitation is widely practised, groundwater is vulnerable to contamination. While groundwater is often distributed and consumed without treatment, safety disinfection (e.g. chlorination) would be recommended in such settings (ARGOSS, 2001).

Since aquifers by their nature allow long contact periods between pore waters and rocks and sediments, groundwater frequently has higher levels of dissolved minerals than does surface water or rainwater. Under the right geochemical conditions, different elements can reach undesirable levels in groundwater. This manual describes contamination with fluoride and arsenic in detail, but other elements commonly found in groundwater can include sodium and chloride (major components of salinity), calcium and magnesium (which make up hardness), and iron and manganese (metals which can stain materials and give an unpleasant taste to water).

Removal of salinity and hardness is complicated and relatively expensive. However, simple sand filters can be optimised to remove iron and manganese, as described in Hartmann (2001).

Even though groundwater extracted from one aquifer may be contaminated with arsenic or fluoride, other aquifers (deeper or shallower) in the same area may provide completely uncontaminated water. This could be due to differences in the mineralogy of the aquifer material or changes in dissolved oxygen concentrations, which can influence the mobility of redox-sensitive contaminants such as arsenic. A classic example of this is the widespread geogenic arsenic contamination in deltaic areas of Bangladesh. Here, shallow wells in young sediments under reducing conditions yield very high arsenic concentrations, whereas deep tube wells usually provide water with a completely different chemistry, with little arsenic (Hug et al., 2011).

A vast number of technologies exist for the abstraction of groundwater. These are described in a range of resources and manuals. A good overview of water-lifting devices is given in WHO/IRC (2003) and Baumann (2000). In addition, UNESCO has produced several documents describing groundwater resources. Particularly useful are "Groundwater resources of the world and their use" (UNESCO/IHP, 2004) and "Non-renewable groundwater resources: A guidebook on socially-sustainable management for water policy makers" (UNESCO, 2006).

Rainwater

Rainwater is the ultimate source of all drinking water in the long term, since it replenishes both surface water and groundwater. Rainwater can also be captured directly and used as drinking water. However, rainwater is highly variable in its spatial and temporal distribution, so the use of rainwater for drinking often requires significant storage or distribution capacity. Whether rainwater harvesting is viable in a certain region depends very much on the yearly amount and distribution of rainfall. Rainwater is a main drinking-water source for relatively few people, but in some settings on ocean shores or islands, it can be the only source of drinking water.

Rainwater is free from pathogens, at least until it reaches the ground, and except in some urban areas, is of excellent chemical quality. When properly collected and stored, rainwater can provide a safe and acceptable source of drinking water for at least part of the year. Rooftop water harvesting has been extensively researched by the Development Technology Unit of the University of Warwick, which has produced an excellent handbook on the topic of "Roofwater harvesting: A handbook for practitioners" (Thomas et al., 2007). A wealth of additional information on rainwater harvesting can be found at the SSWM portal: Rainwater Harvesting (Rural).

Household water treatment and safe storage

Regardless of its source, drinking water can easily become contaminated with pathogens through unhygienic distribution, collection, handling and storage (Wright, et al., 2004). One approach to minimising the adverse health impacts of such contamination is to promote microbial treatment at the household level, or Household Water Treatment, combined with safe storage (HWTS).

A growing body of evidence demonstrates that the use of HWTS methods improves the

microbial quality of household water and reduces the burden of diarrhoeal disease in users (Fewtrell et al., 2005; Clasen et al., 2007; Waddington and Snilstveit 2009). Several HWTS methods have been proven to improve drinking-water quality significantly, both in the laboratory and in field trials in developing countries (Clasen et al., 2007; WHO, 2011). These HWTS methods include filtration, chemical disinfection, disinfection with heat (boiling, pasteurisation) and the use of flocculants and/or disinfectants. The role of the International Network on Household Water Treatment and Safe Storage (the "Network") is in part to coordinate the effective implementation of such options. The Network, established in 2003 by WHO, and as of 2011 co-hosted by WHO and UNICEF, includes over 100 international, governmental and non-governmental organisations, private sector entities and university research departments that are actively involved in household water treatment and safe storage policy, research, implementation, monitoring and evaluation.

Additional resources can be found in the WHO/UNICEF toolkit (WHO/UNICEF 2012) and at the SSWM portal (Sustainable Sanitation and Water Management Toolbox).

7.2 Arsenic treatment technologies

Technologies for arsenic removal rely on basic physical and chemical processes that are summarised in the following sections. More details can be found in the scientific literature and more information and references in one of the several reviews of arsenic removal technologies (e.g. Mohan and Pittman, 2007).

The review here focuses on decentralised (community or household) arsenic removal methods. Particular emphasis is on technologies which have been validated through independent verification programmes (Johnston, 2002; USEPA, 2005). The following chapters present and summarise the principal steps and procedures for arsenic removal.

Pre-treatment (oxidation)

Arsenic in groundwater is mainly present in two oxidation states, As(III) and As(V), depending on the environmental conditions in the aquifer. Most arsenic removal technologies are most effective at removing As(V) (arsenate), since As(III) (arsenite) is predominantly non-charged below pH 9.2. Therefore, many treatment systems include an oxidation step to convert arsenite to arsenate. Oxidation alone does not remove arsenic from solution; it must be coupled with a removal process such as coagulation/precipitation, adsorption or ion exchange.

Air oxidation

Atmospheric oxygen is readily available as an oxidising agent; however, the kinetics of air oxidation of arsenic are very slow (taking weeks), and the reaction needs to be catalysed. Metals such as iron or manganese, which are naturally present in groundwaters, catalyse the oxidation of As(III), but oxidation is normally not complete without additional oxidants or the repeated addition of Fe(II).

Chlorine

Chlorine is widely available and is a rapid and effective oxidant for arsenite. Dosing can be difficult, since locally available chlorine can be of uncertain quality in developing countries. When enough chlorine is added for effective disinfection of water from microbial contamination, arsenite oxidation is normally complete. Doses generally range from 1.0 to 5.0 mg/L, with the goal of approximately 0.5 mg/L residual chlorine to provide protection against microbial contamination after treatment.

Manganese compounds

Potassium permanganate (MnVII) effectively oxidises As(III), along with Fe(II) and Mn(II). Filtration of water through a bed of solid Mn(IV) oxides can rapidly oxidise arsenite to arsenate without the need for adding a liquid or gas oxidant. Oxidation is efficient over a wide range of pH and does not release excessive manganese into solution.

Other more advanced oxidants (e.g. ozone, ultraviolet lamps) are not considered here, as they are difficult to use in developing countries.

Adsorption and ion exchange

Ion exchange is a reversible chemical reaction between an insoluble solid and a solution during which ions may be interchanged. The ions can be relatively easily exchanged. Adsorption, on the other hand, involves the formation of a bond between a dissolved ion and the solid-phase surface. These bonds are not so easily broken. Various solid materials have a strong affinity for dissolved arsenic. Arsenic is strongly attracted to sorption sites on the surfaces of these solids, and is effectively removed from solution.

Ion exchange resins

Ion exchange is a physico-chemical process by which an ion in the solid phase is exchanged for an ion in the feed water. The solid phase is typically a synthetic resin which has been chosen to preferentially adsorb the particular contaminant of concern. To accomplish this exchange of ions, feed water is continuously passed through a bed of ion-exchange resin beads in a down-flow or up-flow mode until the resin is exhausted. A good example is the READ-F ion exchange filter (Fig. 7.2).

Fig. 7.2 *READ-F household ion-exchange filter used in Bangladesh (see also document on "Verified Arsenic Removal Technologies in Bangladesh")*

Most commonly, the resins are composed of a matrix of polystyrene cross-linked with divinylbenzene. Charged functional groups are attached to the matrix by covalent bonding. These functional groups determine the resin's affinity to certain ions such as arsenate. Conventional sulphate-selective resins are particularly suited for arsenate removal. Nitrate-selective resins also remove arsenic, but arsenic breakthrough occurs earlier (USEPA, 2003c). Only arsenate can be removed using ion-exchange filters, as arsenite is not charged. A pre-oxidation step might therefore be necessary.

Arsenic removal: Various strong-base anion exchange resins are commercially available which can effectively remove arsenate from solution, producing effluent with less than 1 µg/L arsenic (Clifford, 1999). Arsenate removal is relatively independent of pH and influent concentration. On the other hand, competing anions, especially sulphate, can have a strong effect. In low-sulphate waters, ion-exchange resins can easily remove over 95% of arsenate and treat from several hundred to over a thousand bed volumes, before arsenic breakthrough occurs. However, when sulphate is present and saturates the exchange sites, it can lead to desorption of large amounts of exchanged arsenate – so-called "arsenic dumping". Accordingly, the USEPA recommends that ion-exchange resins only be used for low-sulphate waters (USEPA, 2000b).

Regeneration: Exhaustion occurs when all sites on the resin beads have been filled by contaminant ions. At this point, the bed is regenerated by rinsing the column with a regenerant, a concentrated solution of the ions initially exchanged from the resin. The number of bed volumes that can be treated before exhaustion varies with resin type and influent water quality (USEPA, 2000b). Ion-exchange resins are easily regenerated by flushing with concentrated salt solutions (1.0 M NaCl is commonly used). Brine can be reused 20–30 times, in spite of increasingly concentrated

arsenic levels in the regenerant. Spent regenerant is loaded with arsenic and needs to be treated or disposed of safely (USEPA, 2000d).

A hybrid anion exchanger (HAIX) containing hydrous ferric oxide has been used to remove arsenic from drinking water in West Bengal for 10 years now. The initial investment in this material appears to be offset by the long filter life. Please see German et al. (2014) for further details.

Advantages

- High adsorption capacity
- Commercially available
- Regeneration possible

Disadvantages

- Moderately expensive
- Risk of "arsenic dumping" of waters with high sulphate concentrations
- Interference from sulphate and total dissolved solids
- Water rich in Fe and Mn might require pre-treatment to prevent filter clogging
- Regeneration produces arsenic-rich brine

Activated alumina

Activated alumina (AA) is a commercially available granular form of aluminium oxide which can be used as a filter medium to remove a range of contaminants from water, including arsenic. The contaminant ions are exchanged with the surface hydroxides on the alumina. When adsorption sites on the AA surface become filled, the bed must be regenerated. Activated alumina has a much higher affinity for As(V) than for As(III). Therefore, depending on the prevalence of As(III), filtration might need to be preceded by an oxidation.

Arsenic removal: The arsenic adsorption capacity of AA (mg As/g AA) varies significantly with water pH and influent arsenic concentrations and speciation. Arsenate removal capacity is highest within a narrow range of solution pH from 5.5 to 6.0, in which the alumina surfaces are protonated, and in which other anions are not concentrated enough to compete with arsenic (USEPA, 2000b). In large systems, pH adjustment is often applied to optimise treatment.

Regeneration: Regeneration of AA beds is usually accomplished using a strong basic solution of concentrated NaOH. Arsenic is more difficult to remove during regeneration than other ions such as fluoride. Therefore, higher base concentrations are used, typically, 4% sodium hydroxide. After regeneration with strong base, the AA medium must be neutralised using strong acid (e.g. 2% sulphuric acid). Arsenic-rich wastes must be processed before disposal (USEPA, 2001).

Advantages

- High arsenic removal efficiency
- Commercially available
- Regeneration possible
- Tested in community and household application

Disadvantages

- Moderately expensive
- Strong acid and base needed for regeneration
- Arsenic-rich waste produced
- Optimal arsenic removal within a limited pH range

Iron-based solids

Iron, especially in the ferric state (Fe(III)), has a strong affinity for arsenic. It also has an affinity for other ions. Phosphate, arsenate and silicate bind equally strongly, followed by negatively charged ions (Balistrieri and Chao, 1990; Hsu et al., 2008; Hug, 2014):

phosphate = arsenate ≈ silicate > (bi)carbonate > humic acid >fluoride > sulphate > chloride

This sequence indicates that arsenic will compete for binding sites with phosphate and silicate, but not with ions such as fluoride, sulphate or chloride.

Fig. 7.3   *SIDKO community arsenic removal filter installed in Bangladesh*

Granular iron-based media have been developed relatively recently for arsenic removal (e.g. Driehaus et al., 1998). Several commercial iron-based materials are available, including granular ferric hydroxide (e.g. AdsorpAs®, see SIDKO filter, Fig. 7.3). Iron-

based solids can effectively remove arsenate, arsenite and phosphate from water. Before the water is passed over the active medium, it is aerated and pre-filtered to oxidise and remove iron flocs (USEPA, 2003b).

Sands coated with iron oxides have been synthesised by various researchers and tested for their arsenic removal capacity. UNESCO-IHE has developed a household filter which uses coated sand from Dutch iron removal plants (Petrusevski et al., 2008).

Advantages
- High arsenic removal efficiency
- Works well over a broad range of pH
- Removes both As(V) and As(III): pre-oxidation may not be needed
- Commercially available

Tested in community and household application

Disadvantages
- Moderately expensive
- Regeneration is possible but usually not done
- Arsenic-rich waste produced

Zero-valent (metallic) iron

When metallic, or zero-valent, iron corrodes, it produces dissolved ferrous iron (Fe(II)). The ferrous iron reacts with oxygen to form ferric iron (Fe(III) that precipitates as iron hydroxide (Fe(OH)$_3$), which acts as a sorbent for arsenic. Reactive oxygen species produced during iron corrosion also oxidise As(III) to the more strongly sorbing As(V) (Leupin and Hug, 2005). A household filter (the_SONO_filter, Fig. 7.4) has been developed which makes use of metallic iron to remove arsenic from drinking water in Bangladesh (Hussam and Munir 2007). This filter consists of two buckets placed on top of each other, with the top bucket containing sand, iron filings and brick chips and the bottom bucket containing sand, charcoal and brick chips. It has been verified through the BETV-SAM programme (see document on Verified_Arsenic_Removal_Technologies_in_Bangladesh).

Fig. 7.4   *SONO filter using metallic iron for arsenic adsorption*

Advantages
- High arsenic removal efficiency
- Continuous generation of ferric adsorption sites prolongs filter lifetime
- Removes both As(V) and As(III)
- Relatively inexpensive

Disadvantages
- Iron corrosion may lead to clogging and low filtration rates
- Limited field experience, mainly in household filters
- Limited commercial availability
- Arsenic-rich waste produced

Choice of filter medium

The choice of filter medium is primarily related to its use.
- Ion exchangers and granular ferric oxides, though relatively expensive, remove arsenic quickly and can be used for high throughput situations providing that As(III) has been oxidised.
- Filters using metallic iron, which also oxidises As(III), are less expensive and need to be run slowly and are more suited to household or community filters with limited water volumes.

Since contamination levels for arsenic in drinking water are in the microgram per litre range, the capacity of a filter for arsenic is a secondary issue (unlike for fluoride).

Precipitation, co-precipitation and coagulation

Precipitation methods reduce dissolved arsenic concentrations by the precipitation of low-solubility solid minerals such as calcium arsenate. But these cannot normally lower arsenic to drinking-water limits. Co-precipitation refers to the precipitation of solid particles in the arsenic-containing water – normally aluminium or iron (hydr)oxides – that can sorb and incorporate arsenic.

Coagulation is the clumping of fine particles in solution to larger ones that can settle. Metal salts, such as alum, ferric chloride or ferric sulphate, are widely used coagulants to remove arsenic from drinking water (USEPA 2000a). These salts initially dissolve upon addition to water and then rapidly form fine precipitated flocs of metal hydroxides. These flocs coagulate and settle out of solution, scavenging many dissolved and particulate materials in the process. Vigorous stirring is required immediately after coagulant addition to ensure uniform mixing. Once the coagulant is dispersed, slow mixing allows the flocs to collide and grow (flocculate) without breaking up. Much of the floc matter will settle by gravity, but filtration is essential to remove small particles which can remain in suspension, as these can contain significant amounts of arsenic. If water is soft and of low alkalinity, it may be necessary to increase alkalinity (e.g. by adding lime addition) to ensure good floc formation.

Alum ($Al_2(SO_4)_3$) is effective for removing As(V) but ineffective for As(III), so pre-oxidation is often necessary. Alum has a narrow effective range, from pH 5–7; if the pH is above 7, removal may be improved by adding acid to lower the pH. Typical doses are 10 to 50 mg alum per litre.

Ferric (Fe(III)) salts (e.g. $FeCl_3$ and $Fe_2(SO_4)_3$) coagulate best between pH 5 and pH 8. Typical doses are 5 to 50 mg/L ferric salts. Ferric salts can remove both As(III) and As(V), but As(V) is retained more strongly, so pre-oxidation is often carried out.

Ferrous (Fe(II)) salts (e.g. $FeSO_4$) can also be used to remove arsenic, but oxygen (in air) and time are required to let the Fe(II) oxidise to Fe(III), which forms the arsenic-sorbing Fe(III) (hydr)oxide particles. At pH 7, it takes 1–4 hours for Fe(II) to oxidise completely to Fe(III) and to precipitate. Less time is required at a higher pH. During the oxidation of Fe(II) to Fe(III) by oxygen from air, a part of the As(III) is also oxidised to As(V), so the overall removal of As(III) with Fe(II) is better than with Fe(III), if no additional oxidant is used (Roberts et al., 2004). Groundwater often contains naturally dissolved Fe(II). If the natural concentration of Fe(II) is high (>15 mg/L), then this Fe(II) alone might be sufficient to remove the arsenic.

Coagulation also improves turbidity and colour and can also reduce levels of organic matter, bacteria, iron, manganese and fluoride, depending on operating conditions. If concentrations of phosphate or silicate in the source water are high, coagulation may be less effective.

Coagulation is operationally complex and is more commonly practised in centralised water-treatment plants. Chile has been removing arsenic from drinking water by coagulation for a long time – in 1970, the world's first arsenic removal plant was constructed along the Toconce River. Since then, numerous plants have been built in Chile, most of which use ferric chloride coagulation with chlorine pre-oxidation (Sancha,

2006).

Some household coagulation systems have been developed, typically using an upper bucket for coagulation and flocculation and a lower bucket with filter material (e.g. charcoal and sand) for the removal of suspended solids, including metal (oxy)hydroxide particles containing arsenic (e.g. Cheng et al., 2004). The performance of the Shawdesh_Aqua_Filter, a two-bucket system using ferric sulphate, was verified in the Bangladeshi BETV-SAM project (see "Verification Programmes" below).

Electrocoagulation, in which aluminium or iron flocs are produced by passing a current through metal plates in contact with the water to be treated, is an emerging technology. Electrocoagulation offers certain advantages over conventional treatment with salts: removal of As(III) may be superior due to at least partial oxidation, the need for chemical supply and addition is greatly reduced and sludge volumes are smaller (e.g. Kumar et al. 2004; Emamjomeh and Sivakumar 2009a). As electrocoagulation is a relatively new approach for the removal of arsenic (and fluoride), current research is focusing on optimising the many design factors which can influence treatment efficiency and cost (Addy et al., 2011).

Common to all (co)precipitation techniques are:

Disposal: The use of coagulants produces arsenic-rich sludge which needs to be safely disposed of, away from drinking-water sources (USEPA, 2000d). Wastes may be thrown into latrines that are well separated from drinking-water wells. However, centralised landfilling is probably the best disposal route.

Costs: Coagulation using metal salts requires simple chemicals that are readily available and cost-effective. Filter material generally consists of sand and charcoal, materials which are also cheap and easy to obtain.

Advantages
- Relatively inexpensive
- Simple chemical reagents, widely available
- Usually applied in batch treatment; effectiveness should remain constant over time (i.e. no "breakthrough" or saturation issues)

Disadvantages
- Requires rigorous and time-consuming operation and maintenance
- Usually requires pre-oxidation
- Generates arsenic-rich sludge
- Phosphate and silicate may reduce arsenic removal rates
- Treatment adds ions (sulphate, chloride) to the water, which may affect its taste
- Limited optimal pH range
- Limited field experience with electrocoagulation, processes not yet optimised

Co-precipitation with naturally occurring iron

High dissolved iron concentrations in groundwater pumped from anoxic aquifers can be utilised to remove arsenic. When the iron to arsenic mass ratio is greater than 40–50 (Meng et al., 2001), oxidation and filtration of iron will generally reduce arsenic to acceptable levels (USEPA, 2000c; USEPA, 2006). If groundwater also contains high phosphate concentrations, the iron:arsenic ratio should be even higher (Hug et al., 2008). If this criterion is met, then the system can function from its first use.

In Vietnam, household sand filters are commonly used for iron removal. An upper chamber is filled with locally available sand, while a lower chamber serves to store the filtered water. Groundwater pumped from a tube well trickles through the sand filter into the underlying storage tank (Fig. 7.5). Arsenic removal is governed by the precipitation of iron (hydr)oxides, which form a coating on the surface of the sand grains. Arsenic is then absorbed by the iron (hydr)oxides and remains immobilised under oxic conditions. The efficiency of the method is dependent on the concentration of the naturally occurring iron, as well as on the concentration of competing ions (especially with phosphate >2 mg/L) (Luzi et al., 2004; Roberts, 2004). Fe/As ratios of ≥50 or ≥250 are required to ensure arsenic removal to concentrations below 50 or 10 µg/L, respectively. In Vietnam, where 93% of tube wells contain >1 mg/L iron and <2 mg/L phosphate, the sand filters' median arsenic removal efficiency was 91%. Estimates for Bangladesh indicate that a median residual level of 25 µg/L arsenic could be reached in 84% of the contaminated groundwaters (Berg et al., 2006).

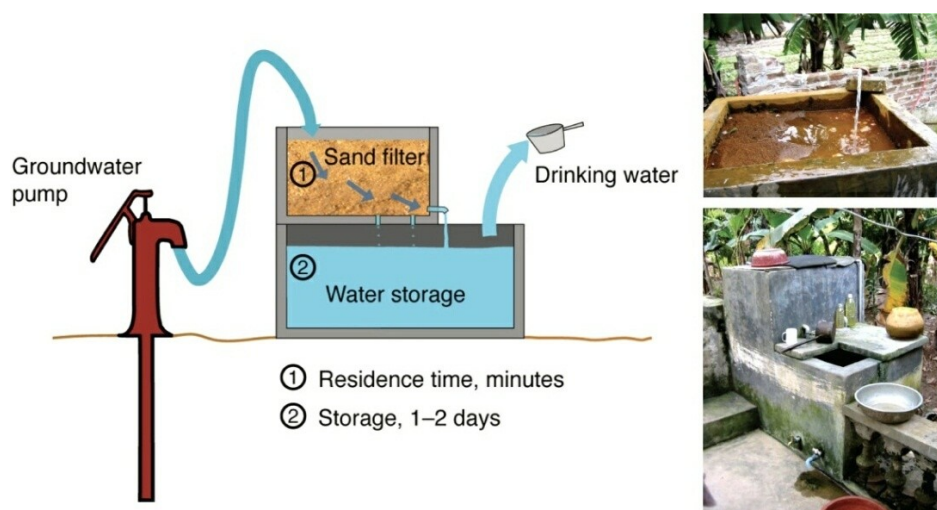**Fig. 7.5**   *Sand filter for arsenic removal in Vietnam*

Advantages

- Relatively inexpensive
- Achievable using locally available materials
- No consumables or regeneration needed
- Efficiency improves with time, as ferric iron accumulates in sand filter
- Taste and appearance of water is markedly improved through iron removal

Disadvantages
- Arsenic removal is limited, requires high Fe/As ratio
- Poor performance where phosphate concentrations are high
- Lack of standard design parameters can lead to inefficient "homemade" systems
- Stored water may be vulnerable to faecal contamination

Membrane methods

Selectively permeable synthetic membranes can remove a variety of contaminants, including arsenic. Reverse osmosis and nanofiltration are two membrane technologies suitable for arsenic removal, operating with membrane pore sizes of less than 0.01 micron, which is sufficient to remove metal ions. These membranes need to be operated with pressure gradients ranging from about 3 to 10 bar (Johnston et al., 2002).

Membrane techniques require that inflowing water be of relatively high quality to prevent membrane fouling, meaning that a preceding filtration step is often necessary. Arsenic removal is possible over a wide pH range.

The percentage of treated water that can be produced from the feed water is known as the recovery. In municipal systems, recovery can be up to 85% for nanofiltration and 30–85% for reverse osmosis. In household systems, this value is typically significantly lower (e.g. 10–25%), which can be seen as a disadvantage, as a large amount of raw water is needed to produce the desired amount of treated water (USEPA, 2003a).

Advantages
- Additional removal of other chemical contaminants and pathogens
- Arsenic removal over a wide pH range

Disadvantages
- Complex and maintenance-intensive process
- Membrane fouling needing pre-treatment and chemical cleaning
- Operation at high pressures
- Low recovery rate
- High capital and operating costs

Reverse osmosis for contaminant removal is described in more detail in Section 7.3.

Verification programmes

The performance of a number of commercial technologies for arsenic removal has been independently verified by different agencies.

The USEPA, through its Environmental Technology Verification programme, has evaluated twelve commercial arsenic removal systems, in cooperation with NSF International Technologies (USEPA, 2007) include coagulation/filtration, ion exchange, adsorption onto iron-based solids or iron-modified activated alumina, and reverse osmosis.

In Bangladesh, the project Bangladesh Environmental Technology Verification – Support to Arsenic Mitigation (BETV-SAM) evaluated fifteen technologies between 2005 and 2009. Six of the technologies were issued verification statements and have been certified for sale in Bangladesh. These six technologies are briefly profiled in the file, Verified_Arsenic_Removal_Technologies_in_Bangladesh.

More detailed reports on the six technologies can be downloaded:

- Shawdesh_Aqua_Filter

- Nelima_Filter

- MAGC/Alcan_Filter

- READ-F_Filter

- SONO_Filter

- SIDKO_Filter

In addition to the six technologies described above, the BETV-SAM project tested a further seven technologies but denied them verification.

Four of the verified technologies (MAGC/Alcan, READ-F, SONO, and SIDKO) were distributed at scale for the purpose of a social assessment through the Deployment of Arsenic Removal Technologies (DART) project. Experiences with these filters are described in detail by Hanchett and Khan (2009).

7.3 Fluoride treatment technologies

A range of technologies are available for the removal of fluoride from drinking water. These can be divided into three categories based on the underlying fluoride-removal process:

- Adsorption (Filter Materials)
- Precipitation and Coagulation
- Membrane Methods

In the following section, we profile the technologies that are suitable for application at household and community scales for decentralised systems in developing countries. We focus on technologies that have already been successfully implemented in the field. Many other technologies exist that have been tested in the laboratory but have not yet proved successful in the field or were never pursued. It is important to remember the following:

→ The ideal technology, suited for all types of conditions, does not exist!

The particular challenge for fluoride-removal technologies is the fluoride concentration in contaminated waters, which is roughly 50–150 times higher than arsenic concentration in arsenic-contaminated waters. This particularly affects the costs of the adsorption and precipitation/coagulation methods, as more filter materials, chemicals and maintenance are required.

The choice of the most suitable technology will be influenced by a range of factors, such as the fluoride concentrations in the input water, the funds available for implementation, operation and maintenance requirements (O&M), the local availability of raw materials and whether the technology is accepted by the population. Cost issues are usually at the forefront when decisions concerning the selection of a technology are made. The life cycle costs that will need to be considered (e.g. capital expenditures, maintenance expenditures etc.) are described in more detail in IRC (2011) and on the WASHCost website. Readers interested in technologies that are mentioned here may consult several reviews on defluoridation methods, for example Fawell et al. (2006), Ayoob et al. (2008), Mohapatra et al. (2009), KEBS (2010), Bhatnagar et al. (2011) and Jagtap et al. (2012).

Adsorption (filter materials)

A widely used method for the removal of fluoride is to pass the contaminated water through a filter bed that retains the fluoride. The binding of fluoride to the surface of granular filter materials is an adsorptive process. In developing countries, most filter materials that have a high affinity for fluoride are aluminium- or calcium-phosphate-based. On the following pages, we describe two commonly used materials: activated alumina and bone char.

Activated alumina

Activated alumina (AA) is a commercially available granular form of aluminium oxide (Al_2O_3) that can be used as a filter medium to remove a range of contaminants, including fluoride, from water (Fig. 7.6).

In contact with water, the surface of the AA becomes hydrated and forms $Al(OH)_3$ with surface hydroxide groups ($\equiv Al\text{-}OH$). Negatively charged ions can replace the hydroxide (OH^-) ion, as shown for fluoride below:

$$\equiv Al\text{-}OH + F^- \rightarrow \equiv Al\text{-}F + OH^-$$
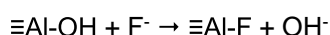
The strength of binding with the sites is reported by Amy et al. (2000) to be:

$$OH^- > H_2AsO_4^- > F\text{-} > SO_4^{2-} > HCO_3^- > Cl^- > NO_3^-$$

This means that while hydroxide binds most strongly, the binding strength of fluoride is stronger than most ions in drinking water. This means that there will be little competition from these ions.

The highest removal capacities using AA are achieved within the narrow pH range of 5.5–6, when the attraction of fluoride ions to the AA surface is at its greatest and interference with competing ions is minimised. At higher pH values, the bed capacity is significantly lower, and fluoride breakthrough occurs earlier (Rubel and Woolsley, 1979). Activated alumina is used in industrialised countries in municipal plants, but also in developing countries at community and household scales (see e.g. Venkobachar et al., 1997; Daw, 2004).

Fig. 7.6   *One type of activated alumina: Compalox (Albemarle®)*

Production: Activated alumina is a commercially available product.

Fluoride Removal Efficiency: AA is highly efficient in reducing fluoride concentrations in treated water to levels below 0.3 mg/L. Fluoride removal of 85–95% can be achieved in well-maintained systems running at optimum conditions (Pickard and Bari, 2004). However, filter function is dependent on input-water quality and especially its pH. AA fluoride uptake capacity is at a maximum between pH 5.5 and pH 6, and it decreases considerably with increasing pH values. Waters with high alkalinity and high pH therefore need to be acidified before they are passed over the AA bed. There are many different types of activated alumina with different uptake capacities on the market.

Regeneration and re-use: When the AA is exhausted, it needs to be regenerated. (The filter material may also be replaced, but regeneration is generally more cost-effective.) Regeneration is typically done by passing a sodium hydroxide solution (1–4%) over the AA bed, followed by rinsing with clean water. This results in caustic waste water rich in total dissolved solids, aluminium and fluoride, which needs treatment to remove these ions before disposal. The filter is then reactivated using sulphuric acid or CO_2 gas, followed by flushing with water until the bed is at a pH of ~6.

The fluoride removal capacity appears to be lower after each regeneration cycle. Complete replacement of the filter material is generally necessary after 3–5 regeneration cycles (e.g. Chauhan et al., 2007; Fawell et al., 2006), while application in South Africa has shown AA media to still be efficient after 6 or more regeneration cycles (Schoemann, 2008). A step-by-step documentation on activated alumina regeneration as carried out in villages in India is given in the UNICEF Report, "Regeneration Manual for Activated Alumina used in Domestic Defluoridation Units".

Costs: The initial costs of the filter material are generally relatively high, though efficient regeneration may bring down overall costs considerably. However, it will be necessary to establish a "regeneration centre" at a central location where spent media from household and community filters can be brought for regeneration.

Advantages
- High fluoride uptake capacity (at pH 5.5–6)
- Filter medium can be regenerated

Disadvantages
- Skilled operator needed for supervision of plant (community filter) and for centralised regeneration
- Expensive filter material, not cost-effective if not regenerated
- Pre-treatment necessary if pH of input water is too high

Bone char

The charring and crushing of animal bones produces a granular material that has been used successfully in several countries (e.g. Kenya, Ethiopia, Thailand) as a filter material to remove excess fluoride from drinking water. The removal of fluoride from water by bone char (BC) is an adsorptive process, allowing the exchange of fluoride ions with hydroxide ions (OH^-) at the surface of the main mineral constituent of BC (the calcium phosphate, hydroxyapatite, $Ca_5(PO_4)_3OH$), releasing OH^- into solution:

$$Ca_5(PO_4)_3OH + F^- \leftrightarrow Ca_5(PO_4)_3F + OH^-$$
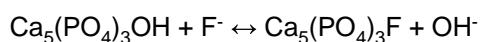
Bone char filters can be implemented at both the community and household scales (Fig. 7.7). Raw water is fed into columns or filters and is allowed to percolate through the system. Once the 1.5 mg/L fluoride threshold has been reached, the material needs to be regenerated or replaced.

Filter Material Production: The bone material needs to be largely free of flesh before it is charred. The charring is carried out in a kiln in a low-oxygen atmosphere at a temperature of 300 to 500°C for approximately 10 days, to produce bone char with the highest fluoride removal capacity with no organic remains (CDN, 2007). If the temperature is too high, the hydroxyapatite contained in bones changes to another mineral, and the resulting bone char has a significantly reduced uptake capacity. The desired product should be grey in colour. A soot-coloured product indicates the presence of organic material, and a white-coloured product indicates that the temperature was too high.

After charring, the bones are crushed to size fractions between 0.4 and 4 mm, which are washed with solution a solution of sodium hydroxide (6 g/L, pH 13) to remove remaining organic substances. The bones are then rinsed with water and acidified with CO_2 gas.

In Ethiopia and Kenya, large kilns capable of charring several tonnes of bone per batch are used. In Thailand, small household furnaces have been tested in which householders can produce their own bone char (Smittakorn et al., 2010). Generally, in small-scale production, it may be harder to maintain quality control of the bone char produced than in larger, standardised processes.

Note: Production requires skill and may take some time to perfect. Should a faulty batch containing residual organic substances that taint the water be used, it is probable that the trust of users will be irrevocably destroyed.

Fluoride Removal Efficiency: The fluoride uptake capacity of the filter material depends on the quality of the bone char and particle size. The smaller the particles, the higher their uptake capacity (Mjengera and Mkongo, 2002). Implementation in Kenya has shown that BC filters can reduce fluoride concentration from over 6 mg/L to less than 0.1 mg/L after filtration, with a fluoride uptake capacity of ~1.2 mg/g determined in both field and laboratory studies (Mutheki et al., 2011).

Regeneration: Once the WHO drinking-water standard for fluoride (1.5 mg/L) or a national standard has been reached in the treated water, the filter material needs to be replaced or regenerated. Regeneration is typically done by passing a sodium hydroxide solution (0.25%–1%) through the BC bed, followed by rinsing with clean water. This results in caustic waste water rich in total dissolved solids and fluoride, which needs either to be neutralised or strongly diluted. The filter is then reactivated using CO_2 gas followed by flushing with water until the effluent has a pH of ~6. The fluoride removal capacity is lower after each regeneration cycle.

The caustic waste can be treated with $CaCl_2$ or $Ca(OH)_2$ (lime) to produce a highly insoluble solid CaF_2 precipitate, which needs to be disposed of safely.
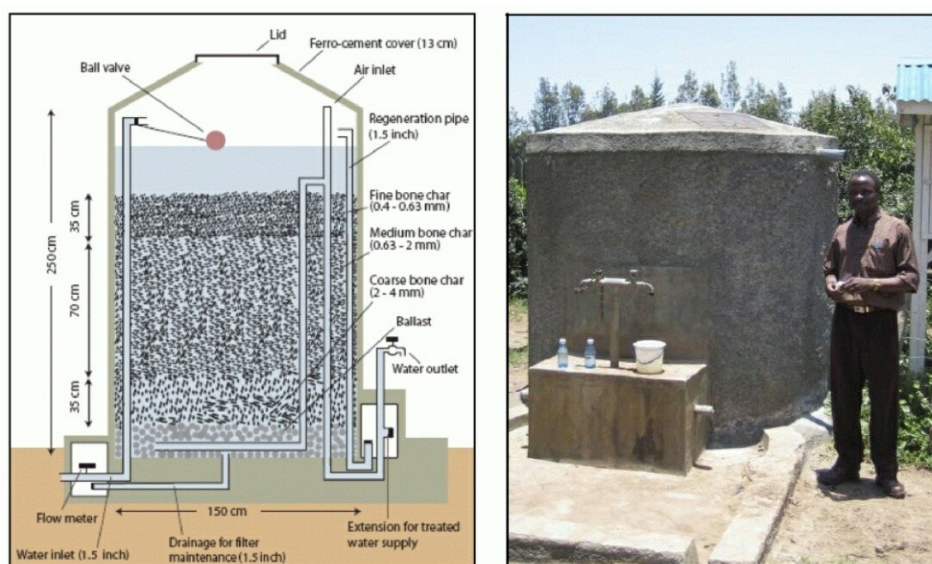

Fig. 7.7 *Community BC filter used in Kenya by the Nakuru Defluoridation Company (NDC)*

Costs: Production of bone char is intensive in terms of infrastructure and labour. These costs and the cost of raw bones are the main contributors to the total costs. Charcoal for starting the charring process, electricity for crushing and sieving the charred bones, caustic soda for washing the bone char and bags for packing the final material are of comparatively minor importance.

Advantages
- Bones as raw material are locally available at relatively low cost
- Filtered water is neutral in taste and colour (if the BC has been correctly produced)
- Relatively short contact time required (around 30 minutes)

Disadvantages

- Initial investments and experience needed for setting up bone char production (building of kiln etc.)
- The use of animal bones as a filter material is not acceptable in some regions for religious or cultural reasons
- Use of low quality bone char with a high organic content might result in the treated water having an unacceptable taste.
- Relatively low fluoride uptake capacity (around 1.2 mg/L), which can necessitate frequent filter media replacement and lead to high transportation costs

Synthetic "bone char": HAP

Bone char essentially consists of hydroxyapatite ("HAP", $Ca_5(PO_4)3OH$). This material can also be produced synthetically using simple raw materials (lime and phosphoric acid). Laboratory studies have shown that synthetic HAP can have a clearly higher fluoride uptake capacity than BC. Synthetic HAP is already used for fluoride removal in Germany and Italy and was used in the past in the USA. Recently, the Nakuru Defluoridation Company in Kenya has also started producing HAP and is now testing the material in the field.
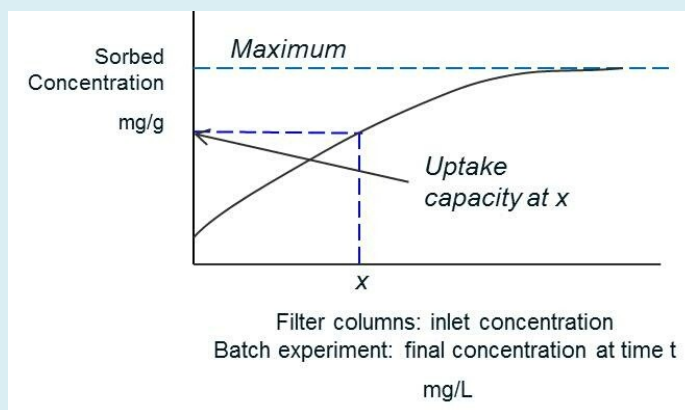
Uptake capacities of filter materials

Fig. 7.8   *An adsorption isotherm for fluoride*

The uptake capacity of a filter material is important, because it provides information on how long a filter material will last. The maximum uptake capacity is attained when all available sites are occupied and occurs only at high dissolved arsenic or fluoride concentrations. At lower dissolved concentrations, the amount that is sorbed is proportional to the amount in solution:

$$K_d = C_{solid}/C_{solution}$$

where K_d is the distribution coefficient, and C_{solid} and $C_{solution}$ are the solid-phase and

dissolved fluoride or arsenic concentrations, respectively.

Thus the uptake capacity will be high at higher inflow concentrations. The uptake capacity is also influenced by solution pH, time and temperature.

!! Handling acids and bases !!

The regeneration of filter materials is usually carried out using sodium hydroxide and concentrated acids, such as sulphuric acid, for neutralisation. The handling and storage of such chemicals requires occupational health training and skills development, careful supervision and strict enforcement of rules and regulations.

Guidelines

Wear safety goggles to avoid permanent damage of the eyes when working with acids and bases.

Wear suitable clothing that will protect you against spilled chemicals. Hard-soled, covered footwear must be worn at all times.

Wear gloves to protect your hands.

In case of spills, wash chemicals from skin straightaway.

   i) Wash your hands and face quickly and thoroughly whenever they come into contact with a chemical.
   ii) If you receive a chemical burn from an acid or base, immediately wash the burned area with large quantities of water.
   iii) Chemicals spilled over a large part of the body require immediate action. Remove all contaminated clothing and rinse with water. Do not use creams or lotions, etc. Get medical attention.

   Note: If you wear contact lenses, they must be removed for effective cleansing. It is better to wear glasses in case of a spill.

Work in well-ventilated surroundings to avoid inhaling of toxic fumes. Acid fumes in particular can cause permanent damage to the lungs.

Always pour concentrated acids into dilute solutions or water and **never** the other way round. Heat is generated by the mixing process, and by controlling the amount of acid in the mixture, you can prevent the temperature from rising too much. Quick mixing can cause the mixture to boil and splash the surroundings.

Further Reading

National Research Council (1995) Prudent practices in the laboratory: Handling and disposal of chemicals. Washington, DC: The National Academies Press.

Precipitation and coagulation

Fluoride can be removed from solution by precipitation and coagulation processes, followed by the settling (or flotation) of the precipitates. This usually involves the addition of chemicals that act as precipitating agents. Established techniques involving precipitation or coagulation include the Nakuru technique, the Nalgonda technique and

electrocoagulation.

Contact precipitation (the Nakuru Technique)

The contact precipitation technique is also a filter method and works by adding calcium (Ca) and phosphate (PO4) compounds to untreated water, with fluoride concentrations being reduced by both sorption and precipitation reactions when the fluoride comes into contact with hydroxyapatites. Bone char provides a surface for the precipitates to form.

One method, implemented in Tanzania, is to add $CaCl_2$ and NaH_2PO_4 to the water. These dissolve, releasing Ca and PO_4. The resulting solution is then passed through a bone char bed (Dahi, 1996). It is relatively high in maintenance, as frequent addition of chemicals is required.

To combat this drawback, another contact precipitation approach has been developed and successfully implemented by the Nakuru Defluoridation Company (formerly the Water Quality Group of the Catholic Diocese of Nakuru (CDN WQ)) in Kenya, involving the production of calcium phosphate pellets, which slowly release Ca and PO4 when in contact with water (Fig. 7.9). This technology is known as the Nakuru Technique. The water passes through a pellet and BC mixture (3:1 ratio) and then through a bone char bed. The Nakuru Technique has successfully been implemented in fluoride removal filters in Kenya and Ethiopia (Fig. 7.10). In the following paragraphs, we will describe this method in more detail.

Fig. 7.9 *Calcium phosphate pellets that are used in combination with bone char in fluoride removal filters ("Nakuru Technique")*
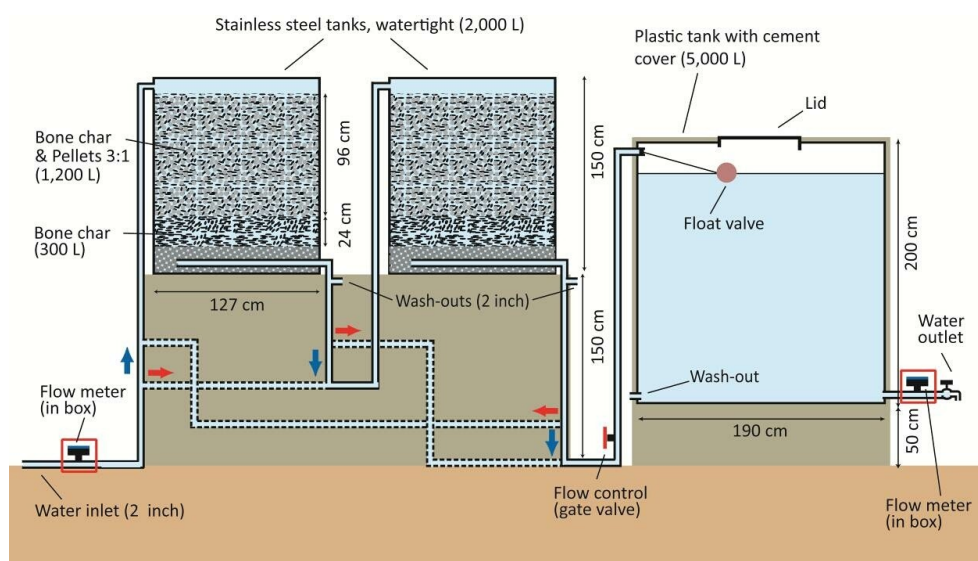
Fig. 7.10 *Design of a Nakuru Technique filter implemented in the Ethiopian Rift Valley by Eawag and Oromia Self-Help Organisation (OSHO)*

Filter Material Production: To the authors' knowledge, Ca-PO$_4$ pellets for use in fluoride removal filters are currently only produced by NDC in Kenya. Pellets are produced in a cement mixer using Ca(OH)$_2$, Kynofos21 (a commercially available Ca-PO$_4$ mixture sold as animal feed) and bone dust as raw materials. Subsequent curing, washing and drying steps follow. Readers that are interested in more details should contact the Nakuru Defluoridation Company Ltd. For details on bone char production, see the "Bone Char" section in this document.

Fluoride Removal Efficiency: Monitoring has shown that the fluoride uptake capacity of a bone char filter can be increased up to threefold, to 2–4 mg/L, when Ca-PO$_4$ pellets are added (Korir et al., 2009; Mutheki et al., 2011) (see Fig. 7.11). The fluoride removal efficiency of the Nakuru Technique is highly dependent on the flow rate. The filters have to be designed in a way that allows the water to stay in contact with the filter medium for a long time (at least 3 hours).

Regeneration and Disposal: Regeneration of contact precipitation filter material is not possible. It therefore needs to be replaced when the pellets are exhausted and fluoride breakthrough occurs (>1.5 mg/L). Whether spent filter material could be valuable as a phosphate fertiliser to increase crop yields is still being investigated. Preliminary research has shown that spent filter material has a lower fluoride content than commercially available fertilisers and similar phosphate availability (Hukari, 2011).

Costs: A clear advantage over regular bone char systems is that the filter medium lasts longer, thereby reducing replacement and transportation requirements. On the other hand, the pellet costs depend highly on the cost of the calcium phosphate used for its production. If phosphate prices increase in future, the costs for pellet

production will rise as well. The filter material costs (without regeneration for BC) for treating water with an initial fluoride content of 5 mg/L in Kenya are currently around 2.5 USD/m³ for CP and 4.2 USD/m³ for BC (Mutheki et al., 2011).
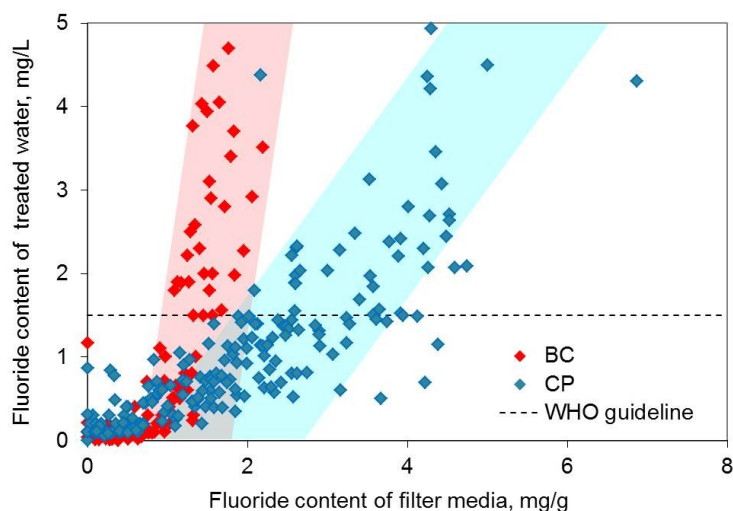

Fig. 7.11 *Fluoride uptake as a function from field tests in Ethiopia and Kenya. Shaded areas show ranges for BC and CP obtained from laboratory tests done at NDC (Kenya) and Eawag (Switzerland) (Johnson et al., 2011)*

Advantages

- Prolonged lifespan of filter material in comparison to filters containing only bone char
- Non-toxic raw materials
- Research suggests that the spent medium can be reused as fertiliser

Disadvantages

- Regeneration of the filter medium is not possible
- Fluoride removal efficiency is highly dependent on the flow rate, which makes its application in household filters difficult
- Pellets used in the Nakuru Technique are not widely available commercially, as they are currently produced only by NDC in Kenya
- Skill and experience are needed for pellet and bone char production
- Kynofos21 (calcium phosphate raw material) might not be available locally and would have to be imported, or a local alternative found

Nalgonda technique

The removal of fluoride using alum as a coagulant was first proposed in the United States in the 1930s. It was later adapted by the National Environmental Engineering Research Institute (NEERI) in India in the 1970s and named the "Nalgonda Technique" (Nawlakhe et al., 1975, Fig. 7.12). It is an alum-based coagulation-flocculation method that requires

alum (aluminium sulphate, $Al_2(SO_4)_3$) and lime (calcium hydroxide, $Ca(OH)_2$):

$$Al_2(SO_4)_3 + 3Ca(OH)_2 \rightarrow 2Al(OH)_3 + 3Ca^{2+} + 3SO_4^{2-}$$
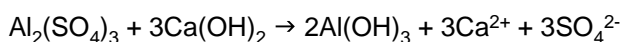
Alum is first dissolved and is then added to the untreated water, forming aluminium hydroxide flocs. Fluoride binds to these flocs, which are left to settle.

The dose of chemicals required depends on the quality of the raw water. Although rough dose rates exist based on theoretical models and field trials (Lyengar, 2000; UNICEF, 2008; Fawell et al., 2006), these cannot be taken as standard in every case. Field trials will therefore be necessary to determine the correct dose.
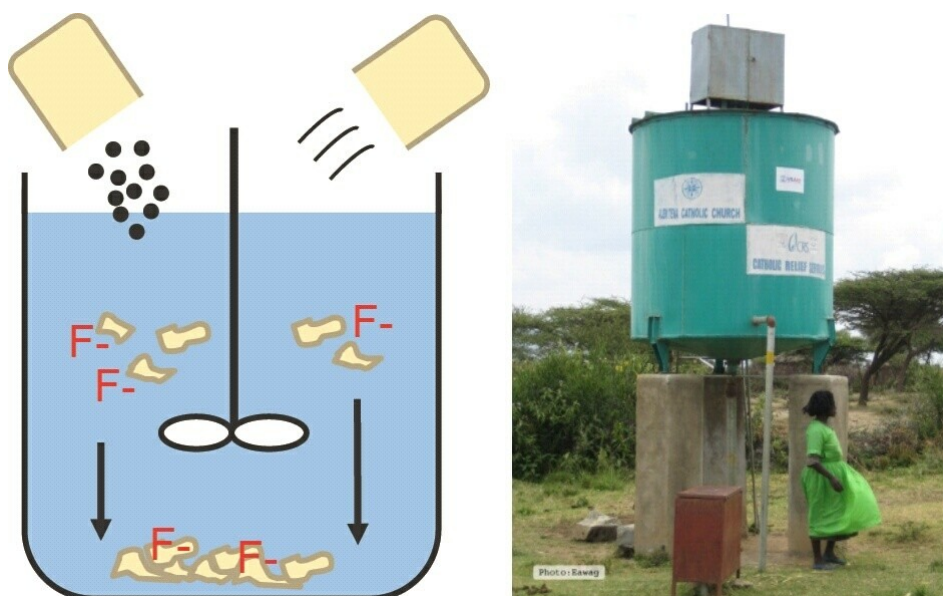

Fig. 7.12 **Left:** *Principle of Nalgonda technique (left): Alum and lime are added to the high fluoride water, the mixture is stirred and precipitates containing fluoride settle as sludge to the bottom of the solution*
Right: *Community Nalgonda Unit installed by the Catholic Relief Service in the Ethiopian Rift Valley*

Fluoride Removal Efficiency: The Nalgonda technique may be insufficient to reduce F-values to below 1.5 mg/L when alkalinity and fluoride values in the untreated water are high. Use of the Nalgonda technique in Tanzania only reduced fluoride concentrations to 2.1–3 mg/L in water initially containing between 8 and 12 mg/L fluoride (Dahi et al., 1996).

Fluoride and alkalinity levels in raw water need to be monitored frequently, as the chemical dosage needs to be adjusted according to the quality of the inlet water.

Disposal: Fluoride- and aluminium-rich sludge is produced, which needs to be disposed of safely, out of reach of children and animals and away from drinking-water sources, preferably landfilled. Disposal in latrines is possible if these are well separated from groundwater resources.

Costs: The chemicals needed (alum and lime) are relatively cheap and readily available in most countries, making the Nalgonda technique an inexpensive fluoride removal

method if conditions are such that fluoride guidelines are met. Additional costs for a generator need to be taken into account for community units which require an electrical stirrer.

Advantages

- Chemicals readily available in most countries
- Relatively inexpensive in comparison to other technologies

Disadvantages

- Insufficient fluoride removal efficiency when concentrations in raw water are high
- The method is labour intensive and requires rigorous and time-consuming operation and maintenance
- Some community filter units require power for the electrical stirrer
- Electrical stirrers include movable parts, which are prone to mechanical failure
- Perceived taste of the treated water may be affected by high sulphate concentrations (~ 600 mg/L)
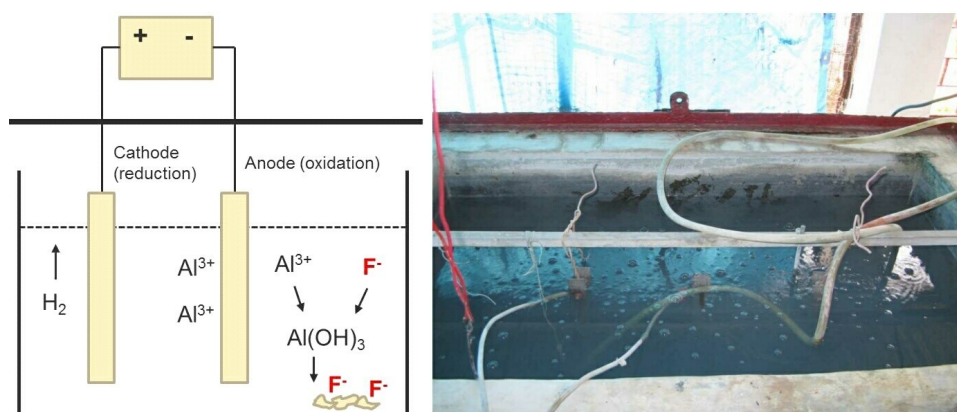- Large amounts of waste are produced that are often deposited onsite

Electrocoagulation

Fig. 7.13 *Schematic principle of electrocoagulation (left) and EC community plant operated by NEERI, India (right)*

The electrocoagulation (EC) method has been used to remove fluoride and other ions from industrial wastewaters for some time (e.g. Shen et al., 2003; Hu et al., 2008) and is now increasingly receiving attention as a suitable technology for fluoride removal from drinking water in developing countries. This technology lies at the intersection of three more fundamental technologies: electrochemistry, coagulation and precipitation.

The method utilises metal (e.g. aluminium) plates that act as anode and cathode. When a potential is applied to the electrodes, a current flows and Al3+ is released at the anode and reacts with water at neutral pH to form precipitate of Al(OH)3, a compound which has a high affinity for fluoride (Fig. 7.13). The resulting $Al(OH)_3$-F flocs settle at the bottom of the solution and can be removed as sludge.

Fluoride Removal Efficiency: Fluoride removal efficiency depends on the initial fluoride concentration, the initial pH of the influent water and the current density (Emamjomeh and Sivakumar, 2009b; Gwala et al., 2011; Ghosh et al., 2008; Zuo et al., 2008; Zhao et al., 2011). The optimum pH for fluoride removal lies between 6 and 7. Laboratory studies have shown that fluoride concentrations can be lowered from 15 mg/L to below 1.5 mg/L within 40 min (Gwala et al., 2011; Mameri et al., 1998). Field implementation in India has accomplished fluoride removal from 4.5 mg/L to below 1 mg/L within 2 hours using solar energy as an electricity source (Gwala et al., 2011).

Disposal of Waste: The fluoride- and aluminium-rich sludge settling at the surface needs to be removed and disposed of safely, out of reach of children and animals and away from drinking-water sources. Disposal in latrines is possible if these are well separated from groundwater resources. Another possibility may be to stabilise sludge in cement or bricks.

Costs: Electrocoagulation uses simple and readily available materials (e.g. aluminium plating). An electricity source is needed (solar panels or a generator), which can result in high initial costs and, in the case of a generator, high operational costs as well.

Advantages

- High fluoride removal efficiency (at pH 6–7)
- Simple system, no moving parts
- No hazardous chemicals used (unless pH adjustment with acid needed)
- Relatively small amounts of sludge generated

Disadvantages

- High SO_4^{2-} concentrations in raw water can inhibit fluoride removal
- Aluminium levels in treated water may exceed the level recommended by WHO level (200 µg/L)
- Energy source needed (e.g. solar energy)
- pH may need to be controlled
- Requires relatively skilled staff

Membrane methods

Membranes with fine pores can be used to separate contaminants from water physically. As the fluoride ion is very small, most membranes are not fine enough to retain it. **Reverse osmosis** is a technique utilising very fine membranes coupled with high pressures to remove fluoride from drinking water efficiently.

Reverse osmosis

Reverse osmosis utilises a synthetic, semipermeable membrane, which allows the passage of water but not of ions or larger molecules. In principle, during the process of osmosis, water molecules move through the membrane along a concentration gradient

from a high to a low dissolved salt concentration. The opposite effect is desired in the reverse osmosis process: pressure is applied on the membrane to overcome the osmotic pressure and to force water molecules from the concentrated solution to the fresh water side (Fig. 7.14). Reverse osmosis is widely applied for desalination and water purification purposes, including the removal of fluoride. More information on the principle of reverse osmosis and other membrane methods can be found in a range of documents, including Elimelech and Phillip (2011), Greenlee et al. (2009), Mulder (2000), Pontié et al. (2006) and Shannon et al. (2008).

Compared to other technologies for fluoride removal, reverse osmosis has the advantage that it removes not only ions, such as fluoride, but also pathogens (viruses, bacteria, protozoa). There are two major limitations of the reverse osmosis technology:

1   High energy requirements;
2   Membrane fouling.

Membrane fouling occurs when suspended particulate matter, colloids, bacteria and organic material are deposited on the surface of the membrane. To control fouling, a pre-filtration step or conventional pre-treatment (e.g. coagulation and disinfection) may be needed to remove the particulate, colloidal and dissolved organic matter causing the fouling. Chemical cleaning is used to restore the permeability of the fouled membranes. During reverse osmosis filtration, feed water is recirculated, and only a certain percentage (around 20–50%, depending on the system used) of the raw water ends up as treated water (permeate), the rest being waste. Reverse osmosis therefore has a high water demand and should not be used in areas of known water scarcity.
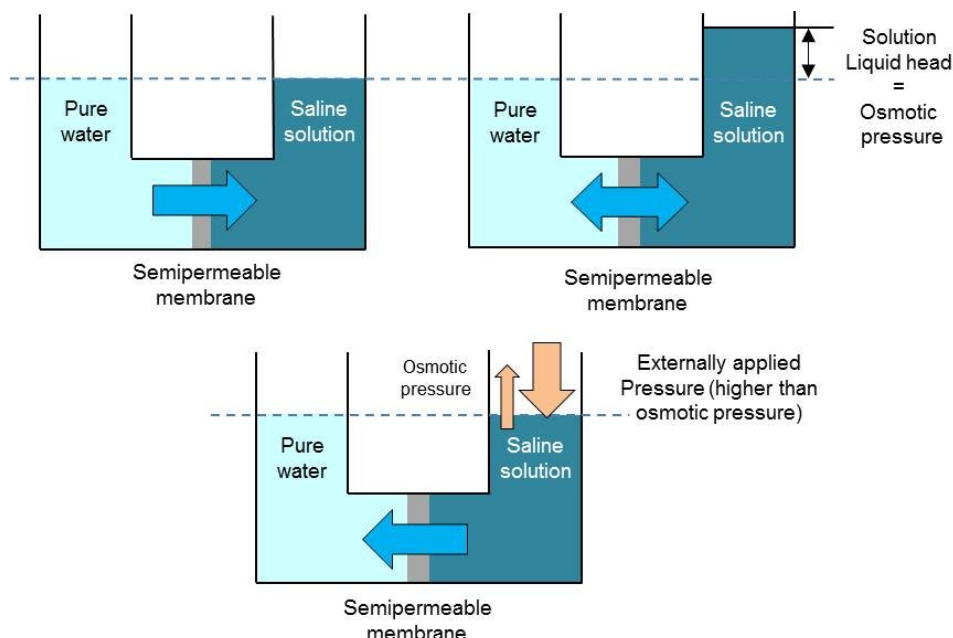

Fig. 7.14 *Principle of reverse osmosis*

Fluoride Removal Efficiency: Reverse osmosis can remove fluoride almost completely. Treated water can be deficient in minerals serving as essential micronutrients to humans and generally needs to undergo remineralisation before distribution.

Costs: Reverse osmosis is a high-tech process needing skilled operators. Capital and operational costs are high. It is an energy-intensive technology, requiring the generation of high pressures. Electricity costs can therefore be substantial.

Advantages

- Efficient fluoride removal
- Reduction in salinity
- Additional removal of chemical contaminants and pathogens

Disadvantages

- Complex and high maintenance process
- Membrane fouling needing pre-treatment and chemical cleaning
- High energy consumption
- High water use
- Cost-intensive

References and further reading

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Links to alternative water resources information

International Network on Household Water Treatment and Safe Storage. www.who.int/household_water

Operation & Maintenance Network: www.operationandmaintenance.net

Rural Water Supply Network (RWSN): www.rural-water-supply.net

Samsamwater: www.samsamwater.com/library.php

SSWM Toolbox: www.sswm.info/category/implementation-tools/water-purification/hardware/point-use-water-treatment/hwts

Technology Applicability Framework (TAF): www.washtechnologies.net/en