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Arsenic reduction to < 1 µg/L in Dutch drinking water

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ABSTRACT

Arsenic (As) is a highly toxic element which naturally occurs in drinking water. In spite of substantial evidence on the association between many illnesses and chronic consumption of As, there is still a considerable uncertainty about the health risks due to low As concentrations in drinking water. In the Netherlands, drinking water companies aim to supply water with As concentration of < 1 µg/L – a water quality goal which is tenfold more stringent than the current WHO guideline. This paper provides (i) an account on the assessed lung cancer risk for the Dutch population due to pertinent low-level As in drinking water and cost-comparison between health care provision and As removal from water, (ii) an overview of As occurrence and mobility in drinking water sources and water treatment systems in the Netherlands and (iii) insights into As removal methods that have been employed or under investigation to achieve As reduction to < 1 µg/L at Dutch water treatment plants. Lowering of the average As concentration to < 1 µg/L in the Netherlands is shown to result in an annual benefit of 7.2–14 ME. This study has a global significance for setting drinking water As limits and provision of safe drinking water.

1. Introduction

Chronic arsenic (As) poisoning related to prolonged exposure through drinking water is a global issue. Most of the affected population lives in South Asia, particularly in Bangladesh, India, Pakistan and Nepal (Ahmad and Bhattacharya, 2018a; Ahmed et al., 2004; Bhattacharya et al., 2002; Mukherjee et al., 2006; Mushtaq et al., 2018; Ahmad and Bhattacharya, 2018b). Nevertheless, major reports of As contaminated drinking water have also been emerged from several countries in the Eurasian region such as Turkey, Greece, Romania, Slovakia, Croatia, Serbia, Hungary, Spain (Baba and Sözbilir, 2012; Bundschuh et al., 2013; García-Sánchez et al., 2005; Lindberg et al., 2006; Papić et al., 2012; Rowland et al., 2011; Varsányi and Kovács, 2006) and several other Latin American countries such as Argentina, Bolivia, Chile and Uruguay (Bundschuh et al., 2010; Lima et al., 2019; Litter et al., 2010; Ormachea Muñoz et al., 2016; Ormachea Muñoz et al., 2013; Ramos Ramos et al., 2012; Litter et al., 2019a, 2019b)

(Fig. 1).

Several types of cancer and other detrimental effects on human health due to chronic As exposure have been reported (Das and Sengupta, 2008; Ferreccio et al., 2013; Gibb et al., 2011; IARC, 2012; Islam et al., 2012; Jovanovic et al., 2013; Marshall et al., 2007; Parvez et al., 2011; Rahman et al., 2011; Sengupta et al., 2008; Smith et al., 1992) (Fig. 2). However, there is a considerable uncertainty about the health risks due to exposure to low As concentrations, including the concentrations within WHO guideline for As in drinking water (< 10 µg/L) (Ahmad and Bhattacharya, 2019; Kozisek, 2017; Saint-Jacques et al., 2018; Schmidt, 2014). In the Netherlands, drinking water is of high quality and As concentrations are much lower than the WHO guideline, nevertheless, drinking water companies aim to supply drinking water with < 1 µg/L As.

In this paper we provide i) an account on the assessed lung cancer risk for the Dutch population due to pertinent low-level As in drinking water and cost-comparison between health care provision and As

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Fig. 1. Arsenic contamination of drinking water in different parts of the world.

removal from water, ii) insights into As occurrence and mobility in Dutch drinking water sources and water treatment systems and iii) insights into As removal methods that have been employed or are under investigation to achieve As reduction to $< 1 \mu\text{g/L}$ at (municipal) water treatment plants (WTPs) in the Netherlands.

2. Rationalizing reduction of As to $< 1 \mu\text{g/L}$ in Dutch drinking water

In 2015 the Association of Dutch Drinking water Companies (Vewin) voluntarily agreed on a guideline of $< 1 \mu\text{g/L}$ for As in drinking water (Van der Wens et al., 2016). This policy is based on a two-step assessment of As in drinking water, including i) an assessment of excess lung cancer risk for Dutch population and ii) a cost-comparison between the health care provision for lung cancer and As removal from water to avoid lung cancer (Van der Wens et al., 2016). The two-step assessment is described in the following sections.

2.1. Health risk assessment for Dutch population

In the Netherlands, like most other Western European countries, As concentrations in drinking water are much lower than the WHO guideline. The dose-response relationship at these low exposure levels is still unclear (Ahmad and Bhattacharya, 2019; EFSA, 2009). Consequently, no safe threshold value for As exposure is known below which health effects are negligible. The general consensus in the Netherlands is that, in cases like this, linear extrapolation from high-dose studies can be used to assess particular health risks.

Since lung cancer is the most sensitive end-point of As exposure (Celik et al., 2008; EFSA 2009; Gibb et al., 2011), an assessment of excess life-time lung cancer risk due to low-level As in drinking water was carried out for Dutch population (Van der Wens et al., 2016). The risk assessment was done by linear extrapolation of risk using a Chilean study (Ferrecio et al., 2000) which had been previously found appropriate with respect to quality and applicability for the American population by the National Research Council (National Research Council, 2001). It was concluded that based on Ferrecio et al. (2000), with a lifelong exposure to $50 \mu\text{g/L}$ As, the lung cancer risk would increase by 41 (28–54)%, where the numbers in brackets indicate 95% confidence interval (National Research Council, 2001). Considering the average life-time risk for developing lung cancer of 3.0% in the Netherlands (in 2015), this implies a 1.23 (0.84–1.62)% increase in life-time lung cancer risk for Dutch population. Further, linear extrapolation was applied to assess the risks at lower As concentrations (Table 1). This showed that the WHO guideline ($10 \mu\text{g/L}$ As) is not sufficiently

protective and does not correspond to the excess life-time cancer risk of 10^{-6} in the Netherlands which is considered as Virtually Safe Dose (VSD) according to Dutch policy. Based on calculations, exposure to each additional $\mu\text{g/L}$ As through drinking water can increase the risk for lung cancer development in the Dutch population by 0.025 (0.017–0.032)%. Considering the drinking water As concentrations in some areas in the Netherlands which are as high as $6 \mu\text{g/L}$ according to 2015 survey (Ahmad et al., 2015), the excess risk of developing lung cancer due to As exposure is 0.15 (0.10–0.19)%. It is noteworthy that the linear extrapolation shows that the theoretical VSD of As in drinking water would be $0.004 \mu\text{g/L}$. Accurately determining such a low concentration of As in water is beyond the sensing capability of most analytical methods. But on the other hand, the linearly extrapolated risks might be an overestimate (Schmidt, 2014). Therefore, a second step assessment was carried out to determine a reasonable As guideline for the Netherlands (see following section).

2.2. Cost-benefit analysis of arsenic removal

In the second-step of assessment, the health care costs related to the treatment of additional cases of lung cancer and the financial investments required to remove As to lower than 1 and $2 \mu\text{g/L}$ were compared. The total annual costs for achieving the target level of $< 1 \mu\text{g/L}$ As in Dutch drinking water was estimated at 3.8–7.2 M€ (Van der Wens et al., 2016). This estimate was based on As removal pilot studies carried out at several WTPs in the Netherlands. On the other hand, for the health related costs, the Disability-Adjusted Life-Years (DALYs) approach of the Dutch National Health Council was used (Hanninen et al., 2014; NHC, 2007; Prüss-Üstün et al., 2003). DALYs are a measure for burden of disease of a certain population and can be calculated by adding the ‘Years of Life Lost’ (YLL) to the ‘Years Lived with Disability’ (YLD). The YLL can be obtained by multiplying the number of deaths per year by the remaining life expectancy at the moment of dying, whereas the YLD can be obtained by multiplying the incidence of a certain disease by the product of the so-called ‘disability weight’ and the disease duration (Stouthard et al., 1997). It is noteworthy that since the mortality rate of lung cancer is the highest of all forms of cancer in the Netherlands and the expected life duration is significantly reduced by lung cancer (Hoeymans et al., 2010; Volksgezondheidzorg, 2018), the largest part of disease burden was calculated for As-induced lung cancer incidences and mortality during the assessment (Van der Wens et al., 2016). Based on the Dutch total population of 16.656.000 (in 2011) and an average life expectancy of 80 years (Havelaar and Melse, 2003), it is estimated that every $\mu\text{g/L}$ As in drinking water can annually result in 51 (35–68) cases of extra lung cancer which corresponds to

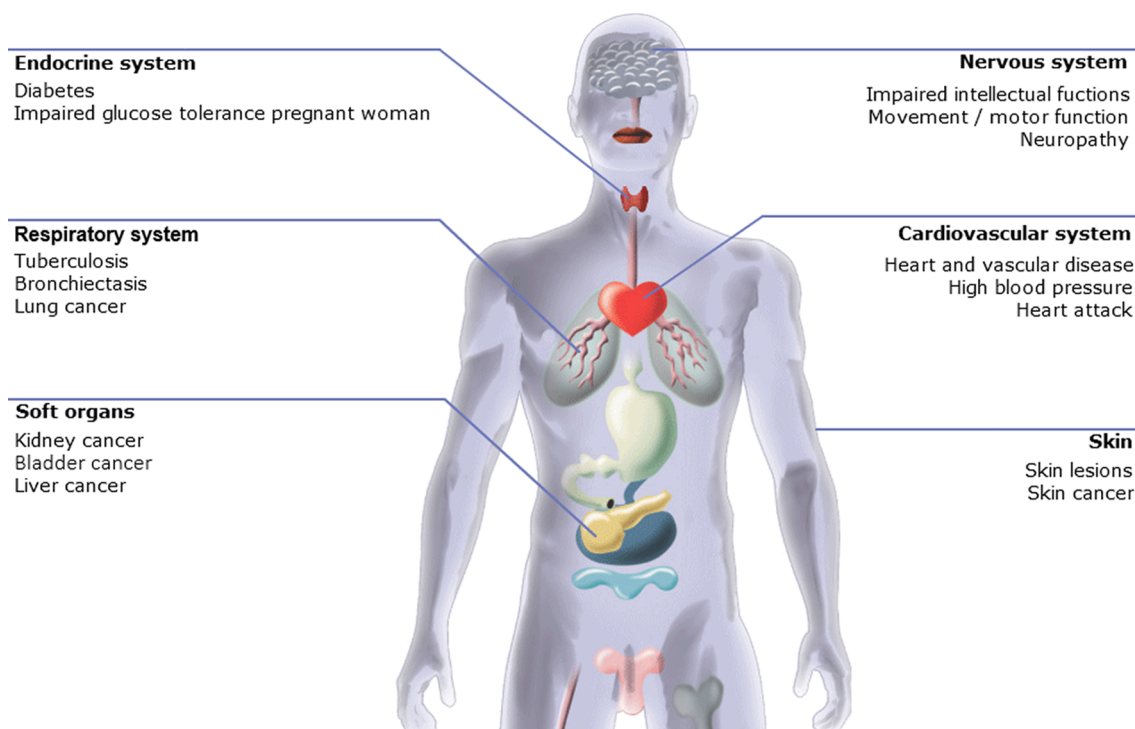


Fig. 2. Adverse health effects due to chronic As exposure through drinking water.

Table 1

Estimated excess life-time lung cancer risk due to different As concentrations in drinking water for Dutch population.

Arsenic concentration in drinking water ($\mu\text{g/L}$)	Average extra life-time lung cancer risk in % (95% confidence interval)
50	1.23 (0.84–1.62)
20	0.49 (0.34–0.65)
10	0.25 (0.17–0.32)
5	0.12 (0.08–0.16)
3	0.07 (0.05–0.10)
1	0.02 (0.02–0.03)

756 (516–996) DALYs (provided that the total population would be exposed lifelong to this concentration). Taking into account the costs of health care for cases of lung cancer caused by As (approximately 1 M€ for each $\mu\text{g/L}$ As) and the value of avoidance of a DALY (€ 60,000 on average in the Netherlands (Pomp et al., 2014)), a cost-benefit analysis of reduction of the average As levels in drinking water (1.2 $\mu\text{g/L}$ As) to < 1 $\mu\text{g/L}$ revealed an annual benefit of 10.7 M€ (7.2–14 M€) (Van der Wens et al., 2016). This justifies the ambition of water sector to supply drinking water with As concentration of < 1 $\mu\text{g/L}$ in the Netherlands.

3. Arsenic in Dutch drinking water sources

In the Netherlands, fresh water from different sources is treated for drinking water production at about 180 municipal WTPs. Most of the drinking water is obtained from deep groundwater, supplemented by surface water which is used directly or after infiltration (e.g. dune infiltration, river bank filtration, Fig. 3). Five types of Public Supply Well Fields (PSWFs) are recognized in the Netherlands (Table 2) (Mendizabal and Stuyfzand, 2009). A PSWF is typically a coherent set of pumping wells delivering natural groundwater, artificially recharged surface water or river bank filtrate to the public as drinking water, mostly with and rarely without treatment. The spatial distribution of As in PSWFs is

given in Fig. 4(a) (Mendizabal et al., 2011; Stuyfzand et al., 2008). The mean As concentrations of the raw water from 241 PSWFs range between < 0.5 and 69 $\mu\text{g/L}$ which has been stable since 2008 (Ahmad et al., 2015). The mean As concentrations for each groundwater type are quite low (0.3–2.9 $\mu\text{g/L}$; Table 2), but there are 8 well fields with As concentrations > 10 $\mu\text{g/L}$ (Fig. 4A). These higher concentrations are observed in anoxic sand aquifers and deeply anoxic river bank filtration systems. The highest As concentration at the abandoned PSWF Oostrum (69 $\mu\text{g/L}$; Fig. 4A) is associated with oxidation of pyrite (FeS_2 , a common natural sulfide mineral) through excessive manure and fertilizer applications on an agricultural catchment area. This situation is encountered in other well fields also, like in Vierlingsbeek (a phreatic well field, discussed in detail by Stuyfzand et al. (2008)), but the mixed raw water remains < 10 $\mu\text{g/L}$ with respect to As due to effective well field adaptation measures such as cautiously raising the pumping rate of low As wells. The high As concentrations (> 10 $\mu\text{g/L}$) also correlate with abstraction from relatively deep situated, glauconitic sand aquifers of Late Tertiary age (Fig. 5), such as Dorst and Klotputten (Fig. 4A). The exact cause for this correlation is still unclear (Coetsiers, 2007; Flink, 1985), because reductive weathering of glauconite ($[\text{K,Na}][\text{Fe}^{3+},\text{Al,Mg}]_2[\text{Si,Al}]_4\text{O}_{10}(\text{OH})_2$, a natural aluminum silicate mineral), can be overshadowed by dissolution of syngenetic apatite ($\text{Ca}_5(\text{PO}_4)_3[\text{OH,F}]$). The well fields pumping from cretaceous limestone typically have very low As concentrations (Table 2). There are no clear relations between As on the one hand and PO_4^{3-} , SO_4^{2-} , HCO_3^- , Fe, Cl⁻, Mn and DOC on the other hand, but high concentrations correspond with pH 6–7.5, $\text{NO}_3^- < 2\text{mg/L}$ and screen depths of approximately 0–50 m below sea level (Fig. 5; (Stuyfzand et al., 2008)). To gain further insights into As speciation in source water a limited sampling campaign including 14 PSWFs was carried out in 2015 (Ahmad et al., 2015). The high As concentrations mainly coincides with prevalence of As(III) in (deeply) anoxic environment. A relatively high percentage of As(V) corresponds in most cases with NO_3^- and low Fe(II) concentration.

In some local situations a strong As mobilization (up to 1500 $\mu\text{g/L}$) can take place and affect drinking water or its treatment in future

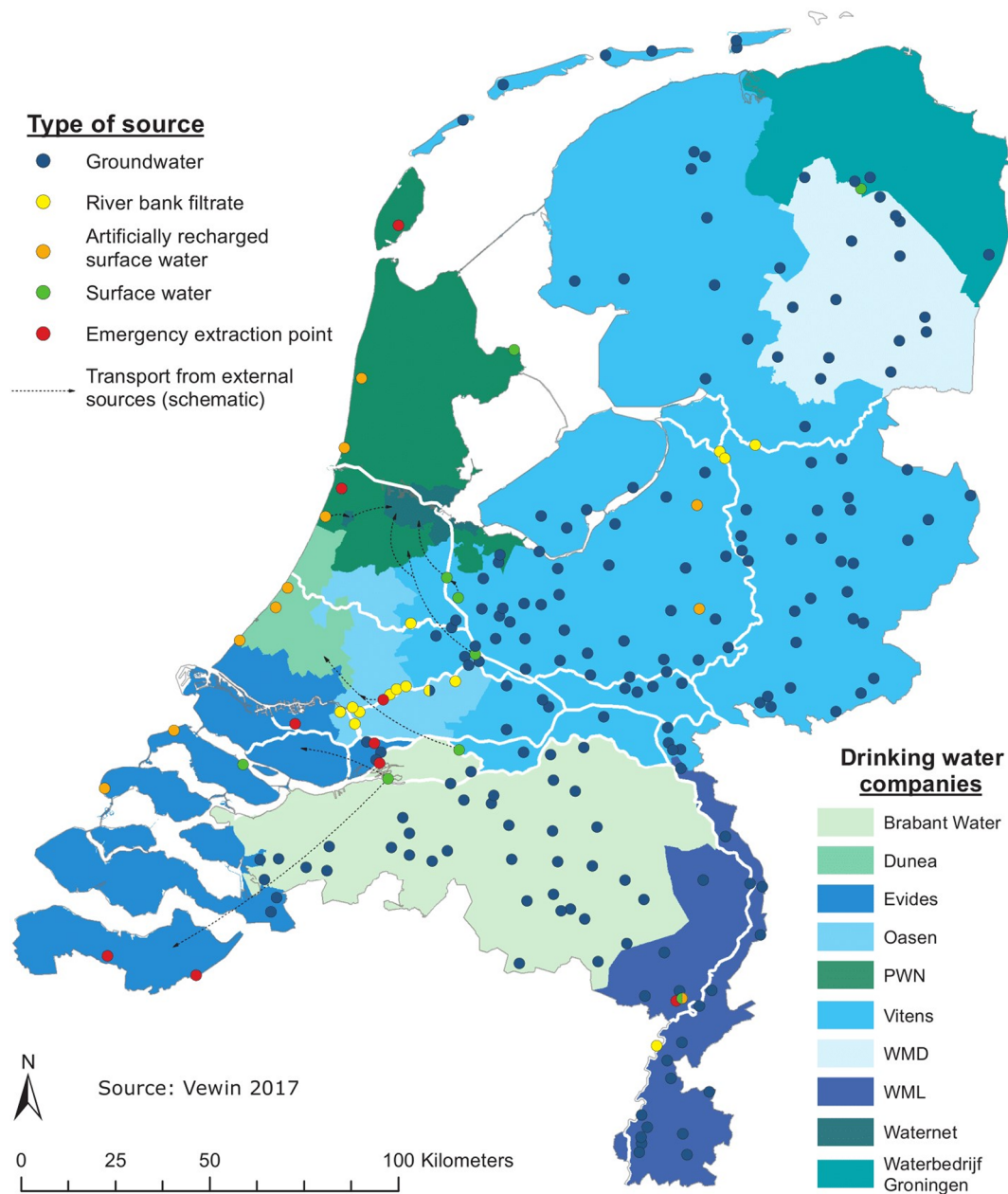


Fig. 3. The water companies and type of raw water sources used for drinking water production in different parts of the Netherlands.

(Ahmad et al., 2015; Stuyfzand et al., 2008). This is concluded from detailed studies of the following young hydrological systems in sandy aquifers of Quaternary age: (i) a partly decalcified, pumped aquifer system with strong agricultural inputs (PSWF Vierlingsbeek); (ii) artificial recharge using basins (PSWF Leiduin and Scheveningen), (iii) artificial recharge using injection wells (a pilot near Langerak), (iv) river bank filtration in the Hollandsch Diep, and (v) a polder system south of Amsterdam City, composed of a reclaimed lake surrounded by an influent eutrophic river, and underlain by Holocene peat. The most probable As mobilizing processes in the systems i–v are: raised NO_3^- inputs on agricultural plots underlain by a pyritiferous aquifer (i); the introduction of O_2 and NO_3^- containing surface water into deeply anoxic aquifers containing pyrite (ii–iii); (sub)recent changes in quality of infiltrating river water (ii, iv), especially regarding rises of PO_4^{3-} , SO_4^{2-} , HCO_3^- , DOC and temperature (which contribute to desorption of As); (sub)recent mud accumulations in infiltrating parts of lakes, rivers and recharge basins (ii, iv) where the oxidation of fresh organic matter is producing a lot of CH_4 which may reduce iron(hydr)oxides

and As(V) (to As(III)), while also producing a lot of PO_4^{3-} , HCO_3^- and DOC which compete for sorption sites with As; and the reductive dissolution of iron(hydr)oxides plus desorption in peat rich polder areas (v) where the oxidation of peat is producing elevated quantities of CH_4 , PO_4^{3-} , HCO_3^- and DOC.

The cumulative frequency distribution of total dissolved As concentrations in the raw water pumped by all PSWFs in the Netherlands (Figs. 6 and 7) indicates that if As is tackled at the source level, e.g. by managing flow, well combinations or even shutting down some wells, ca. 45% of all the PSWFs in the Netherlands would require (substantial) efforts to meet the $1 \mu\text{g/L}$ As target.

4. Fate and removal of arsenic during water treatment

According to the 2015 survey (Ahmad et al., 2015), mean As concentrations in Dutch drinking water were in the range of < 0.5 – $6.2 \mu\text{g/L}$ (average $1.2 \mu\text{g/L}$) (Fig. 4B), thus lower compared to the As concentrations in source water (Fig. 4A). This indicates that some As is

Table 2
Hydrological classification of PSWF types in the Netherlands, with data referring to the year 2008.

PSWF type	Unit	Well field type				
		A	B	i	K	U
		Phreatic	Confined	Artif. Rech.	Limestone	Bank filtrate
First year of operation of first PSWF		1853	1893	1940	1904	1890
Number of active PSWFs in 2008 \$		67	126	12	9	27
Total volume of drinking water produced #	Mm ³ /a	187	449	192	23	74
Mean volume production per PSWF #	Mm ³ /a	2.8	3.7	16.0	2.6	2.7
% of total volume of drinking water produced #	%	20	49	21	3	8
Mean number of wells / sampling points		11	12	131	7	17
Mean land surface altitude	m ASL	18	14	9	68	3
Mean abstraction level	m BLS	26–60	82–135	10–31	24–74	24–56
Age spectrum	a	10–200	60–25,000	0.1–50	2–500	1–200
EC 20 °C	µS/cm	371	389	584	771	687
pH		7.01	7.25	7.60	7.03	7.25
Temp	°C	11.0	11.6	12.3	10.4	12.5
O ₂	mg/L	2.2	0.8	3.5	3.8	0.7
CH ₄ ⁺	mg/L	0.65	2.7	0.01	0.26	1.1
Cl ⁻	mg/L	26.9	24.8	69.4	31.2	80.6
HCO ₃ ⁻	mg/L	141	206	178	367	258
SO ₄ ⁻²	mg/L	41.1	11.6	58.9	62.8	40.1
NO ₃ ⁻	mg/L	5.3	0.1	3.9	22.2	1.5
PO ₄ ⁻³	mg/L	0.44	0.71	0.30	0.16	1.70
SiO ₂	mg/L	17.4	20.3	7.2	16.4	24.1
Fe	mg/L	3.8	3.8	0.5	0.4	3.3
Mn	mg/L	0.23	0.17	0.11	0.01	0.47
NH ₄ ⁺	mg/L	0.26	0.48	0.15	0.03	2.01
As	µg/L	1.8	1.5	2.9	0.3	2.0
As-max (max for well field type)	µg/L	13.2	26.3	7.6	< 0.5	7.4

Water production in year 2004; \$ In the number of active PSWFs (241 in total), a PSWF with separate well strings pumping from a shallow and deep aquifer respectively, was counted twice (generally A and B) because they were sampled separately. A,B = groundwater formed by natural rainwater infiltration, in sandy aquifers; Artif. Rech = artificially recharged surface water; Limestone = as A or B, in limestone aquifer; ASL = Above Sea Level; BLS = Below Land Surface

removed during water treatment. Interestingly, the design of the WTPs is not traditionally based on As removal and the observed removal of As at certain WTPs in the Netherlands is rather “incidental”. Detailed studies to understand the natural As removal mechanisms at WTPs have been carried out in recent years. Here we summarize briefly some recent findings.

Anaerobic groundwater is typically treated by aeration and rapid sand filtration (Fig. 8) to remove dissolved Fe(II), Mn(II) and NH₄⁺ (Ahmad et al., 2018; de Moel et al., 2006). Removal of the co-occurring As in groundwater has been found to be due to co-precipitation with naturally present Fe in groundwater (Gude et al., 2016), in agreement with McNeill and Edwards (1995) and Lytle et al. (2007) in the United States, Sorlini et al. (2014) in Italy and Katsoyiannis et al. (2008) in Greece. When groundwater is aerated, Fe(II) oxidizes ($4\text{Fe}^{2+} + \text{O}_2 + 10\text{H}_2\text{O} \rightarrow 4\text{Fe}(\text{OH})_3 + 8\text{H}^+$) and undergoes hydrolysis to produce Fe(III) hydroxide precipitates (Fe(III)-precipitates) which can adsorb As from water (Hering et al., 1996; McNeill and Edwards, 1995). The As bearing Fe(III) precipitates are retained in the rapid sand filter bed (Jessen et al., 2005; Van Beek et al., 2012; Vries et al., 2017) and therefore an effluent with reduced As concentration is produced (Fig. 8). The differences in As removal efficiencies between different WTPs in the Netherlands can be explained by taking into account the composition of source water (Ahmad et al., 2019, 2020; Gude et al., 2016). The adsorption efficiency of As onto Fe(III)-precipitates strongly depends on pH, As speciation and co-occurring ions (Jain and Ali, 2000; Pierce and Moore, 1982; Raven et al., 1998; Senn et al., 2017; Wilkie and Hering, 1996). The adsorption of As(V) is generally more efficient compared to As(III) in the pH range of most natural waters (Hering et al., 1996; Hsu et al., 2008; Lakshmanan et al., 2008; Lytle et al., 2005) because As(V) is negatively charged and As(III) is neutral. Co-occurring anions in water, such as PO₄⁻³ and SiO₄⁻⁴ as well as humic acids can reduce As adsorption to Fe(III) precipitates due to competition for adsorption sites (Ahmad et al., 2020; Dixit and Hering, 2003; Hering

et al., 1996; Meng et al., 2000; Su and Puls, 2001; Wilkie and Hering, 1996; Youngran et al., 2007). Calcium-hardness, on the other hand, improves the removal of As (Ahmad et al., 2020; Guan et al., 2009; Hering et al., 1996; Senn et al., 2017).

The design and operation of rapid sand filters has been found to play a critical role in determining As removal efficiency at WTPs. The supernatant depth, filter media grain size and bed configuration are all important factors to control As removal. With higher supernatant depth on the rapid sand filters, homogeneous Fe(II) oxidation and concomitant As uptake was improved in a pilot study (Gude et al., 2018a). Also, the use of double media rapid sand filter, including a coarse granular top layer and finer bottom layer, was found to result in a higher As removal efficiency due to formation of As-Fe(III) precipitates deeper in rapid sand filter bed (Gude et al., 2018a, b).

The role of ripened rapid sand filter media for the oxidation and removal of As(III) has been studied in detail. It has been observed that As(III) rapidly oxidizes to As(V), in a matter of few minutes, during rapid sand filtration (Ahmad et al., 2018, 2014; Gude et al., 2016). Similar rapid oxidation of As(III) to As(V) in rapid sand filters has also been reported in previous studies (Jessen et al., 2005; Lytle et al., 2007). The accelerated As(III) oxidation in rapid sand filters is attributed to bacteria (Gude et al., 2018c), though the potential role of manganese oxides on the granular filter media has not been convincingly rejected (Gude et al., 2016, 2017, 2018c).

5. Methods to reduce arsenic to < 1 µg/L in drinking water

At several WTPs small operational adjustments, as discussed in section 4, may increase the As removal efficiency to achieve the desired < 1 µg/L As in produced drinking water. However, considerable changes in the treatment are inevitable at some WTPs because of the relatively high As concentrations or complexity of the water matrix. In this regard, methods based on As and Fe(III) co-precipitation have

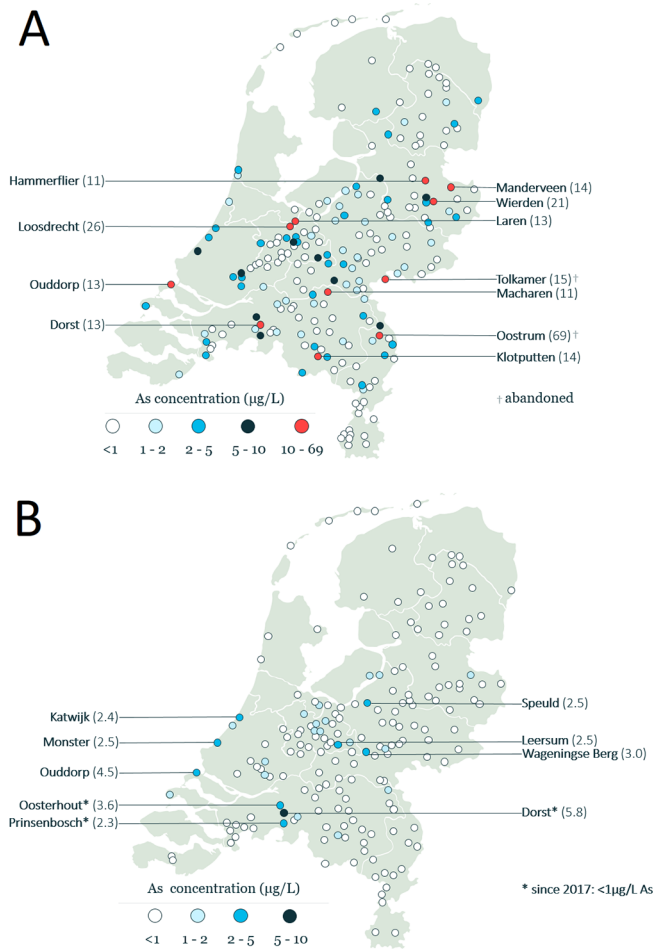


Fig. 4. Mean As concentrations in (A) raw water pumped by the 242 PSWFs in the Netherlands (Stuyfzand et al., 2008) and (B) produced drinking water (Ahmad et al., 2015). PSWFs with As concentrations higher than 10 µg/L are indicated. The abandoned PSWFs are marked with a graveyard icon. WTPs with As concentrations higher than 1 µg/L are also indicated.

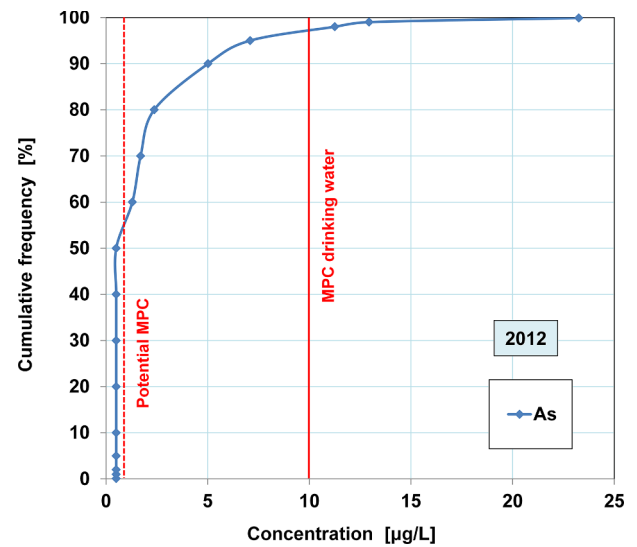


Fig. 6. Cumulative frequency distribution of total dissolved As concentrations in the raw groundwater from 159 PSWFs in 2012. Minimum detection limit was 0.5–1.0 µg/L.

received a significant interest, mainly because of the ease of implementation at existing water treatment plants (Ahmad et al., 2017). The simplest approach is to increase the Fe concentration in water by dosing an Fe(III) coagulant (Edwards, 1994), e.g. FeCl₃, which undergoes hydrolysis to form Fe(III) precipitates that adsorb As from water (Hering et al., 1996, 2017; Meng et al., 2000). However, these (additional) As bearing Fe(III) precipitates have been found to significantly reduce run time of rapid sand filters (Ahmad et al., 2014). A reduction in rapid sand filter run time results in a higher frequency of filter backwashing and hence a higher use of water and energy. Such consequences are generally undesired, but can be resolved by redesigning the rapid sand filters.

Arsenic removal by dosing Fe(II) (e.g. as FeSO₄) and subsequent oxidation of the added Fe(II) into Fe(III) by oxygen (aeration) is another promising approach, briefly addressed in a recent study (Gude et al., 2018b). This method can have some advantages over Fe(III) dosing. For

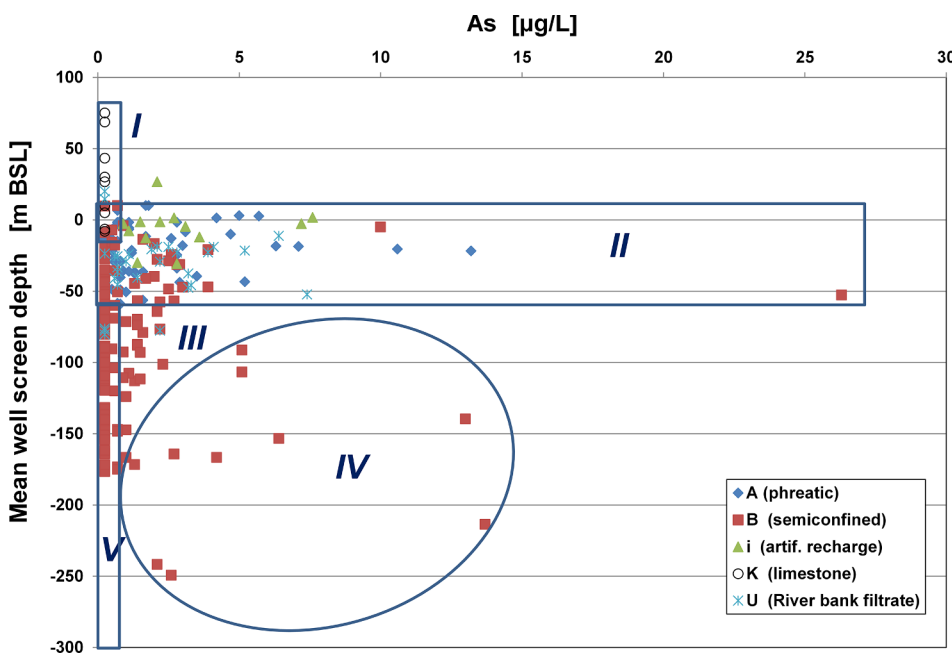


Fig. 5. Concentration of total dissolved As in the raw water of 5 PSWF types (ABIKU), as function of mean well screen depth. Based on data collected in 2008. I = (sub)oxic groundwater in limestone; II = zone where raised concentrations frequently occur due to either pyrite oxidation or reductive dissolution of ferric(hydr)oxide; III = transition/mixing zone from II to V; IV = zone with raised As levels due to mobilization in glauconite rich sediment; V = deep zone with normally low As concentrations.

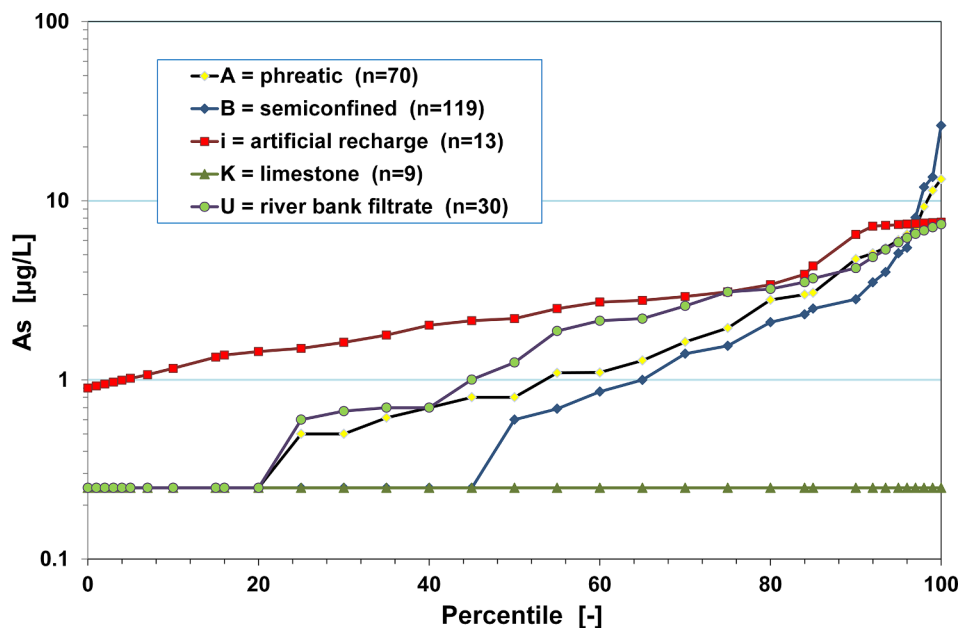


Fig. 7. Cumulative frequency distribution of total dissolved As concentrations in the raw water pumped by the 242 PSWFs in the Netherlands in 2008. Based on data from 2008 sampling campaign.

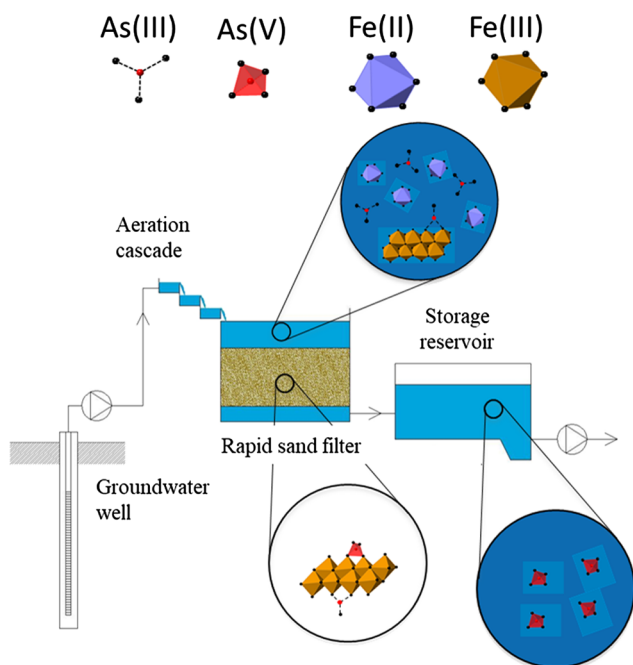


Fig. 8. The mechanisms controlling the fate and removal of As(III) and Fe(II) during conventional groundwater treatment. Modified from (Ahmad et al., 2018). Note that As(III) oxidizes to As(V) in rapid sand filters and the effluent contains predominantly As(V).

example, during the oxidative hydrolysis of Fe(II), co-oxidation of As(III) to As(V) can occur due to the oxidizing radical species that form during oxidation of Fe(II) by dissolved O_2 (Hug and Leupin, 2003; Roberts et al., 2004). Thus, use of a strong chemical oxidant for As(III) oxidation can be avoided. Also, by reducing the rate of Fe(II) oxidation (e.g. by pH reduction), filter run times longer than in case of Fe(III) may be obtained, as preliminarily shown by Sharma (2001). Further research is required to prove these postulated advantages of Fe(II) over Fe(III) dosing at WTPs.

In order to achieve a higher As adsorption onto Fe(III) precipitates and reduce Fe dosages at WTPs, complete oxidation of As(III) into As(V)

is generally required. Rapid oxidation of As(III) can be achieved at WTPs by dosing a strong oxidant, such as potassium permanganate ($KMnO_4$) (Ahmad et al., 2018; Sorlini and Gialdini, 2010). But when $KMnO_4$ (and $FeCl_3$) dosing was adopted at a Dutch WTP for As(III) oxidation ($3H_3AsO_3 + 2MnO_4^- \rightarrow 3H_2AsO_4^- + 2MnO_{2(s)} + H_2O + H^+$), it also reacted with Fe(II) ($3Fe^{2+} + MnO_4^- + 7H_2O \rightarrow 3Fe(OH)_{3(s)} + MnO_{2(s)} + 5H^+$) and Mn(II) ($3Mn^{2+} + 2MnO_4^- + 2H_2O \rightarrow 5MnO_{2(s)} + 4H^+$) and affected their pre-established, O_2 driven and microbial mediated, removal mechanisms in the rapid sand filters (Ahmad et al., 2018). Moreover, the clarification behaviour of the filter backwash water was considerably altered due to $KMnO_4$ dosing (Ahmad et al., 2018). Since it is known that the properties of Fe and Mn oxides, such as surface charge, colloidal stability and reactivity with solutes depend on the (redox) conditions under which they are formed and this behaviour is not yet completely elucidated (Bruins et al., 2015; Cornell and Schwertmann, 2003; Van Genuchten and Pena, 2017; Vries et al., 2017), future research should look into the macroscopic and molecular properties of Fe(II) and Mn(II) oxidation products as a function of oxidant type (e.g. O_2 , $KMnO_4$, Cl_2) so that the above mentioned operational consequences can be better understood.

6. Conclusion and future prospects

The epidemiological effects of As on human health due to chronic exposure to very low concentrations through drinking water are uncertain. While this uncertainty needs to be addressed, this paper presents a two-step approach that has been used in the Netherlands to derive a guideline for As in drinking water. The first step includes health risk assessment based on linear extrapolation from a high-dose study which showed that the current WHO guideline ($10 \mu g/L$ As) is not sufficiently protective for the Dutch population. The second step compares the health care costs related to the medical treatment of excess lung cancer cases to the financial investments required for As removal from drinking water. Lowering of the average As concentration to $< 1 \mu g/L$ in the Netherlands is shown to result in an annual benefit of 7.2–14 M€. Thus, Dutch drinking water companies aim to reduce As concentrations to $< 1 \mu g/L$ in drinking water and have been able to successfully achieve this target in recent years using Fe based co-precipitation methods under different set of conditions.

We acknowledge that in several low income countries compliance

with a low As guideline such as 1 µg/L can be technically challenging and costly. Nevertheless, an integral assessment of As removal costs and resulting health benefits should be considered in guideline setting process. In other words, we are suggesting a reassessment and fixation of the drinking water As limits to a level where health benefits outweigh the removal costs.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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