

Article

Reduction in Arsenic Exposure by Domestic Water Purification Devices in Shanghai Area and Related Health Risk Assessment

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Abstract: Domestic water purification devices (point-of-use) are important choices for people to deal with sudden drinking water contamination. Especially for sudden heavy metal and arsenic contamination, home water purification units can play a key role as a secondary line of defense. Most of the arsenic removal studies are limited to meeting the requirements of local water quality standards and few studies have been conducted on arsenic removal from household water purifiers. In this paper, the distribution characteristics of arsenic in water at the end of the pipeline network in the Shanghai water supply area have been investigated. Three types of household water purifiers, mainly with membrane separation technology, were selected to treat the simulated arsenic contamination in tap water sources. The effects of pH and inorganic salts on arsenic removal were studied in comparison to the results of coexisting ion concentrations in tap water. The results showed that the total arsenic concentrations in different tap water samples range from 0.479–1.195 $\mu\text{g L}^{-1}$, which is lower than the limit value for arsenic concentration set by China's drinking water standard GB 5749–2006 (10 $\mu\text{g L}^{-1}$) and by the World Health Organization (10 $\mu\text{g L}^{-1}$). It is found that reverse osmosis and nanofiltration water purifiers were more effective in removing As(V), with removal rate above 97.7%, and less effective in removing As(III), the rate ranging from 40.1 to 56.3%. Preliminary validation was provided for the safety of household water purifiers in securing universal and sudden drinking water contamination. Domestic water purifiers are effective in reducing the risk of arsenic exposure in drinking water.

Keywords: arsenic; drinking water; point-of-use; health risk; contamination incident

1. Introduction

Arsenic is an element that is carcinogenic to humans, mainly through ingestion [1]. The World Health Organization's recommended limit for arsenic in drinking water is 10 micrograms per liter [2]. The presence of trace amounts of arsenic in water is difficult to avoid due to geological reasons [3] and arsenic contamination in water has always been a widespread concern. At present, the problem of arsenic in water has not been completely solved [4,5] and trace amounts of arsenic in drinking water still pose a threat to people's health [6]. In particular, arsenic contamination has been one of the main threats to the safety of people's drinking water, affecting a large number of people with arsenic contamination of drinking water sources such as lakes, rivers, private well water, etc. [7]. In response to these situations, many scholars and engineers have undertaken a lot of research and engineering practices [8–11] which mainly focus on the municipal tap water production process and the treatment of private well water. The main methods used are membrane filtration [12,13], adsorption [14], coagulation, etc. [15]. In recent years home water purification devices have

become an important solution to prevent and solve sudden pollution incidents [16–18]. Home (domestic) water purifiers, also known as point-of-use (POU) systems, have been widely used in the United States [19]. The purpose of a home water purifier is to further purify drinking water and reduce possible health risks. Domestic water purifiers usually consist of several treatment units. Membranes and adsorbent materials are the primary treatment units in home water purifiers and studies have shown that these materials are effective in removing common contaminants from water, but lack of maintenance can also lead to secondary contamination. Domestic water purifiers have also come into use in recent years in some less economically developed areas [20], mainly to compensate for the lack of public water purification facilities [21].

In recent years, the incidence of drinking water contamination has increased, posing a serious threat to people's health [22]. Most of the research on maintaining the safety of drinking water in response to emergencies has been directed at the operation of water plants and the maintenance of pipe networks [22,23]. In areas where public water treatment facilities are available and tap water is not direct drinking water, few people use home water purifiers. In fact, home water purification devices are also a solution to deal with sudden drinking water contamination. Especially for sudden heavy metal and arsenic contamination, home water purification units can play a key role as a secondary line of defense. Few studies have been conducted on arsenic removal from household water purifiers. Most of the arsenic removal studies are limited to meeting the requirements of local water quality standards and rarely involve the removal and risk assessment of trace amounts of arsenic at concentrations below the required limits of water quality standards.

The objective of this work is to know the health risk of arsenic in tap water and the possibility of risk reduction through the way of home water purifiers. Therefore, the distribution characteristics of arsenic in water at the end of the pipeline network in Shanghai have been investigated. Household membrane purifiers were used to treat the simulated arsenic contamination in tap water samples with arsenate and arsenite. The effects of pH and inorganic salts on arsenic removal were studied. A preliminary evaluation of the human health risk of arsenic in end-of-pipe water and pure water after using the water purifier in each district of Shanghai was made. This research can provide the information about the arsenic levels in tap water and technical support to more efficient household water purifier products.

2. Materials and Methods

2.1. Water Quality Analysis of the End Water of Shanghai Tap Water Network

2.1.1. Sampling

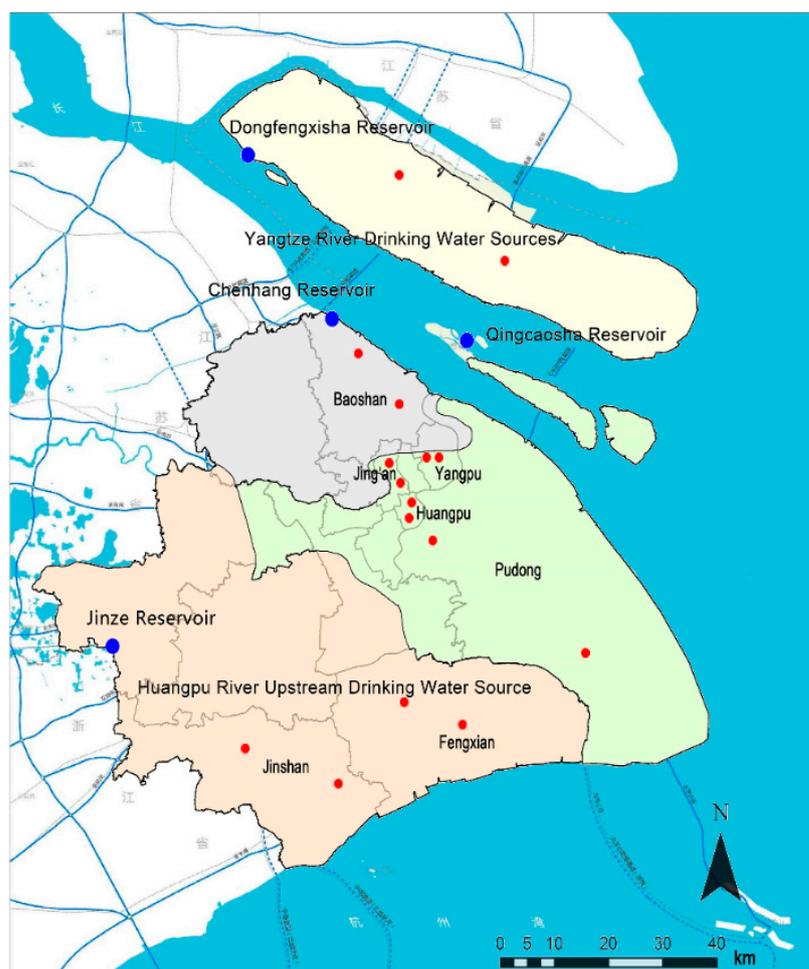
According to the water supply area distribution of Shanghai drinking water sources (Jinze Reservoir, Chenhong Reservoir, Qingcaosha Reservoir, Dongfeng Xisha Reservoir), 16 representative points in Fengxian, Jinshan, Yangpu, Jing'an, Pudong New Area, Huangpu, Chongming and Baoshan District were selected for sampling. The tap water used by the residents of these 16 points was collected for the time period from November 2020 to May 2021. The sampling frequency is once every two months, with a total of four sampling sessions. The distribution and details of the sampling points are shown in Table 1 and Figure 1.

2.1.2. Methods and Instruments

Chinese national standard "Standard Test Method for Drinking Water" GB/T 5750-2006 was used to analyze the water quality of the end water samples collected from the pipeline network. Total organic carbon in the water samples was determined using Analytik Jena multi N/C 2100 (Analytik Jena, Germany). The concentration of anions such as sulfate, nitrate and halide ions in water samples was determined by ion chromatography Metrohm 883 Basic IC plus (Metrohm, Switzerland). The concentrations of heavy metals in water samples were determined using an inductively coupled plasma mass spectrometer Agilent 7900 (Agilent, Japan).

Table 1. The deployment of sampling points for end-of-pipe water in Shanghai.

Drinking Water Source	District	Sampling
Jinze Reservoir	Jinshan	Tap water in a resident, tap water in industrial park
	Fengxian	Tap water in two residents
	Yangpu	Tap water in a university, tap water in a resident
Qingcaosha	Jing'an	Tap water in a resident, tap water in industrial park
	Pudong	Tap water in two residents
	Huangpu	Tap water in two residents
Chenhang Reservoir	Baoshan	Tap water in two residents
	Chongming	Tap water in two residents

**Figure 1.** Distribution of sampling points.

2.2. Simulation of Arsenic Contamination Experiments

2.2.1. Chemical Reagents and Instruments

Arsenic salt $\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$ and NaAsO_2 (purity > 98%) were purchased from Sigma-Aldrich. Stock solutions with As(V) and As(III) concentration of 100 mg L^{-1} , respectively, were prepared with the pure water from a Milli-Q device ($18.2 \text{ M}\Omega \text{ cm}$ at 25°C). The two stock solutions were stored in brown glass bottles under refrigeration for later experiments on drinking water contamination patterns. The concentrations of arsenic in water samples were determined using an inductively coupled plasma mass spectrometer Agilent 7900. The redox states and content of arsenic species in aqueous solution were measured by high-performance liquid chromatography-inductively coupled plasma-mass spectrometry Agilent 1200 infinity LC-Agilent 7900 ICP-MS (Agilent, Japan). For the

household water purification experimental device, a reverse osmosis water purifier from manufacturer A, a nanofiltration water purifier from manufacturer B and an ultrafiltration water purifier from manufacturer C were selected. Process diagrams are shown in Figure 2a–c and Tables S1 and S2

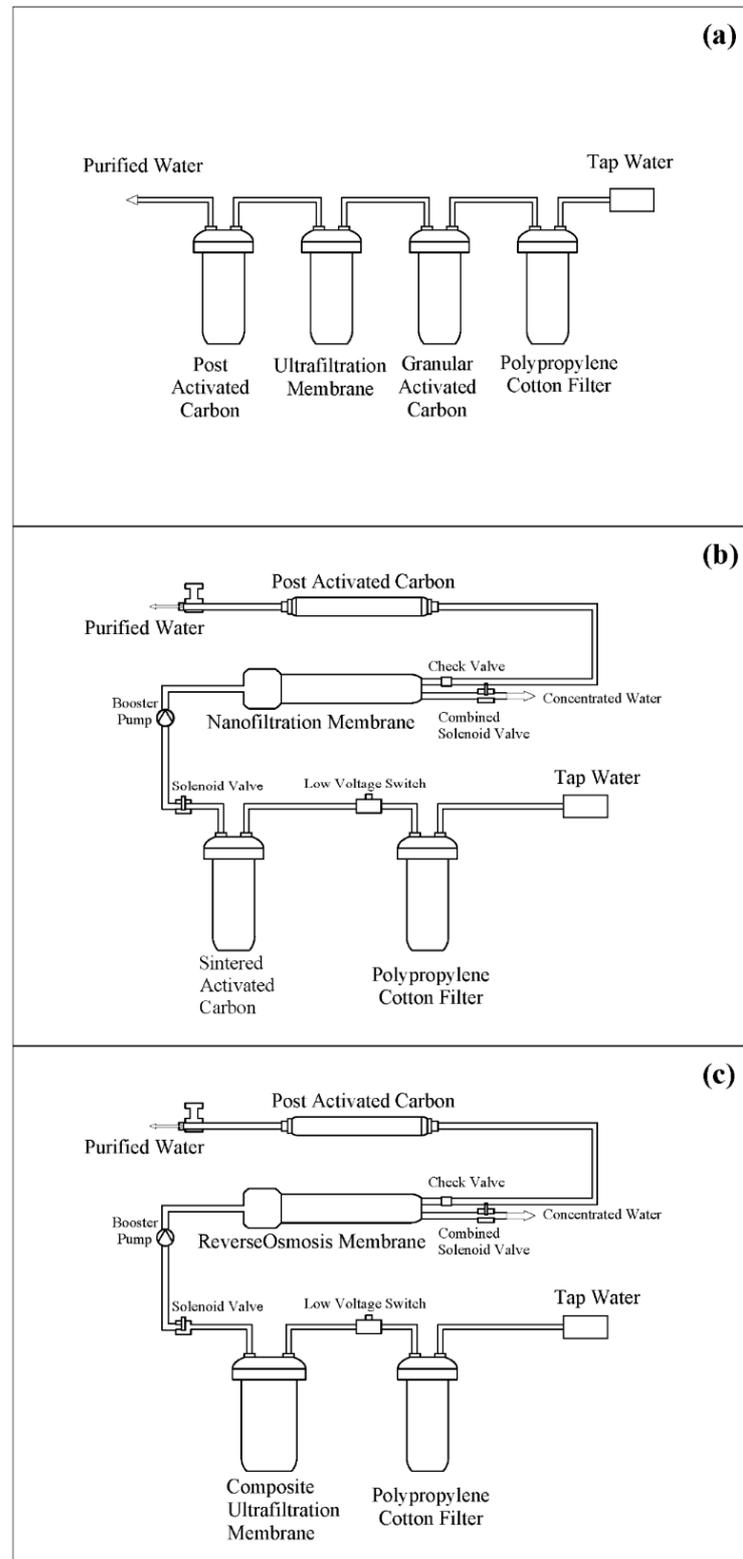


Figure 2. Domestic water purifier process flow chart: (a) ultrafiltration, (b) nanofiltration and (c) reverse osmosis.

2.2.2. Experimental Method of Pollutants Removal

Different amounts of As(III) or As(V) stock solutions were added to the tap water samples to simulate the arsenic contamination (pollution concentration range of 10–100 $\mu\text{g L}^{-1}$). Water samples containing arsenic were passed through several types of household water purifiers (0.24 MPa, 25 °C). The arsenic concentration in the effluent of each process unit and in the effluent of the water purifier was determined. HCl and NaOH were used to adjust the pH in the removal experiments under different pH conditions. In the experiments on the effect of coexisting ions, $\text{Ca}(\text{NO}_3)_2$, MgSO_4 and KCl were added to simulate different water conditions.

2.3. Health Risk Assessment

The lifetime carcinogenic risk via drinking freshly purified water was calculated on the following Equations (1) and (2) [24], and for the values of the parameters within the equations refer to the Chinese Handbook of Population Exposure Parameters (Adult Volume) [25]:

$$R_i^c = \frac{[1 - e^{-D_i \times q_i}]}{A_{ge}} \quad (1)$$

$$D_i = \frac{C_i \times IR}{BW} \quad (2)$$

where R_i^c is the lifetime carcinogenic risk via drinking freshly purified water, D_i is the daily As exposure dose due to oral intake of As-contaminated drinking water ($\text{mg kg}^{-1} \text{d}^{-1}$) and is calculated by the Equation (2), IR is the water ingestion rate (L day^{-1}) and is assumed to be 2.2 L, A_{ge} is the regional average life expectancy (year) of 83.6 year in Shanghai as recommended by Shanghai's Ministry of Health, q_i is the slope intensity factor of carcinogenicity by oral ingestion of carcinogens ($15 [\text{mg}/(\text{kg d})]^{-1}$) as recommended by USEPA [24], BW is the body weight (kg) and is assumed to 70 kg, C_i is the concentration of carcinogens.

3. Results and Discussion

3.1. Distribution of Heavy Metals in Water at the End of Shanghai Water Pipeline Network

The distribution of heavy metals in the water at the end of the water pipeline network in Shanghai is shown in Figure 3 and Figures S1–S13. The water quality of the end water of the Shanghai water pipeline network is good, and the concentration of heavy metals is below the limit value specified in China's drinking water standard GB 5749-2006. The concentrations of zinc, aluminum and iron in the end water of the pipe network are among the highest levels but the regional distribution is extremely uneven, relating to the sampling point pipe network materials and aging degree. Before the renovation of the pipe network in the old district of Huangpu, the pipe network may have rusted severely and thus the iron content in the end water of the pipe network was high, but after the renovation, the iron concentration in the end water of the pipe network dropped sharply. Chongming and Jinshan have higher concentrations of zinc and aluminum, which may be related to the materials of the pipe network at the user's home and appear to be elevated for a short period of time in the sampling month.

It is worth noting that arsenic was detected in all the tap water samples and, although the concentration is far below the requirements of the drinking water standards, its toxicity and carcinogenicity still require our attention. The distribution of arsenic concentrations can be seen in Figure 4 and Table S3, with total arsenic concentrations in tap water ranging from 0.479–1.195 $\mu\text{g L}^{-1}$, which is lower than the limit value for arsenic concentration set by China's drinking water standard GB 5749-2006 (10 $\mu\text{g L}^{-1}$) and by the World Health Organization (10 $\mu\text{g L}^{-1}$). It is noteworthy that the concentration of arsenic in the tap water samples from two water supply areas, Chen Hang Reservoir and Jin Ze Reservoir, was significantly higher than that of other samples.

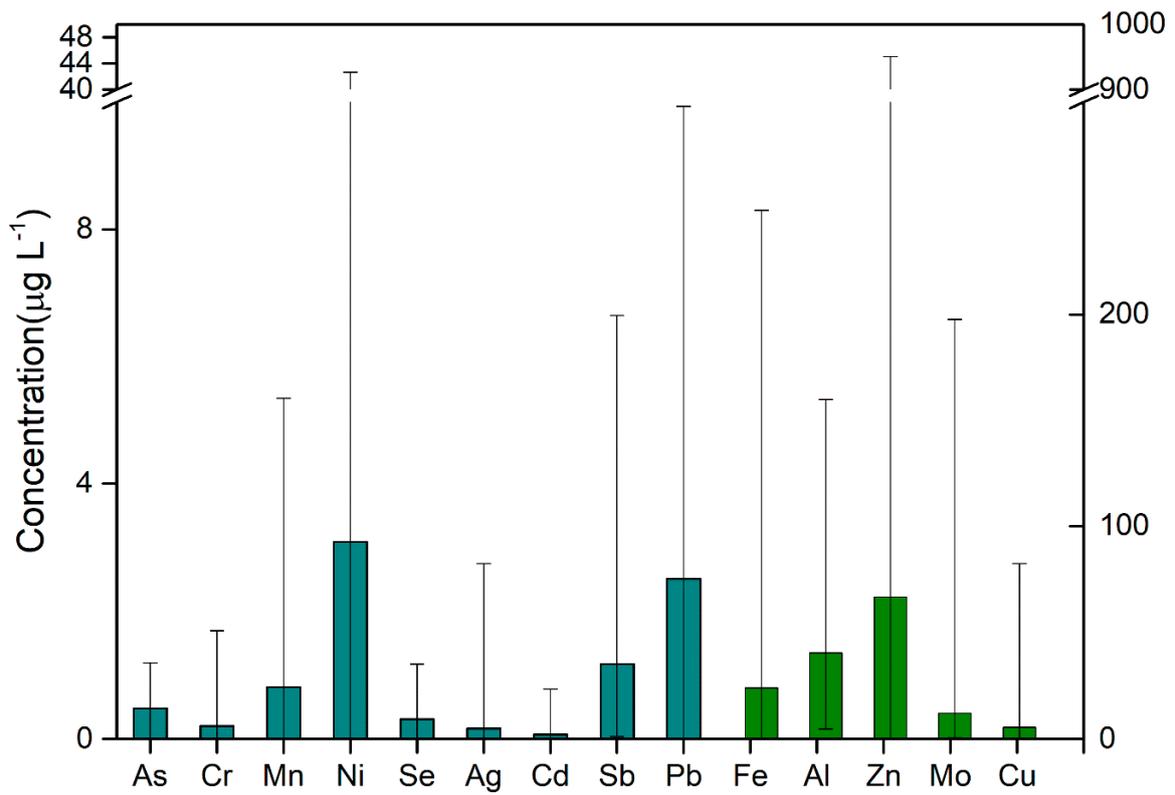


Figure 3. Distribution of heavy metal concentration in the end water of Shanghai pipeline network.

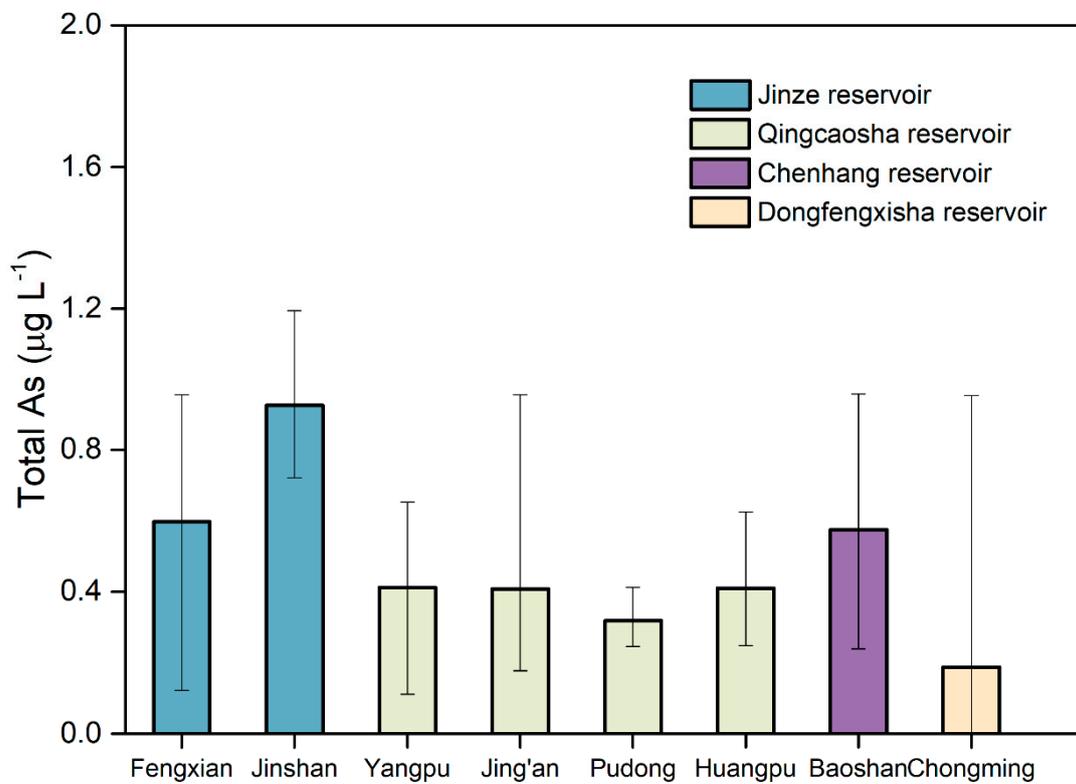


Figure 4. Distribution of total arsenic in the end water of Shanghai pipeline network.

3.2. Evaluating the Effectiveness of Home Water Purifiers for the Removal of Arsenic Contamination

3.2.1. Evaluation of Water Purifiers for the Removal of Arsenic in Different Valence States

As shown in Figure 5a,b, the removal rate of As(III) by ultrafiltration water purifiers ranged from 3.39 to 6.63%, and the removal rate of As(V) ranged from 6.07 to 15.3%. For reverse osmosis and nanofiltration water purifiers, the removal rate of As(III) is 40.1–56.3% and the removal rate of As(V) is 97.7%.

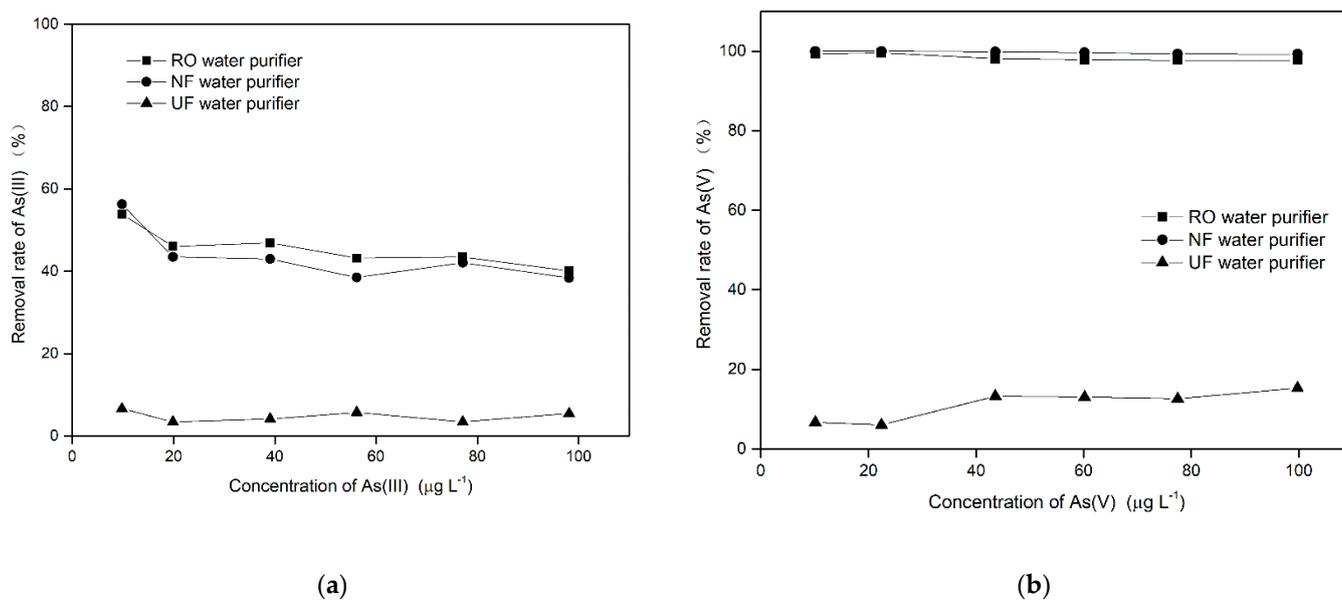


Figure 5. Removal efficiency of three types of water purifiers for (a) As(III) and (b) As(V).

The purification effect of different types of water purifiers varies somewhat and the removal efficiency increases when the initial concentration of arsenic contamination is high. However, water purifiers are not effective in the removal of arsenite (As(III)).

3.2.2. Evaluation of As(III) Removal Effect of Each Unit of Water Purifier

The treatment unit of a water purifier is generally divided into three parts, the pre-treatment unit, main unit and post-treatment unit. The main unit of a water purifier is the membrane treatment unit. The pre-treatment unit is generally a synthetic fiber cotton, sintered activated carbon and granular activated carbon; these three types of pre-treatment units mainly rely on coarse filtration and adsorption wherein the removal of arsenic is mainly related to the pore size, activated carbon type, hydraulic retention time, water volume and other factors. In general, granular activated carbon \approx sintered activated carbon $>$ synthetic fiber cotton. The post-treatment unit of the water purifier uses granular activated carbon which is intended for further protection after the core unit membrane, so the effect of removing contaminants is relatively weaker than that of the pre-treatment unit. It is worth noting that if the front core unit membrane is contaminated, broken or other accidents occur, the post activated carbon will adsorb the excess arsenic, thus causing secondary contamination of arsenic in the effluent water.

Since reverse osmosis water purifiers and nanofiltration water purifiers were more effective in removing As(V) (removal rate basically above 99.0%) and less effective in removing As(III), the detailed assessment of the removal effect of each unit of the water purifier on As(III) was evaluated and the results are shown in Table 2. The pre-treatment and post-treatment units were less effective in removing As(III), with removal rates of 0.83–9.74% for the pre-treatment unit. The removal rates of As(III) by nanofiltration and reverse osmosis membrane units ranged from 47.55 to 86.25%. The retention rate of As(III) by both membranes decreased significantly with the increase in use time. At present,

the ultrafiltration membrane used in the market is generally a composite membrane of activated carbon and ultrafiltration and the removal rate of As(III) has been improved; the removal rate is 9.98–15.48%.

Table 2. Removal effect of each unit of water purifier for different concentrations of As(III).

Water Purifier Process	Unit	Concentration of As(III)		
		10 µg L ⁻¹	50 µg L ⁻¹	100 µg L ⁻¹
Pre-treatment	Synthetic fiber	3.48–5.42%	1.42–3.85%	0.83–1.93%
	Sintered activated carbon	7.51–8.62%	4.51–7.77%	4.28–7.96%
	Granular activated carbon	4.82–8.59%	6.73–9.74%	4.15–6.69%
	Composite ultrafiltration membrane	12.34–14.55%	9.98–15.48%	10.94–13.04%
Main	Ultrafiltration membrane	9.40–11.17%	8.62–12.59%	9.92–11.94%
	Reverse osmosis membrane	57.48–86.25%	50.71–79.04%	47.55–65.00%
	Nanofiltration membrane	53.12–85.65%	49.9–79.39%	48.39–66.20%
Post-treatment	Post Activated carbon	3.73–4.90%	4.04–6.20%	2.07–3.01%

The effectiveness of membrane units in removing arsenic from water is related to the mass transfer process of the membrane and the morphology of arsenic in water [26,27]. The separation characteristics, mechanisms and operating conditions of three different membranes used in domestic water purifiers are shown in Table 3.

Table 3. Characteristics, mechanism and operating conditions of membrane separation, the core unit of water purifier.

Membrane	Pore Size	Category	Propulsion	Mechanisms	Interception Molecular Weight
Ultrafiltration membrane	5 nm–0.1µm	Composite	0.1–1.0 MPa	Screening, adsorption	1000–300,000
Nanofiltration membrane	1–5 nm	Composite	0.2–1.5 MPa	Spatial barrier effect, Donnan effect, adsorption/solubilization	100–1000
Reverse osmosis membrane	<1 nm	Composite	0.1–10 MPa	Diffusion	All ions

Ultrafiltration membranes generally retain substances with molecular weights greater than 1000 because of their larger pore sizes, and because As(III) is present as H₃AsO₃ and H₂AsO₃⁻ with molecular weights of 125–126, while As(V) is dominated by H₂AsO₄⁻ and HAsO₄²⁻ with molecular weights of 140–141, they are therefore less effective in retaining As(III) and As(V).

The mass transfer process of reverse osmosis and nanofiltration membranes is mainly determined by the spatial site resistance effect and the Donnan effect, which usually play a decisive role. The spatial site resistance effect is mainly realized through the pore size sieving effect, and the effective radius of the solute and the size of the membrane pore size determine whether the solute can enter the membrane or not. Configuring tap water with pH 7.33–7.65, As(III) is mainly present as H₃AsO₃ neutral molecules and As(V) as H₂AsO₄⁻. The radius of the hydrated ion of H₃AsO₃ is 0.24 nm and that of H₂AsO₄⁻ is 0.59 nm. In addition, the “critical potential” and “isoelectric potential” of RO and NF membranes occur in aqueous solutions with pH 4–6. The pH is less than this point, the membrane is positive, and if it is greater than this point, due to the anti-protonation effect, the surface of the membrane with negative points. The pH of drinking water is between 6–8 and the negatively charged membrane has a repulsive effect on negatively charged ions. Therefore, the nanofiltration membrane has a removal rate of 50–85% for As(III) and up to 99.0% for As(V).

3.2.3. Evaluation of Water Purifiers for the Removal of Arsenic in Mixed Valence

Trace arsenic in tap water exists in a mixed valence state [28] with trivalent arsenic accounting for a major portion of generally 50–90%. As shown in Figure 6a,b, the removal

rates of both As(III) and total arsenic from the household water purification units increased as the percentage of As(III) decreased. The reason for this may be the result of the interaction between trace organic matter in tap water and arsenic. This interaction usually requires complexation by small amounts of cations such as Ca^{2+} , Fe^{3+} , Mg^{2+} , etc., as cation bridges to form complexes and thus be intercepted by the membrane, hence the increased removal of arsenic [29].

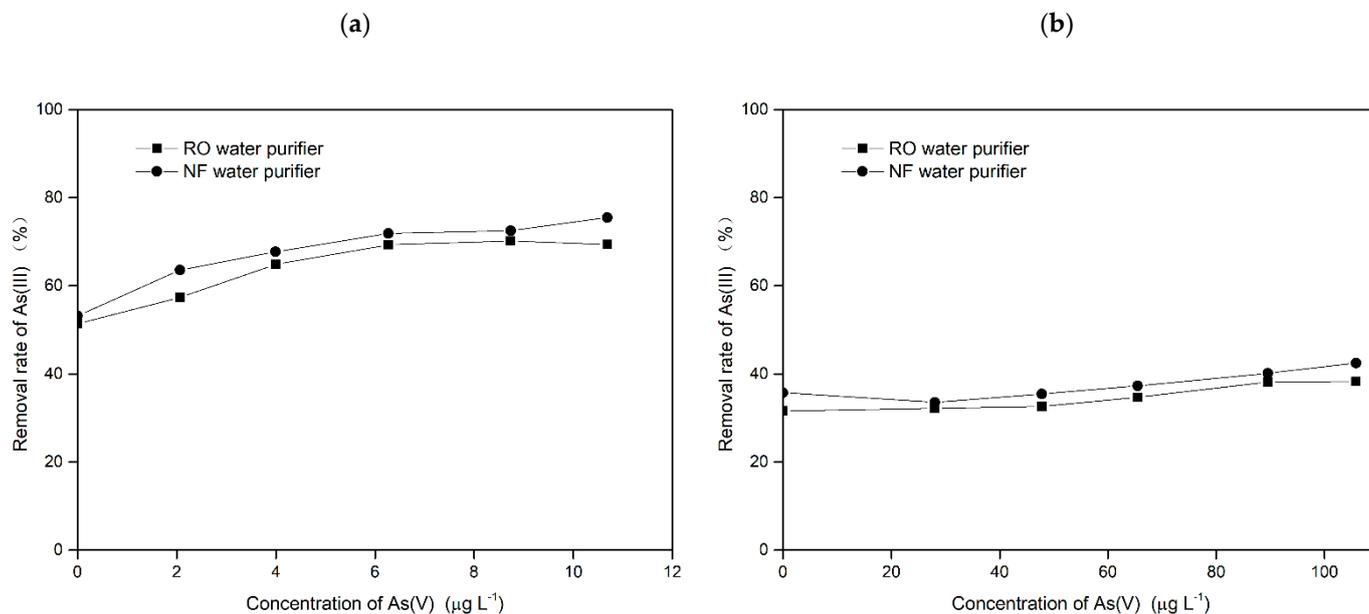


Figure 6. Effect of the percentage of As(V) in the mixed valence arsenic on the removal efficiency of As(III), (a) $C_{\text{As(III)}} = 10 \mu\text{g L}^{-1}$, (b) $C_{\text{As(III)}} = 100 \mu\text{g L}^{-1}$.

3.2.4. Effect of pH and Coexisting Ions

The effect of pH on the removal rate of arsenic in an environment where tap water simulates low concentration of arsenic pollution is shown in Figure 7a,b. When the pH is 6–8, the removal rate of As(III) by RO and NF membrane water purifiers is basically around 45%, and as the pH increases, the removal rate of As(III) by NF membrane water purifiers gradually increases. When the pH was 10, the removal rate of As(III) by NF membrane water purifier reached 88.2%, which doubled the removal effect. The presumed reason is that, when pH is 3.5–8.0, As(III) in tap water exists in the form of H_3AsO_3 and the removal mechanism of RO and NF membranes for As(III) is mainly particle size selective sieving, so the removal rate is basically unchanged. However, when pH increases, H_3AsO_3 is converted into the form of H_2AsO_3^- , and the negatively charged RO and NF membranes, to the negatively charged negatively charged ions, produce charge rejection, so the removal rate of As(III) increases. When the tap water is contaminated with arsenic, raising the pH of the incoming water may be beneficial to the removal of trace amounts of arsenic in the water by the home water purifier.

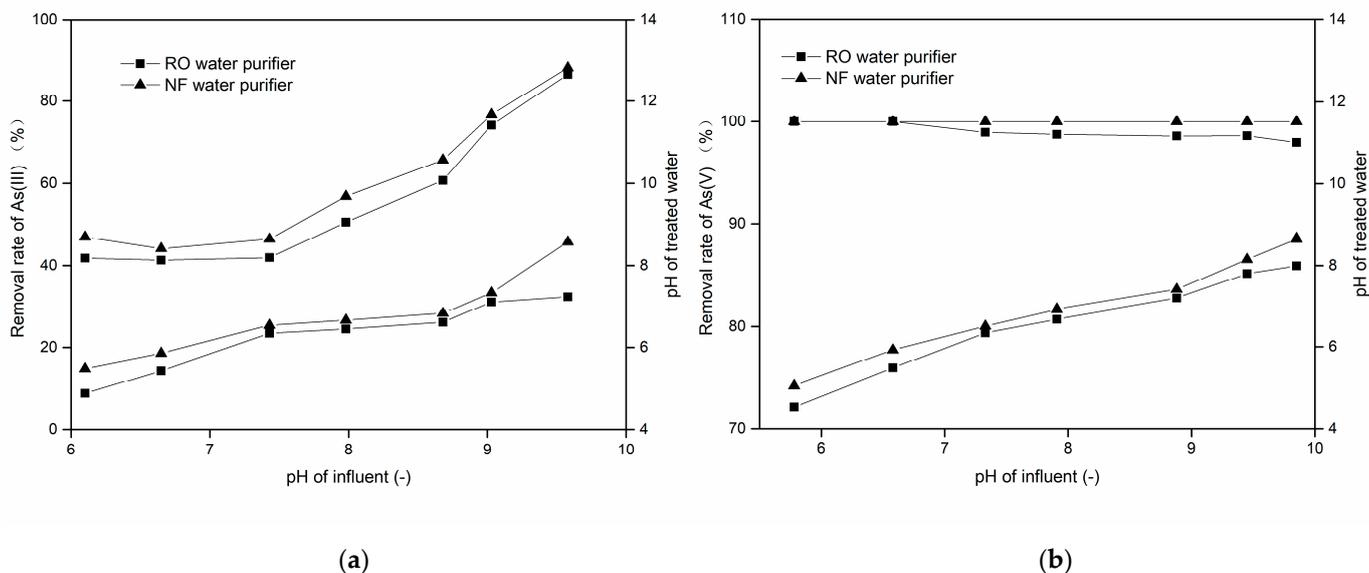


Figure 7. Effect of pH on the removal rate of (a) As(III), (b) As(V).

From Figure 8, it can be seen that the removal of As(III) increased with the increase in cation concentration, and the removal efficiency of arsenite was more significantly improved by the high valent salt. The reason for this may be that the presence of inorganic ions can compress the thickness of the double layer on the membrane surface, thus neutralizing or weakening the negative charge on the membrane surface, weakening the repulsion between the functional groups on the membrane surface and making the membrane pore size smaller to improve the removal capacity [30]. The effect of different cations on the removal of As(III) differs, with divalent cations having a greater effect on As(III) removal than monovalent cations. Notably, the effect of coexisting ions on nanofiltration water purifiers was greater than that of reverse osmosis water purifiers. In trace arsenic contaminated water, reverse osmosis and nanofiltration water purifiers, tap water with high hardness and high TDS were more effective in removing arsenic.

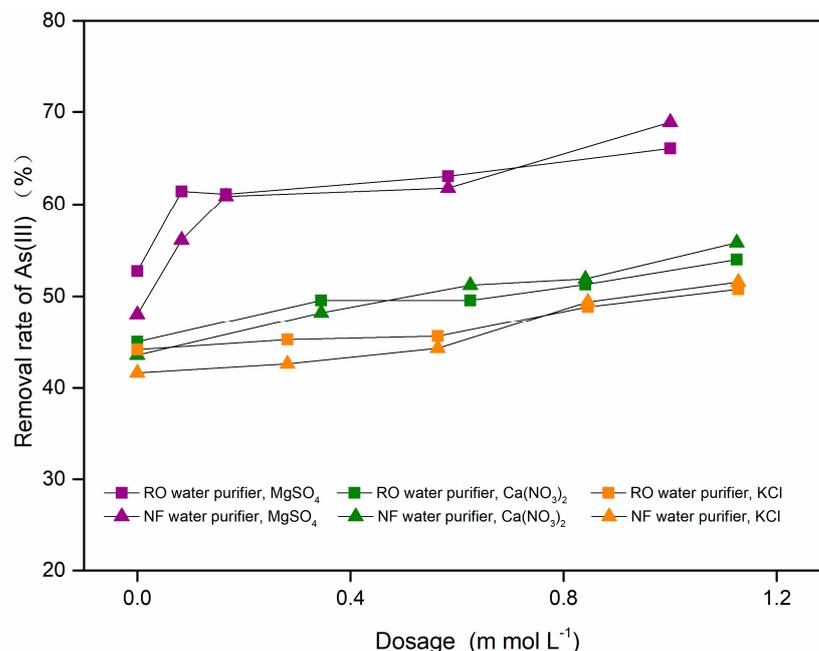


Figure 8. Effect of inorganic salt on the removal efficiency of As(III). The X-axis represents the molar concentration of the added salts (MgSO₄, Ca(NO₃)₂, KCl).

3.3. Health Risk Assessments

The results obtained from the carcinogenic health risk assessment of arsenic concentrations in tap water samples and household water purifier effluent, respectively, are shown in Table 4 and Table S4. As shown in Table 4, the assessed carcinogenic risk of tap water in Shanghai ranges from 0.254×10^{-6} – 6.74×10^{-6} . The carcinogenic risk is below the maximum acceptable risk level recommended by the US EPA for exposed populations. However, the risk values are still higher than the risk levels recommended by the Royal Society and the Swedish Environmental Protection Agency [31]. After passing through the home water purifier, the cancer risk assessment values were reduced for water from the water purifier with nanofiltration and reverse osmosis processes, which greatly reduced the risk of cancer caused by arsenic in drinking water.

Table 4. Carcinogenic risk at different concentration levels in each zone (year^{-1}).

District	Tap Water	RO	NF	UF
Fengxian	4.65×10^{-6}	1.61×10^{-6}	1.39×10^{-6}	3.86×10^{-6}
Jinshan	6.74×10^{-6}	2.18×10^{-6}	1.83×10^{-6}	5.10×10^{-6}
Yangpu	3.68×10^{-6}	1.09×10^{-6}	7.05×10^{-7}	2.72×10^{-6}
Jing'an	6.59×10^{-6}	1.69×10^{-6}	1.17×10^{-6}	5.24×10^{-6}
Pudong	1.78×10^{-6}	2.53×10^{-7}	2.54×10^{-7}	1.40×10^{-6}
Huangpu	1.40×10^{-6}	2.54×10^{-6}	2.54×10^{-7}	8.68×10^{-7}
Baoshan	4.96×10^{-6}	1.40×10^{-6}	1.17×10^{-6}	3.45×10^{-6}
Chongming	2.54×10^{-7}	2.54×10^{-7}	2.54×10^{-7}	2.54×10^{-7}

4. Conclusions

The sampling survey found that the survey area of the end of the pipe network water quality meets the standard. The total arsenic concentrations in different tap water samples range from 0.479 – $1.195 \mu\text{g L}^{-1}$. There is no risk of heavy metal overload. However, arsenic was detected in tap water. In addition, the concentration distribution had spatial and temporal distribution characteristics, with higher levels of arsenic in the water supply areas of Chenhang and Jinze reservoirs, and a need to reduce the carcinogenic risk of arsenic in tap water. The simulated and actual tap water samples were used to evaluate different types of household water purifiers. It was found that reverse osmosis and nanofiltration water purifiers were more effective in removing As(V), with a removal rate above 97.7%, and less effective in removing As(III), with a rate range from 40.1 to 56.3%, thus reducing the carcinogenic risk of arsenic in tap water. Raising the pH of the water was beneficial to the removal of trace amounts of arsenic in the water by the home water purifier.

Supplementary Materials: The following are available online at <https://www.mdpi.com/article/10.3390/w13202916/s1>. Figure S1: Distribution of total Ag in the end water of Shanghai pipeline network; Figure S2: Distribution of total Al in the end water of Shanghai pipeline network; Figure S3: Distribution of total Cd in the end water of Shanghai pipeline network; Figure S4: Distribution of total Cr in the end water of Shanghai pipeline network; Figure S5: Distribution of total Cu in the end water of Shanghai pipeline network; Figure S6: Distribution of total Fe in the end water of Shanghai pipeline network; Figure S7: Distribution of total Mn in the end water of Shanghai pipeline network; Figure S8: Distribution of total Mo in the end water of Shanghai pipeline network; Figure S9: Distribution of total Ni in the end water of Shanghai pipeline network; Figure S10: Distribution of total Pb in the end water of Shanghai pipeline network; Figure S11: Distribution of total Sb in the end water of Shanghai pipeline network; Figure S12: Distribution of total Se in the end water of Shanghai pipeline network; Figure S13: Distribution of total Zn in the end water of Shanghai pipeline network; Table S1: General filter elements and related technical parameters of water purifiers in market; Table S2: The technical parameters of the three water purifiers; Table S3: Spatial and temporal distribution of As in drinking water in each district; Table S4: Concentrations of total arsenic in drinking water and in the effluent of water purifiers in each district ($\mu\text{g L}^{-1}$).

Author Contributions: Q.Q.: Investigation, Data curation, Visualization, Writing—Original draft preparation; Z.Z.: Conceptualization, Validation, Supervision, Reviewing, Project administration, Funding acquisition; H.L.: Visualization, Writing—Original draft preparation, Editing; M.S.: Validation, Funding acquisition; Y.Q.: Validation; D.Y.: Supervision. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest.

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