

Original Research

Occurrence and Health Risk Assessment of Trace Metals in Desalinated Seawater Using Two Desalination Technologies

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Abstract

Desalination of seawater is an effective way to alleviate the world's water shortage. However, the transmission and health risk of trace metals in the different processes of seawater plants are highly uncertain. Here, we investigated the concentrations of 29 ions and trace elements in desalinated water based on two desalination techniques – multiple effect distillation (MED) and reverse osmosis (RO) – and evaluated the potential health risks (carcinogenic risk (CR) and non-carcinogenic risk (n-CR)) of trace metals in these processes. The results showed that most ions and trace elements were efficiently removed using both the RO and MED methods. After desalination, the trace metal concentrations in the desalinated water treated with the MED method were higher than those in the desalinated water treated with the RO method. The n-CR values for the trace metals in the desalinated water using the MED and RO methods were within the neglectable level. The CR values based on the two methods were lower than the maximal acceptable risk levels designated by the US Environmental Protection Agency. The total health risks using the MED method were higher than those using the RO method. Our results indicated that seawater desalination using both treatment methods is safe.

Keywords: desalination, seawater, health risk assessment, trace metals, reverse osmosis

Introduction

Water demand and the water shortage crisis are increasing worldwide due to the population and rapid

growth of the global economy, which are becoming very serious social and environmental problems [1]. According to the World Health Organization, by the year 2025 approximately 50% of the world's population will live in water-stressed regions. Therefore, other alternative methods (utilizing desalted seawater or recycling wastewater) have been invented for efficient water consumption [2]. Desalination of seawater is

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believed to be a reliable and pragmatic option to alleviate this situation, which is an emerging water resource to meet fresh water demands [3]. According to the International Desalination Association, 150 countries with a total of 18,426 desalination plants are operating worldwide, producing 40 million cubic meters per day (Mm^3/day) in 2013, 86.8 Mm^3/day in 2015, and reaching 88.6 Mm^3/day in 2016, providing water for 300 million people [4, 5]. Many countries with water shortage problems, such as Japan, Australia, Spain, China, and the USA, have already developed this new water resource [6]. China is also one of the largest water shortage areas in the world [1]. In 2014, more than 400 of 561 Chinese cities were deficient in water to some extent [7]. Statistically, by the end of 2016 more than 100 seawater desalination projects were completed in China, with a water production scale of 1.89 million tons per day and the largest seawater desalination project scale of 200,000 tons per day [8]. Electrodialysis (ED) and reverse osmosis (RO) methods are the two commonly used seawater desalination technologies [9, 10]. In fact, research on this topic was started in 1958 and 1965 in China [11, 12]. The Institute of Seawater Desalination and Multipurpose Utilization was also established in Tianjin in 1984. Since then, desalination technologies have significantly improved. In China, most of the completed desalination plants are located in Liaoning, Shandong, Hebei, Tianjin, and Zhejiang [7, 13].

Although seawater desalination has been widely used in many countries worldwide, its drawbacks are also obvious. For example, low water alkalinity and hardness and high salinity can lead to the corrosion of the facilities and water-transmission pipeline [14]. If desalinated seawater enters the municipal metal pipeline, the health risk of trace metals in the drinking water will be increased significantly. Therefore, health risk assessment of trace metals in desalinated water is essential for drinking water safety. However, previous studies have mainly focused on desalination technologies [15, 16] and the economic cost accounting and evaluation of project investments [17, 18]. Little research has been conducted on evaluating the health risks caused by trace metals during the process of seawater desalination, a topic that deserves further evaluation.

In this study, water samples were collected in two desalination plants in Northern China based on two typical desalination techniques (multiple effect distillation (MED) and RO). The objectives of the present study were to: (1) investigate the occurrence of trace metals in desalinated water based on the two desalination methods; (2) compare the trace metal concentrations in different process sections with the drinking water sanitary standard; and (3) evaluate the potential health risk (carcinogenic risk (CR) and non-carcinogenic risk (n-CR)) of trace metals in desalinated seawater.

Materials and Methods

Sample Collection

Water samples were collected from two desalination plants in Northern China based on the MED and RO methods in March 2017. In one desalination plant using the MED method, both seawater and desalted water were collected. In another desalination plant using the RO method, seawater, primary RO, secondary RO and desalted water were collected. At each sampling site, three water samples were collected. The water samples were collected in polymer polyethylene bottles, and transported to the laboratory. Thereafter, the water samples were filtered through 0.45- μm filter membranes and then acidified with ultrapure concentrated HNO_3 to a pH lower than 2. All the water samples were stored at 4°C until analyzed. The trace metal concentrations and other ions were measured using inductively coupled plasma-mass spectrometry (ICP-MS, Agilent 7700x) and ion chromatography (ICS-3000, Dionex). Hg was measured using an atomic fluorescence spectrometer (AFS-2202E, Beijing Haiguang Instrument Co., Ltd., Beijing, China). For quality control, the calibration curve was prepared using the mixed standard sample (GB1767). A standard reference material (Trace Element in Water, Agilent) was analyzed to check the accuracy of measurement. The recovery rate was within the recommended range (90-110%).

Health Risk Assessment

In this study, human health risk was assessed by applying US EPA methodology [19]. Human beings can be exposed to trace metals in water through direct ingestion and dermal absorption by the skin [20]. Compared with the other pathway, direct ingestion through drinking water is the most direct exposure route. According to the toxic characteristics of pollutants, health risk assessment includes n-CR and CR. In this study, Pb, Zn, Cu, Mn, Ni, Hg, and Se are non-carcinogens, and Cd, As and Cr are carcinogens. Human life can be divided into four stages: infant, teenager, adult, and elderly populations.

The n-CR is calculated by Eq. (1):

$$R^n = \sum_{k=1}^k R_k^n \quad (1)$$

$$R_k^n = (D_k/R_f D_k) \times 10^{-6}/Y \quad (2)$$

$$D_k = Q_i \times C_k / W_i \quad (3)$$

...where R^n is the total n-CR caused by non-carcinogens; R_k^n is the average year of n-CR of metal k by ingestion (a^{-1}); and $R_f D_k$ is the reference dose of the metal k ($\text{mg}/(\text{kg}\cdot\text{d})$).

The CR is calculated by the following equations:

$$R^c = \sum_{j=1}^j R_j^c \tag{4}$$

$$R_j^c = [1 - \exp(-D_j \times q_j)] / Y \tag{5}$$

$$D_j = Q_i \times C_j / W_i \tag{6}$$

...where R^c is the total CR caused by carcinogens; R_j^c is the average years of CR of the metal j by drinking water (a^{-1}); D_j is the daily exposure dose of metal j , ($mg/(kg \cdot d)$); q_j is the carcinogenic factor of metal j , ($mg/(kg \cdot d)$); and Y is the average lifetime (75.76 a). Q_i is daily water consumption (L); C_j is the concentration of metal j (mg/L); and W_i is body weight.

The hypothesis is that no synergistic relationship exists among the toxicities of the trace metals, and that total health risk is the sum of the n-CR and CR:

$$R = R^n + R^c \tag{9}$$

The values of model parameters q_i and $R_i D_k$ are exhibited in Table 1, and the values of Y , Q and W are shown in Table 2 [21].

Results and Discussion

Inorganic and Ion Parameters in Seawater before and after Desalination

MED has been used in process industries for a long time. Recently, the MED low-temperature operation using a thermal vapor compressor was developed [22]. The inorganic parameters in the seawater and desalted water are listed in Table 3. Relatively high removal efficiencies existed for the inorganic salt using both RO and MED. The removal of Na^+ , K^+ , Mg^{2+} and other negative ions all reached 99.9% by both the RO and MED methods, indicating that these two desalination

Table 1. Values of model parameters q_i and $R_i D_k$ [18].

Carcinogens	q_i ($mg/(kg \cdot d)$)	Non-Carcinogens	$R_i D_k$ ($mg/(kg \cdot d)$)
Cd	6.1	Pb	1.4×10^{-3}
As	15	Zn	0.3
Cr	41	Cu	5×10^{-3}
		Mn	0.02
		Sb	4×10^{-4}
		Ni	0.02
		Hg	4×10^{-4}
		Se	0.01

Table 2. Associated parameters in health risk assessment [18].

Parameter	Value
Y	75.76a
Q	1.0 L/d _i ; 2.0 L/d _{ii} ; 3.2 L/d _{iii} ; 2.7 L/d _{iv}
W	18.9 kg; 44.4 kg; 63.1 kg; 62.2kg

i: infant; ii teenager; iii: adult; iv: the elderly

techniques have high efficiencies in removing the above inorganic ions. Moreover, the removal of B by the RO method was higher than that by the MED method, resulting from the high elution membrane of B added to the second process of the RO method. Additionally, the pH value of the RO method was higher than that of the MED method (Table 3) and was attributed to the increase in the pH value in the second process of the RO method being the best method to control B concentrations. Thus, if the removal rate of B was increased by the MED method, the subsequent treatment technique should be coupled with the MED method.

Removal rate using the RO method ranged from 55.91% to 100% for most trace metals, values that were higher than those using the MED method. Notably, Al, Fe, Zn and Se concentrations in the second process by the RO method increased compared with that in the first process (Table 3) due to the addition of chemical reagents to prevent the pipeline's erosion by the RO method. A similar result was also found in a previous study [23]. In addition, Hg removal rate using the RO method was only 38.78%, and Hg concentrations in the first (0.015 $\mu g/L$) and second (0.011 $\mu g/L$) processes using the RO method were not obviously different from that of the seawater (0.018 $\mu g/L$), indicating that the residual Hg in the pipeline using the RO method should not be ignored (Table 3). In addition to inorganic contaminants, different types of organics can also be found in water (e.g., pesticides, herbicides, and personal care products) released by human activities [24]. Therefore, organics are important targets for membrane water treatment. In the last decade, the RO method also has been applied to remove synthetic organic matter [25]. However, due to the low-water permeability of its membranes, the removal rate was low. Further technology should develop high-permeability membranes.

Comparison of Trace Metals Concentrations in Desalted Water to Water Quality Standards

Comparison of different water quality standards to desalinated water is shown in Table 4. Before the desalted water was injected into the municipal pipeline, the metal concentrations of the desalted water should be detected to ensure drinking water quality. Comparison of the trace metal concentrations (As, Cd, Pb and Hg) of desalted water to the standard of drinking water quality in China revealed that the trace metal concentrations

Table 3. Removal comparison of inorganic parameters to different treatment methods.

Parameters	RO method					MED method		
	Seawater	First process	Second process	Desalinated water	Removal (%)	Seawater	Desalinated water	Removal (%)
Na mg/L	10781.1	151.403	1.8598	0.2773	99.99	9711.5	1.2021	99.988
K mg/L	392.8	6.406	0.0664	0.0083	99.998	320.8	0.0076	99.998
Mg mg/L	1089.9	3.437	0.0917	0.0306	99.997	963.8	0.0444	99.995
Ca mg/L	421.7	4.651	2.7589	2.4515	99.419	499.6	2.0742	99.585
F ⁻ mg/L	48.4	0.0011	0.0007	0.0384	99.921	8.2	0.0019	99.977
Cl ⁻ mg/L	20206.8	269.432	1.899	0.3597	99.998	15641.1	0.575	99.996
SO ₄ ²⁻ mg/L	2309.8	6.9305	0.1876	0.0545	99.998	1778.3	0.4301	99.976
NO ₃ ⁻ mg/L	2.6	0.0014	0.0012	0.0009	99.965	3.1	0.0003	99.990
CO ₃ ²⁻ mg/L	39.234	9.125	5.435	3.827	90.246	33.845	4.657	86.240
B mg/L	4.107	0.91	0.089	0.087	97.882	3.42	0.37	89.181
Li µg/L	20.533	2.065	0.088	0.045	99.78	37.943	0.114	99.70
Al µg/L	L.D.	L.D.	337.761	L.D.	--	--	--	--
V µg/L	2.608	0.070	0.051	0.007	99.75	18.785	5.431	71.09
Cr µg/L	0.776	0.094	0.063	0.045	94.15	0.926	1.140	
Mn µg/L	2.581	0.311	0.945	0.182	92.95	3.100	0.249	91.97
Fe µg/L	1.836	0.204	2.927	0.810	55.91	--	--	--
Co µg/L	0.157	0.012	0.011	0.011	92.71	0.982	0.012	98.78
Ni µg/L	0.803	0.314	0.094	0.123	84.73	33.553	0.167	99.50
Cu µg/L	2.138	0.013	0.126	L.D.	--	23.373	0.404	98.27
Zn µg/L	0.014	0.490	0.714	0.997	--	2.082	1.845	11.38
Ga µg/L	0.004	0.003	0.055	L.D.	--	0.321	0.068	78.82
As µg/L	1.405	L.D.	L.D.	L.D.	--	22.659	1.379	93.91
Se µg/L	L.D.	0.090	0.218	0.080	--	--	--	--
Rb µg/L	56.890	1.900	0.023	0.026	99.95	--	--	--
Cd µg/L	L.D.	L.D.	L.D.	L.D.	--	0.069	0.024	65.22
Tl µg/L	L.D.	L.D.	L.D.	L.D.	--	0.014	0.006	57.14
Pb µg/L	L.D.	L.D.	L.D.	L.D.	--	0.020	0.012	40.00
U µg/L	1.058	0.004	0.002	0.001	99.94	--	--	--
Hg µg/L	0.018	0.015	0.011	0.011	38.78	L.D.	L.D.	--

were all below the standard values (0.01 mg/L for As, 0.005 mg/L for Cd, 0.01 mg/L for Pb and 0.001 mg/L for Hg) (GB 5749-2006) [26]. For major metals (Al, Fe, Mn, Cu and Zn), the Al and Cu concentrations in seawater treated using the RO method were all lower than the detection limit. The concentrations of other elements were all lower than the standards of the GB 5749-2006 and standard of drinking water quality in Canada (TEL) [27]. The seldom monitored trace metals (Sb, Ba, Be, B, Ni and Tl) showed similar results indicating that seawater treated using these two treatment techniques

(MED and RO methods) all reached the drinking water standard in China (GB 5749-2006). Notably, the metal (As, Mn, Cu, Zn, Be, B and Tl) concentrations of desalted water treated using the MED method were higher than those using the RO method, indicating that the RO method might be better than the MED method.

Health Risk Assessment

The n-CR values for the trace metals in the desalted water using the MED and RO methods are presented in

Table 4. Comparison of trace metals in desalted water to different water quality standards.

Elements	^a GB 5749-2006	^b TEL	MED method	RO method
As (mg/L)	0.01	0.025	0.0014	^c L.D.
Cd (mg/L)	0.005	0.005	0.00002	L.D.
Pb (mg/L)	0.01	0.01	0.00001	L.D.
Hg (mg/L)	0.001	0.001	L.D.	0.00001
Se (mg/L)	0.01	0.01	L.D.	0.0001
Al (mg/L)	0.2		L.D.	L.D.
Fe (mg/L)	0.3	≤0.3	L.D.	0.0008
Mn (mg/L)	0.1	≤0.05	0.0002	0.0002
Cu (mg/L)	1.0	≤1.0	0.0004	L.D.
Zn (mg/L)	1.0	≤5.0	0.0018	0.0010
Sb (mg/L)	0.005	0.0061	0.014	L.D.
Ba (mg/L)	0.7	1.0	L.D.	L.D.
Be (mg/L)	0.002		0.00007	L.D.
B (mg/L)	0.5	5.0	0.03713	0.000001
Ni (mg/L)	0.02		0.00017	L.D.
Tl (mg/L)	0.0001		0.00001	L.D.

a: Standard of drinking water quality in China, GB 5749-2006;

b: Standard of drinking water quality in Canada, TEL;

c: Limit detection.

Table 5. Hg in the desalted water treated using the MED method was not detected; thus, its n-CR was neglected. The n-CR values using the MED method were in the following descending order: Cu>Pb>Ni>Mn>Zn. The total n-CR values for the infant, teenager, adult and elderly populations were 8.12×10^{-11} , 6.92×10^{-11} , 7.79×10^{-11} , and $6.66 \times 10^{-11} \text{ a}^{-1}$ respectively. The n-CR values were 3-4 orders of magnitude lower than the neglectable risk level recommended by The Netherlands and England [21, 28], indicating that n-CR caused by these metals could be neglected. For the RO method, Pb and Cu concentrations in the water samples were lower than the detection limit. Therefore, the n-CR values only considered Zn, Mn, Ni and Hg. The n-CR values of the four metals followed the order Hg>Mn>Ni>Zn, which was different from the result using the MED method. By contrast, Hg was the most significant contributor to non-carcinogenic risks in the desalted water using the RO method. The n-CR value of Hg was an order of magnitude higher than that of the other three metals. The total n-CR values based on the RO method were 3.23×10^{-11} , 2.75×10^{-11} , 3.09×10^{-11} , and $2.65 \times 10^{-11} \text{ a}^{-1}$ for the four sensitive groups, respectively; the values were all lower than that based on the MED method. Thus, the n-CR values using both desalination methods were neglected.

The CR values of Cd, As and Cr in the desalted water using the MED and RO methods are also exhibited in Table 5. The CR values of the three

metals using the MED method were in the following order: Cr>As>Cd. The CR values of Cr and As were two orders of magnitude higher than that of Cd. The result was similar to the CR value in the drinking water in Beijing [29]. The total CR values for trace metals using the MED method were 4.71×10^{-5} , 4.01×10^{-5} , 4.52×10^{-5} , and $3.87 \times 10^{-5} \text{ a}^{-1}$ for infant, teenager, adult, and elderly populations, respectively. The CR values using the MED method were above the maximal acceptable risk levels designed by the Netherlands, Sweden and England ($1 \times 10^{-6} \text{ a}^{-1}$), but in the range of the maximum allowed value provided by the International Commission on Radiological Protection (ICRP, $5 \times 10^{-5} \text{ a}^{-1}$) and the US EPA ($1 \times 10^{-4} \text{ a}^{-1}$) (Table 6). Regarding the RO method, Cd and As were not detected. Therefore, the CR was only caused by Cr. The CR values of Cr were 1.81×10^{-6} , 1.54×10^{-6} , 1.73×10^{-6} , and $1.48 \times 10^{-6} \text{ a}^{-1}$ for the four stages of life. Similar to the result using the MED method, the risks were lower than the maximum limits recommended by ICRP and the US EPA.

The total risks of the trace metals using the MED and RO methods are shown in Fig. 1. According to the assessment result, the total health risks of the trace metals in the desalted water using the MED method for infant, teenager, adult and elderly populations were 4.71×10^{-5} , 4.01×10^{-5} , 4.52×10^{-5} , and $3.87 \times 10^{-5} \text{ a}^{-1}$, respectively. The total risks of the trace metals using the RO method were 1.81×10^{-6} , 1.54×10^{-6} , 1.73×10^{-6} ,

Table 5. Carcinogenic and non-carcinogenic risks of metals through drinking water in the desalted water using the MED and RO method (a⁻¹).

	Infant	Teenager	Adult	The elderly
MED method				
Non-carcinogens				
Pb	5.99E-12	5.10E-12	5.74E-12	4.91E-12
Zn	4.30E-12	3.66E-12	4.12E-12	3.52E-12
Cu	5.64E-11	4.80E-11	5.41E-11	4.63E-11
Mn	8.69E-12	7.40E-12	8.33E-12	7.13E-12
Ni	5.83E-12	4.96E-12	5.59E-12	4.78E-12
Total n-CR	8.12E-11	6.92E-11	7.79E-11	6.66E-11
Carcinogens				
Cd	1.02E-07	8.70E-08	9.80E-08	8.39E-08
As	1.44E-05	1.23E-05	1.38E-05	1.18E-05
Cr	3.26E-05	2.78E-05	3.13E-05	2.68E-05
Total CR	4.71E-05	4.01E-05	4.52E-05	3.87E-05
Total health risk	4.71E-05	4.01E-05	4.52E-05	3.87E-05
RO method				
Non-carcinogens				
Zn	2.32E-12	1.98E-12	2.23E-12	1.90E-12
Mn	6.36E-12	5.41E-12	6.09E-12	5.21E-12
Ni	4.28E-12	3.65E-12	4.11E-12	3.51E-12
Hg	1.93E-11	1.64E-11	1.85E-11	1.58E-11
Total n-CR	3.23E-11	2.75E-11	3.09E-11	2.65E-11
Carcinogens				
Cr	1.81E-06	1.54E-06	1.73E-06	1.48E-06
Total CR	1.81E-06	1.54E-06	1.73E-06	1.48E-06
Total health risk	1.81E-06	1.54E-06	1.73E-06	1.48E-06

and $1.48 \times 10^{-6} \text{ a}^{-1}$, respectively. The health risk using the MED method was higher than that using the RO method. Overall, the n-CR and CR for the four stages of life were in the descending order of infant>adult>teenager>elderly population. The results

suggest that infants are the most sensitive population among human beings; they need more attention. The result was similar to that of Geng et al. [21]. Additionally, the order of magnitude for the total n-CR was 10^{-11} , indicating that n-CR in both sites can be

Table 6. The maximal acceptable risk level and neglectable level recommended by some organizations (a⁻¹) [18, 23].

Organizations	Maximum allowed risk level	Neglectable level	Comments
US EPA	1×10^{-4}	-	-
ICRP	5×10^{-5}	-	Radiation
Netherland	1×10^{-6}	1×10^{-8}	Chemical Contaminants
Sweden	1×10^{-6}	-	Chemical Contaminants
England	1×10^{-6}	1×10^{-7}	-

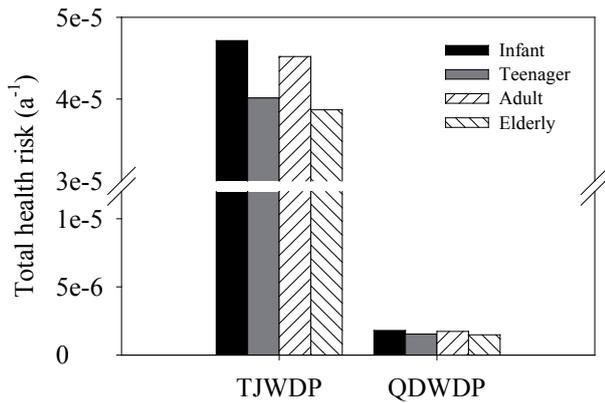


Fig. 1. Total health risks of trace metals in desalination water to four sections of the population using the MED and RO methods.

neglected. By comparison, the values of CR exceeded the limit value recommended by some countries. The results demonstrated that the non-carcinogenic metals rarely have harmful effects on human beings. The main risk came from the CR. Among the carcinogenic metals, Cr was the major contributor to health risk. According to part 3.1, the concentrations of Cr in both sites were within the standard for drinking water, but their CR cannot be ignored. The phenomenon was caused by its high carcinogenic factor ($q_{Cr} = 41$). Therefore, it is imperative to assess the health risk of metals in drinking water.

Conclusions

The RO and MED methods were both efficient in the removal of ions and trace metals, while some metals were not efficiently removed (Hg for the RO method and Zn for the MED method). The trace metal concentrations in the treated seawater all reached the drinking water standard in China (GB 5749-2006). After desalination, according to the trace metal concentrations in the desalinated water, the RO method might be better than the MED method. For both methods, the n-CR values for the trace metals in the desalted water were within the neglectable level, while the CR values were higher than the maximal acceptable risk levels designed by some countries and institutions. The total health risk using the MED method (the order magnitude is 10^{-5}) was higher than that using the RO method (the order magnitude is 10^{-6}). In addition, infants needed more attention due to their sensitivity. Although Cr has a low concentration in desalinated water, it was the major contributor to health risk due to its high carcinogenic factor. Evaluation of the health risks of metals is imperative in drinking water. Both seawater desalination methods are safe for drinking water.

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Conflicts of Interest

The authors declare no conflicts of interest.

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