



# Outdoor PM<sub>2.5</sub> and their water-soluble ions in the Northern part of the Persian Gulf

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## Abstract

**Background:** The environmental conditions potentially predispose the northern part of the Persian Gulf to the occurrence of dust storms. Outdoor PM<sub>2.5</sub> and their water-soluble ions in Bushehr port were studied from December 2016 to September 2017.

**Methods:** A total of 46 outdoor PM<sub>2.5</sub> samples were collected by high-volume air sampler and eight water-soluble ions, including Ca<sup>2+</sup>, Mg<sup>2+</sup>, Na<sup>+</sup>, K<sup>+</sup>, F<sup>-</sup>, Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, and SO<sub>4</sub><sup>2-</sup> in PM<sub>2.5</sub> were also measured by ion chromatography (IC).

**Results:** The 24-hour average concentration of PM<sub>2.5</sub> was in the range of 22.09 to 292.45 µg/m<sup>3</sup>. The mean concentration of water-soluble ions in PM<sub>2.5</sub> was in the range of 0.10 ± 0.14 to 6.76 ± 4.63 µg/m<sup>3</sup>. The major water-soluble ions were the secondary inorganic aerosols (SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup>), which accounted for nearly 41% of total water-soluble ions in PM<sub>2.5</sub>. The total water-soluble ions level of PM<sub>2.5</sub> in winter was higher than that in spring and summer. The positive matrix factorization (PMF) model showed that the source contributions of PM<sub>2.5</sub> were in the order of dust (55.8%), sea salt (17.1%), secondary sulfate (11.8%), industries (7%), vehicular emission (4.7%), and secondary nitrate (3.7%).

**Conclusion:** According to the results, dust and sea salt are the main sources of water-soluble ions in PM<sub>2.5</sub> in Bushehr port, which should attract much attention.

**Keywords:** Aerosols, Bushehr, PM<sub>2.5</sub>, Seasonal variation, Water-soluble ions

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## Introduction

Particulate matter (PM) is one of the air pollutants that is emitted into the atmosphere from different primary or secondary, natural or anthropogenic sources (1,2). Lately, the PM in outdoor air has been categorized as carcinogenic to humans (IARC Group 1) by the International Agency for Research on Cancer (IARC) (3). Outdoor PM can enter the human body via inhalation and ingestion and cause adverse health effects such as lung cancer, atherosclerosis,

and respiratory and cardiovascular diseases (4-8). Particle size, surface area, and chemical composition are among the properties of PM that can influence the health effects (9,10).

PM with an aerodynamic diameter smaller than 2.5 µm (PM<sub>2.5</sub>) can penetrate deeply into the lung area and deposit on the respiratory tract because of their diameter size (11-13). PM<sub>2.5</sub> may be generated and emitted into outdoor air from different sources such as power plants,



waste incineration, crustal and sea salt, vehicle emission construction, agricultural dust, and industrial processes (14,15).  $PM_{2.5}$  emitted directly from a source into the atmosphere are called primary  $PM_{2.5}$  and some process such as the oxidation of primary gases including, nitrogen oxides, ammonia, volatile organic compounds (VOCs) and sulfur dioxide can produce secondary particles (14,16). Outdoor  $PM_{2.5}$  consists of various chemical compounds such as metals, organic carbons, polycyclic aromatic hydrocarbons, and organic and inorganic ions (17-20). It was reported that nitrate and sulfate may be carried by  $PM_{2.5}$  and cause various effects including the formation of reactive oxygen species (ROS), cardiovascular disease as well as oxidative stress (21-24).

Bushehr port with its hot and humid weather is located in southwestern Iran along the northern side of the Persian Gulf. Because of the existence of oil and gas resources as well as related petrochemical industries in Bushehr province, this area is considered a sensitive area to air pollution (25,26). Dust storms may also transfer PM and other pollutants from the Arabian Desert located in the southern part of the Persian Gulf. But to date, there is no information on the water-soluble ion content of  $PM_{2.5}$  in this part of the Persian Gulf. Therefore, the aims of this study were: (1) to determine water-soluble ions contents of  $PM_{2.5}$  including  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $Na^+$ ,  $K^+$ ,  $F^-$ ,  $Cl^-$ ,  $NO_3^-$ , and  $SO_4^{2-}$ , (2) to discuss seasonal variations of water-soluble ions contents in  $PM_{2.5}$ , and (3) to identify the source of  $PM_{2.5}$  using positive matrix factorization (PMF) model.

## Materials and Methods

### Study area and $PM_{2.5}$ sampling

This study was performed to investigate the water-soluble ion characteristics of  $PM_{2.5}$  in Bushehr port. The sampling station was located in Bushehr University of Medical

Sciences building. Figure 1 shows the location of the sampling station in Bushehr port. The outdoor 24-hour samples were collected every six days from December 2016 to September 2017 for 9 months. During the sampling period, 46 samples were collected for investigating water-soluble ions of  $PM_{2.5}$ . Samples were collected on quartz fiber filters (8 in  $\times$  10 in, USA, pore size: 2.5  $\mu m$ ) using a  $PM_{2.5}$  Tisch high-volume sampler (Model TE-6070D, USA) at a flow rate of 1.42 to 1.58  $m^3/min$ . The meteorological data including temperature, relative humidity, and wind speed were collected from meteorological stations near the sampling station in Bushehr port during the sampling period. The range of temperature, relative humidity, and wind speed were 8.12–42.2° C, 31.93–78.12%, and 1.62–9.25 m/s, respectively.

### Chemical analysis

Before and after sampling, each filter was kept in a dehydrated desiccator for 48 h under a relative humidity of 25-30% and temperature of 20-25 °C (27,28). To specify  $PM_{2.5}$  mass concentration, the filters were weighted three times before and after the sampling using an electronic microbalance with a sensitivity of 0.1 mg. The  $PM_{2.5}$  mass concentration was estimated using Eq. (1):

$$PM_{2.5} (\mu g/m^3) = \frac{W_f - W_i \times 10^6}{V} \quad (1)$$

Where  $W_f$  and  $W_i$  are the weights of the filter before and after the sampling (g),  $V$  represents the volume of the air sample ( $m^3$ ), and  $10^6$  is a factor for converting grams to micrograms.

After weighing, the filters containing  $PM_{2.5}$  particles were packed and reserved at  $-18^\circ C$  until the extraction and chemical analysis. Then, the filters were sliced into four equal fractions and one-fourth of the filter was used

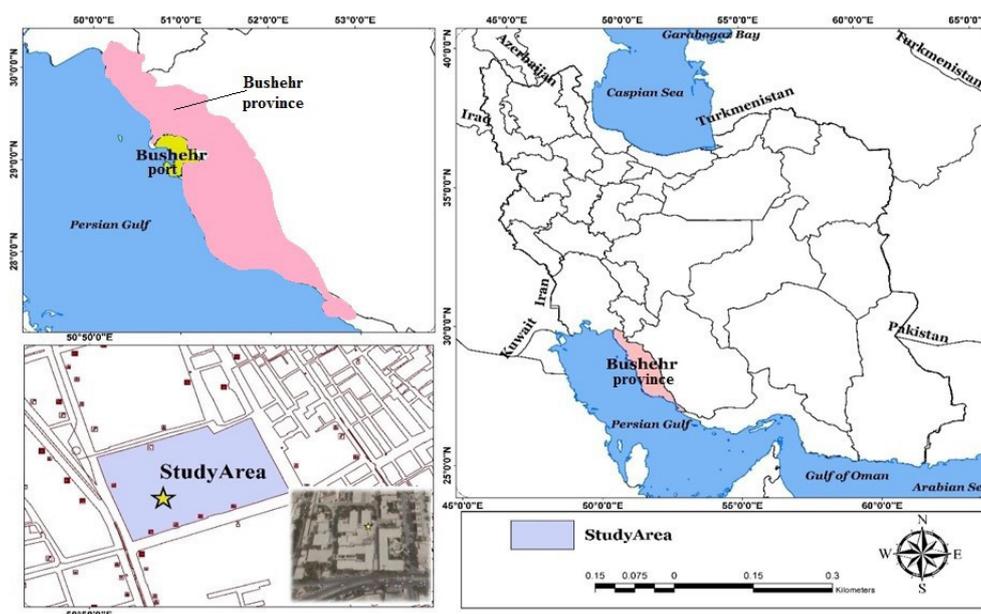


Figure 1. The map of the study area represents the sampling station (17)

to determine water-soluble ions ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{F}^-$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ) contents of  $\text{PM}_{2.5}$  particles. One-fourth of each sample filter was sliced and all fragments were added to a glass vial and 50 ml of ultrapure water was added. Then, the vial was shaken for 60 minutes and after that was ultrasonicated for 30 min to complete extraction. The extracted solution was filtered using a microporous membrane (pore size: 0.45  $\mu\text{m}$  and diameter: 25 mm), and the solution was reserved in plastic vials at  $-4^\circ\text{C}$  until chemical analysis (29). The water-soluble ions including  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{F}^-$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ , and  $\text{SO}_4^{2-}$  were analyzed using an ion chromatograph (Metrohm 850 Professional IC, Switzerland). The data quantification process was performed using an external standard method. The standard curves were graphed according to the five-concentration gradient of the standard. The correlation coefficient ( $r^2$ ) of calibration curves for each ion species was higher than 0.99.

### *PM<sub>2.5</sub> source apportionment*

The PMF is a receptor model developed by the Environmental Protection Agency (EPA), which is used to reconstruct the contribution of emissions from different sources of atmospheric pollutants (e.g.,  $\text{PM}_{2.5}$ ) based on ambient measurement data (i.e.,  $\text{PM}_{2.5}$  chemical composition). The latest version of PMF 5.0 decomposes a matrix of spectate sample data into factor contributions and factor profiles (30,31). In this study,  $\text{PM}_{2.5}$  sources during the sample collection period were quantified by the model. In total, eight ion species ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{F}^-$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ , and  $\text{SO}_4^{2-}$ ) were applied in the PMF model. It should be noted that PMF was created to run in robust mode and eight-factor solution was achieved from the base run. Both Q robust and Q true values were found to be in a fair agreement that this implicates the stability of the model and proficiency to reasonably fit all the variables.

### *Statistical analysis*

The SPSS 19 software and Microsoft Excel 2016 were used for statistical analysis. The descriptive statistics and the spatial-temporal distribution pattern were applied to analyze  $\text{PM}_{2.5}$  and its water-soluble ions contents. Furthermore, Pearson's correlation analysis was employed to identify the relationships between  $\text{PM}_{2.5}$  and ionic species. Statistical significance was accepted at  $P < 0.05$  and  $P < 0.01$ .

## **Results**

### *PM<sub>2.5</sub> mass concentration*

The  $\text{PM}_{2.5}$  concentration ranged from 22.09 to 292.45  $\mu\text{g}/\text{m}^3$  with a mean concentration of 65.77  $\mu\text{g}/\text{m}^3$ . During the sampling period, just 2 samples were lower than the WHO guideline (32) and 8 samples were lower than the EPA guideline (33). The highest concentration of  $\text{PM}_{2.5}$

(292.44  $\mu\text{g}/\text{m}^3$ ) was reported in April (19,34).

### *Mass concentration of water-soluble ions*

The descriptive statistics for  $\text{PM}_{2.5}$  water-soluble ions ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{F}^-$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ , and  $\text{SO}_4^{2-}$ ) during the sampling period are presented in Table 1. The concentrations of water-soluble ions including  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{F}^-$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ , and  $\text{SO}_4^{2-}$  were in the range of 0.97–8.43, 0.07–0.64, 4.29–8.12, 2.63–4.3, 0.02–0.84, 0.004–4.58, 0.004–22.77, and 0.02–23.55  $\mu\text{g}/\text{m}^3$ , respectively.

Figure 2 shows the contribution of each water-soluble ions to the total mass of water-soluble ions and  $\text{PM}_{2.5}$ , respectively. As shown in this figure, the order of contribution of water-soluble ions to the total mass of ions as well as  $\text{PM}_{2.5}$  was as:  $\text{SO}_4^{2-} > \text{Na}^+ > \text{NO}_3^- > \text{Ca}^{2+} > \text{K}^+ > \text{Cl}^- > \text{Mg}^{2+} > \text{F}^-$ . The ions  $\text{SO}_4^{2-}$ ,  $\text{Na}^+$ ,  $\text{NO}_3^-$ ,  $\text{Ca}^{2+}$ , and  $\text{K}^+$  were the most abundant water-soluble ions in the study area.

### *Temporal and seasonal trends of water-soluble ions in PM<sub>2.5</sub>*

Temporal and seasonal variations of the water-soluble ions ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{F}^-$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ , and  $\text{SO}_4^{2-}$ ) in  $\text{PM}_{2.5}$  are shown in Figures 3 and 4. As can be seen in Figure 3, no specific temporal trends were observed for any of the water-soluble ions. Almost the concentrations of all water-soluble ions except  $\text{Na}^+$  and  $\text{K}^+$  had a sharp peak during the sampling period. Six components of water-soluble ions including  $\text{Ca}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ , and  $\text{SO}_4^{2-}$  had a concentration peak in dusty days during the sampling period.

Bivariate correlations were performed to assess the relationships between  $\text{PM}_{2.5}$  and ionic species as shown in Table 2. The correlation analysis indicated that  $\text{PM}_{2.5}$  level was positively correlated with  $\text{NO}_3^-$  and  $\text{Ca}^{2+}$ .  $\text{Na}^+$  showed a positive correlation with  $\text{NO}_3^-$ ,  $\text{Ca}^{2+}$ , and  $\text{K}^+$ . Also, a positive correlation was observed between  $\text{K}^+$  and  $\text{Cl}^-$ .  $\text{SO}_4^{2-}$  illustrated a negative relationship with  $\text{F}^-$  and a positive correlation with  $\text{Cl}^-$ . Moreover,  $\text{Cl}^-$  had a positive correlation with  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , and  $\text{K}^+$ .  $\text{Ca}^{2+}$  was positively related to  $\text{NO}_3^-$  and  $\text{Mg}^{2+}$ .

**Table 1.** Summary statistics of  $\text{PM}_{2.5}$  and their water-soluble ions contents ( $\mu\text{g}/\text{m}^3$ )

Species	Mean	SD	Min	Max	Median	IQR
$\text{PM}_{2.5}$	65.77	49.84	22.09	292.45	52.8	29.13
$\text{Ca}^{2+}$	3.68	1.33	0.97	8.43	3.49	1.25
$\text{Mg}^{2+}$	0.28	0.14	0.07	0.64	0.27	0.25
$\text{Na}^+$	5.38	1.01	4.29	8.12	4.82	1.4
$\text{K}^+$	3.55	0.43	2.63	4.3	3.62	0.55
$\text{F}^-$	0.10	0.14	0.02	0.84	0.05	0.05
$\text{Cl}^-$	2.06	1.21	0.004	4.58	2.33	1.26
$\text{NO}_3^-$	3.88	3.70	0.004	22.77	3.52	3.05
$\text{SO}_4^{2-}$	6.76	4.63	0.02	23.55	6.27	4.59

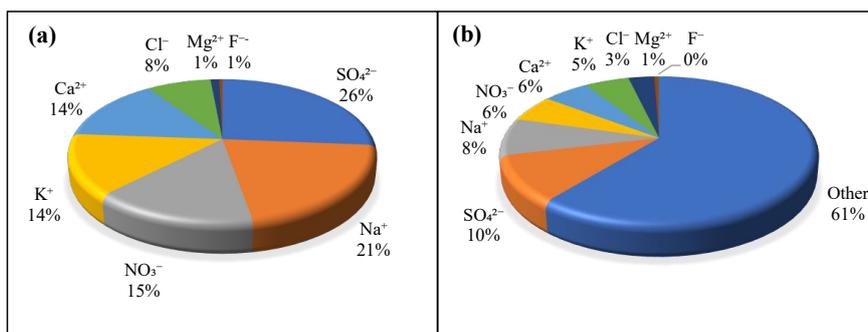


Figure 2. Relative contribution of water-soluble ions to the total mass of ions (a) and  $PM_{2.5}$  (b)

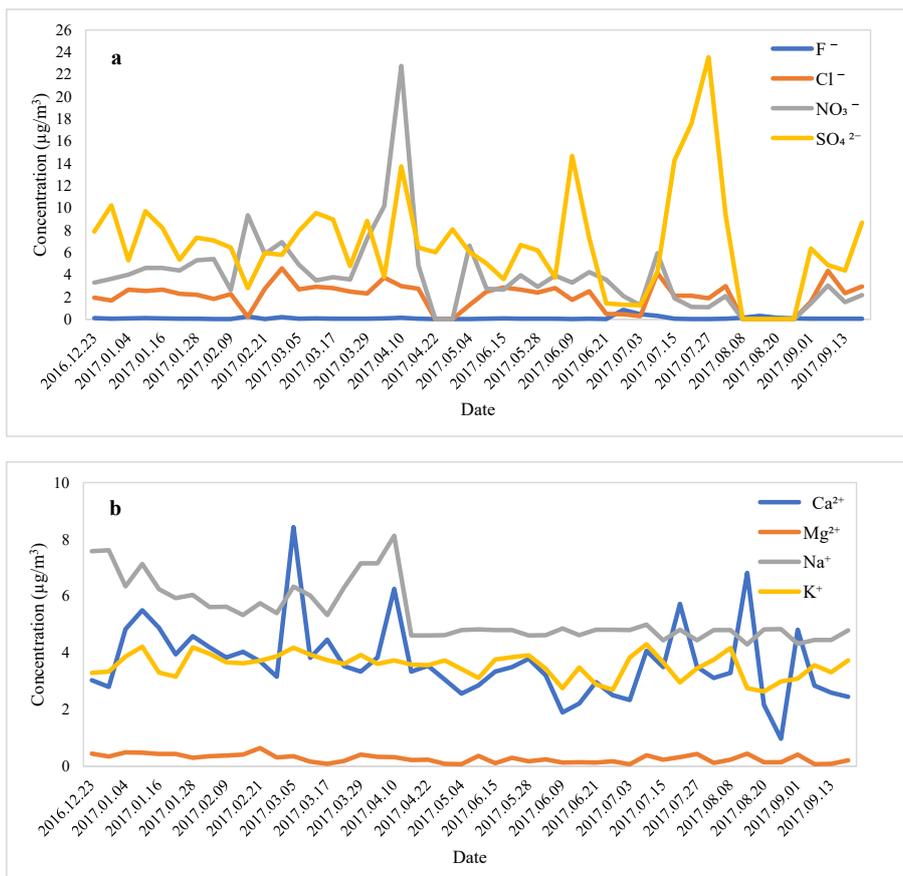


Figure 3. Temporal variations of the water-soluble ions, a: Anions and b: Cations

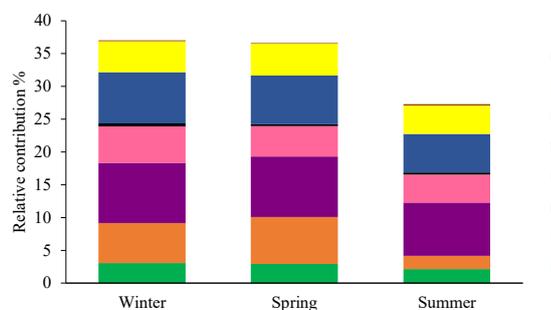


Figure 4. Seasonal variation of water-soluble ions species

Table 2. Correlation coefficient among  $PM_{2.5}$  and ionic species

	$PM_{2.5}$	$F^-$	$Cl^-$	$NO_3^-$	$SO_4^{2-}$	$Ca^{2+}$	$Mg^{2+}$	$Na^+$	$K^+$
$PM_{2.5}$	1								
$F^-$	-0.002	1							
$Cl^-$	0.14	-0.26	1						
$NO_3^-$	0.54**	-0.01	0.41**	1					
$SO_4^{2-}$	0.19	-0.37*	0.3*	0.17	1				
$Ca^{2+}$	0.51**	-0.05	0.18	0.32*	0.21	1			
$Mg^{2+}$	0.06	-0.04	0.16	0.25	0.18	0.51**	1		
$Na^+$	0.25	-0.09	0.27	0.62**	0.17	0.34*	0.46**	1	
$K^+$	0.24	-0.19	0.43**	0.28	0.09	0.29	0.13	0.28	1

\*\*Correlation is significant at the 0.01 level.

\*Correlation is significant at the 0.05 level.

### Source apportionment by PMF model

The sources of  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{F}^-$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ , and  $\text{SO}_4^{2-}$  in  $\text{PM}_{2.5}$  were quantified by PMF 5.0 model, and six factors including secondary sulfate (factor 1), dust (factor 2), industries (factor 3), vehicular emission (factor 4), secondary nitrate (factor 5), and sea salt (factor 6) were identified. The sources of  $\text{PM}_{2.5}$  for the six factors were 11.8%, 55.8%, 7%, 4.7%, 3.7%, and 17.1%, respectively (Figure 5). The factors profile and the percentage of  $\text{PM}_{2.5}$  and species apportion to each source are presented in Figures 6 and 7, respectively.

### Discussion

#### Water-soluble ions in $\text{PM}_{2.5}$ and temporal trends

According to Figure 2,  $\text{SO}_4^{2-}$ ,  $\text{Na}^+$ ,  $\text{NO}_3^-$ ,  $\text{Ca}^{2+}$ , and  $\text{K}^+$  accounted for approximately 90% and 35% of the total mass of water-soluble ions and  $\text{PM}_{2.5}$ , respectively. Shahsavani et al (35) reported that ionic components in Ahvaz, Iran, were in order of  $\text{SO}_4^{2-} > \text{Ca}^{2+} > \text{NO}_3^- > \text{Mg}^{2+} > \text{Cl}^- > \text{Na}^+ \sim \text{NH}_4^+ > \text{K}^+ > \text{F}^- > \text{NO}_2^-$  in TSP and  $\text{Ca}^{2+} > \text{NO}_3^- > \text{SO}_4^{2-} > \text{Mg}^{2+} > \text{Cl}^- > \text{Na}^+ \sim \text{NH}_4^+ > \text{K}^+ > \text{F}^- > \text{NO}_2^-$  in  $\text{PM}_{10}$ . Also,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , and  $\text{Cl}^-$  were respectively the most abundant ions in TSP and  $\text{PM}_{10}$ . Naimabadi et al (36) reported that the most frequent water-soluble ions in  $\text{PM}_{10}$  in Ahvaz, Iran, were  $\text{SO}_4^{2-}$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ , and  $\text{Ca}^{2+}$ , and contributions of water-soluble ions were in order of  $\text{SO}_4^{2-} > \text{NO}_3^- > \text{Na}^+ > \text{Ca}^{2+} > \text{Cl}^- > \text{K}^+ > \text{Mg}^{2+}$ . Goudarzi et al (37) reported that water-soluble ions in Ahvaz, Iran, were in order of  $\text{SO}_4^{2-} > \text{NO}_3^- > \text{Na}^+ > \text{Ca}^{2+} > \text{Cl}^- > \text{K}^+ > \text{Mg}^{2+}$ .

In Table 3, the concentrations of water-soluble ions in the present study are compared with those in other studies in different countries. According to the results, Deng et al (38), Hien et al (39), Sharma et al (40), Khan et al (41), and Remoundaki et al (42) reported  $\text{SO}_4^{2-}$  as the most abundant ion in PM between all water-soluble ions. In contrast, Zhao et al (43), Hassanvand et al (29), and Saraga et al (44) reported  $\text{NO}_3^-$  as the most abundant ion in PM.

The sea salt species of water-soluble ions ( $\text{Na}^+$  and  $\text{Cl}^-$ ) accounted for almost 29% of the total mass of

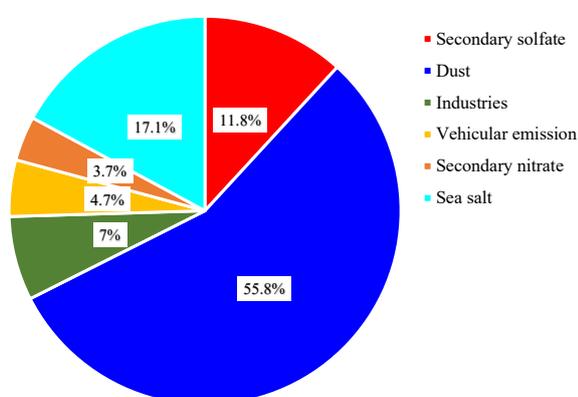


Figure 5. The contribution of each source to the ambient  $\text{PM}_{2.5}$  (%) during the sampling period

water-soluble ions. The high concentrations of these compounds are associated with the proximity of the study area to the Persian Gulf. The contribution of sea salt particles may be a source of high  $\text{Cl}^-$  concentration in many marine environments including the Persian Gulf (46). In agreement with the results of the present study, high concentrations of  $\text{Na}^+$  and  $\text{Cl}^-$  were reported by Gholampour et al (47), and Ho et al (48), who reported the high concentrations of these ions can be due to strong and permanent winds that blow on the beach and can create water droplets and aerosols.

Considering  $\text{Na}^+$  as a tracer of sea salt, excess sulfate (non-sea-salt sulfates [ $\text{nss-SO}_4^{2-}$ ]), excess potassium (non-sea-salt potassium [ $\text{nss-K}^+$ ]), and excess calcium (non-sea-salt calcium [ $\text{nss-Ca}^{2+}$ ]) can be estimated using the following equations (49):

$$\text{nss-SO}_4^{2-} = [\text{SO}_4^{2-}] - [\text{Na}^+] \times 0.2516 \quad (2)$$

$$\text{nss-K}^+ = [\text{K}^+] - [\text{Na}^+] \times 0.037 \quad (3)$$

$$\text{nss-Ca}^{2+} = [\text{Ca}^{2+}] - [\text{Na}^+] \times 0.0385 \quad (4)$$

The non-sea-salt  $\text{SO}_4^{2-}$ ,  $\text{K}^+$ , and  $\text{Ca}^{2+}$  accounted for nearly 80%, 94%, and 92% of the total sulfate, potassium, and calcium of  $\text{PM}_{2.5}$  that may originate from other sources such as dust, industries, and vehicular emissions.

In agreement with the results of the present study, Yan et al, reported that  $\text{nss-SO}_4^{2-}$  in the East China Sea ranged from 1.46–24.9  $\mu\text{g}/\text{m}^3$ , which accounted for 90.4% of total sulfate (50). In another study in the northern South China Sea, Hsu et al, also reported that  $\text{nss-SO}_4^{2-}$  in  $\text{PM}_{2.5}$  accounted for 95.9% of total sulfate, indicating anthropogenic sources of sulfate (51). But in a study around Urmia Lake, Iran, Gholampour et al (47) reported that the amounts of excess sulfate and excess potassium were approximately 65%–75% and 60%–75%, respectively, which are lower than the evaluated values in the present study. As Bushehr port is located in a hot and humid area and has numerous industries, higher concentrations of  $\text{SO}_4^{2-}$  could be because of the increased photochemical oxidation of  $\text{SO}_2$  during warm periods, which is consistent with the results of former studies in Thessaloniki and Budapest (52,53). Also, the particulate forms of sulfate and nitrate may be produced due to the rapid oxidation of  $\text{SO}_2$  and  $\text{NO}_x$  produced from various sources, especially industries (54,55).

The mass ratio of nitrate/sulfate is a good indicator of the relative importance of stationary and mobile sources of nitrogen as well as sulfur in the atmosphere (56). In this ratio, nitrate and sulfate are used as indicators of mobile and stationary emission, respectively. In the present study, the mass ratio of nitrate to sulfate was 0.57, indicating that stationary emission was the predominant source in the study area. It was expected that stationary emission was the dominant source because there is not much traffic in Bushehr port. Also, the mass ratio of nitrate to sulfate

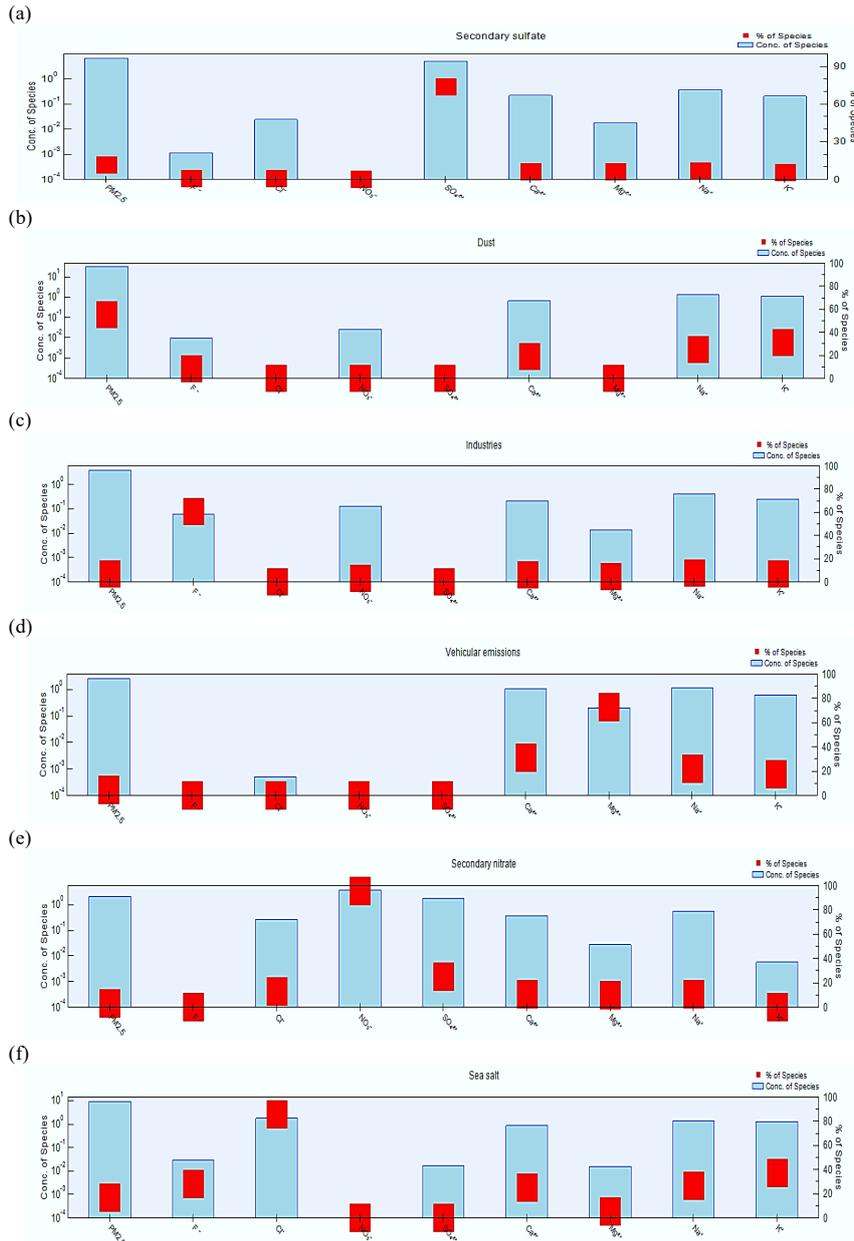


Figure 6. Source profiles of PM<sub>2.5</sub> and other water-soluble ions in the northern part of the Persian Gulf

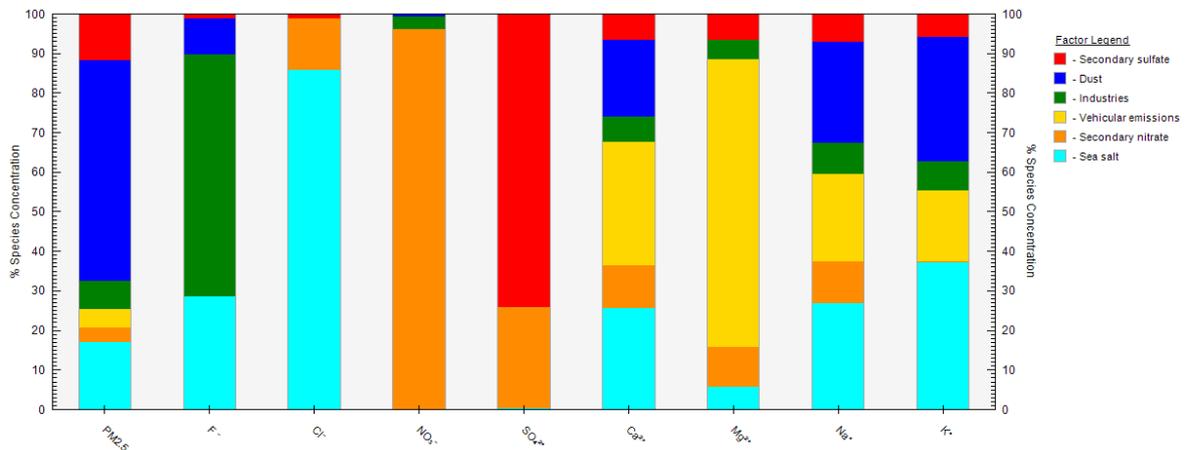


Figure 7. Percentage of PM<sub>2.5</sub> and species apportioned to each source

**Table 3.** Comparative evaluation of water-soluble ions in the present study and former studies ( $\mu\text{g}/\text{m}^3$ )

Country	Site	Year	Ca <sup>2+</sup>	Mg <sup>2+</sup>	Na <sup>+</sup>	K <sup>+</sup>	F <sup>-</sup>	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	Reference
Iran	Retirement home	2012-2013	0.97	0.10	0.60	0.06	-	0.47	4.00	3.40	(29)
	School dormitory		1.10	0.50	0.40	0.08	-	0.70	5.70	4.50	
China	Hefei	2012-2013	5.24	0.30	0.48	0.96	-	1.21	15.14	15.56	(38)
Vietnam	Hanoi	1999-2001	0.33	0.05	0.14	0.61	-	0.03	0.33	6.47	(39)
India	Delhi	2013-2014	2.83	0.96	5.05	4.10	0.91	7.77	10.0	12.9	(40)
Japan	Yokohama	2007-2008	0.20	0.05	0.25	0.13	-	0.21	0.96	3.8	(41)
Greece	Athens	2010-2010	0.34	0.07	0.28	0.10	-	0.23	0.49	3.99	(42)
China	Beijing	2009-2010	1.50	0.20	0.50	1.70	-	2.90	20.50	19.10	(43)
Qatar	Doha	2015	7.42	0.31	1.26	0.23	-	1.4	30.5	15.24	(44)
Iran	Sistan	2014	10.10	0.80	3.14	6.95	-	2.31	1.57	4.06	(45)
Iran	Bushehr	2016-2017	3.68	0.28	5.38	3.55	0.1	2.06	3.88	6.76	This study

in winter, spring, and summer were 0.66, 0.77, and 0.24, respectively. According to the mass ratio of nitrate to sulfate in each season, the stationary emission was the highest in summer followed by winter and spring.

Temporal trends of water-soluble ions are consistent with the reports of previous studies on the ions content of PM<sub>2.5</sub> during dust storms (35,36,46). Also, seasonal variation of total water-soluble ions levels (Figure 4) showed that the highest concentrations of ions were measured in winter (with a mean of 3.64  $\mu\text{g}/\text{m}^3$ ) followed by spring (with a mean of 3.29  $\mu\text{g}/\text{m}^3$ ), and summer (with a mean of 2.69  $\mu\text{g}/\text{m}^3$ ), respectively. These results are consistent with the results of previous studies (56-59). According to Figure 4, the concentrations of Ca<sup>2+</sup>, Mg<sup>2+</sup>, Na<sup>+</sup>, K<sup>+</sup>, F<sup>-</sup>, and SO<sub>4</sub><sup>2-</sup> were slightly different throughout all seasons. Besides the eight species, the concentrations of Cl<sup>-</sup> and NO<sub>3</sub><sup>-</sup> were lower than those in summer. The seasonal variation of PM<sub>2.5</sub> and the consequence of water-soluble ions can be due to atmospheric conditions such as temperature, wind speed, and humidity (58). The concentrations of water-soluble ions are correlated with dust concentration (37,60). Also, the air mass trajectories during the sampling period in Bushehr port can affect the content of PM<sub>2.5</sub>. Based on another study in Bushehr port (34) at the same sampling periods and places of the present study, the air mass from the northwest and southeast had the longest trajectory lines, indicating the importance of the west and southwest regions including Iraq, Saudi Arabia, and Khark Island, with high oil/gas-linked activities. Also, the trajectory analysis indicated that water-soluble ions of PM<sub>2.5</sub> in Bushehr port were originated from distal west regions during the sampling days, especially on dusty days.

The relationships between PM<sub>2.5</sub> and ionic species are illustrated in Table 2. Usually, an R<sup>2</sup> value of almost 0.5 is considered a good correlation between the cations and anions, showing the probability of the formation of that specific cation and anion (61). The results of a study in Durg City, India, were consistent with the results of this

study and showed a positive correlation between PM<sub>2.5</sub> and NO<sub>3</sub><sup>-</sup> (62). The good correlation between PM<sub>2.5</sub> and NO<sub>3</sub><sup>-</sup> may be due to the occurrence of the component of NaNO<sub>3</sub> in PM<sub>2.5</sub> (63). In a study by Shahsavani et al, a good correlation was reported between Na<sup>+</sup> and Cl<sup>-</sup> (0.56), NH<sub>4</sub><sup>+</sup> and SO<sub>4</sub><sup>2-</sup> (0.49), K<sup>+</sup> and Cl<sup>-</sup> (0.58), K<sup>+</sup> and SO<sub>4</sub><sup>2-</sup> (0.56), Ca<sup>2+</sup> and Cl<sup>-</sup> (0.64), Ca<sup>2+</sup> and NO<sub>3</sub><sup>-</sup> (0.56), and Ca<sup>2+</sup> with SO<sub>4</sub><sup>2-</sup> (0.50). They concluded that NaCl, (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, KCl, K<sub>2</sub>SO<sub>4</sub>, CaCl<sub>2</sub>, Ca(NO<sub>3</sub>)<sub>2</sub>, and CaSO<sub>4</sub> may exist in the total suspended particles (35).

#### Source apportionment by PMF model

The sources of PM<sub>2.5</sub> for the six factors including secondary sulfate, dust, industries, vehicular emission, secondary nitrate, and sea salt were identified in Figure 5. Also, Figures 6 and 7 present the factors profile and the percentage of PM<sub>2.5</sub> and species apportion to each source, respectively.

The first factor, weighted by SO<sub>4</sub><sup>2-</sup>, is associated with secondary sulfate origin (Figures 6a and 7). Secondary sulfate is typically related to long-range transportation events. Due to the slow oxidation of SO<sub>2</sub> to SO<sub>4</sub><sup>2-</sup>, this aerosol component is more associated with transportation than local pollution (64), as found in background areas on the Mediterranean coast (65).

The profile of factor 2 is mostly determined by Ca<sup>2+</sup>, Na<sup>+</sup>, and K<sup>+</sup> (Figures 6b and 7), which are associated with dust storms (66).

Factor 3 was heavily weighted by F<sup>-</sup> (Figures 6c and 7), which is mostly used as an indicator for industrial sources (67,68).

Factor 4 shows the vehicular emissions source because of the high portions of Ca<sup>2+</sup>, Mg<sup>2+</sup>, Na<sup>+</sup>, and K<sup>+</sup> (Figures 6d and 7). This factor relates to vehicular emissions sources due to diesel and gasoline-powered vehicles exhaust emissions (69,70).

Factor 5 indicates high burdens for NO<sub>3</sub><sup>-</sup> (Figures 6e and 7), which can be known as a secondary nitrate source. As a result, their gaseous precursors species such as NO<sub>x</sub>

emitted from vehicles, fossil, and industry processes can cause the formation of secondary nitrate sources (69).

The sea salt source (factor 6) is heavily weighted by the attendance of Cl<sup>-</sup> (Figures 6f and 7), indicating the sea spray (71). The presence of sea salt is fully usual in the contaminated coastal and marine atmosphere (72,73), which was also detected in the southeastern part of China in PM<sub>2.5</sub> (74).

### Conclusion

The present study was performed in the northern part of the Persian Gulf (in Bushehr port) for 9 months from Dec 2016 to Sep 2017 to determine the water-soluble ions contents of PM<sub>2.5</sub> including Ca<sup>2+</sup>, Mg<sup>2+</sup>, Na<sup>+</sup>, K<sup>+</sup>, F<sup>-</sup>, Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, and SO<sub>4</sub><sup>2-</sup>. The mean mass concentration of PM<sub>2.5</sub> during the sampling period was 65.77 µg/m<sup>3</sup>. Only 4% and 17% of the samples were lower than the daily mean of the WHO and EPA outdoor air guidelines, respectively. Also, the mean concentration of PM<sub>2.5</sub> on dusty days was nearly 3 times higher than that on other days, indicating that dust storm can be a source of increased PM<sub>2.5</sub> in outdoor air in this area. These water-soluble ions accounted for almost 90% and 35% of the total mass of water-soluble ions and mass of PM<sub>2.5</sub>, respectively. The water-soluble ions including Ca<sup>2+</sup>, Na<sup>+</sup>, K<sup>+</sup>, Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, and NO<sub>3</sub><sup>-</sup> in PM<sub>2.5</sub> had a concentration peak in dusty days during the sampling period. Also, source apportionment results showed that dust, sea salts, secondary sulfate, industries, vehicular emission, and secondary nitrate were the major contributors to PM<sub>2.5</sub> mass. Therefore, due to the geographical location of Bushehr province, the occurrence of dust storms and the presence of numerous industries in this area is highly needed to control various sources of PM<sub>2.5</sub> emissions and choose suitable strategies to reduce the health effects of PM<sub>2.5</sub>.

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### Competing interests

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.

### Ethical issues

The authors certify that all data collected during the study are as stated in the manuscript, and no data from the study has been or will be published separately elsewhere. Ethical approval was obtained from Bushehr University of Medical Sciences (IR.BPUMS.REC.1395.72).

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