

Characteristics of metals in the aerosols of Zhongshan Station, Antarctica

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Abstract Instrumental neutron activation analysis (INAA) was applied to analyze the bulk, high-volume aerosol samples, collected at Zhongshan Station in the Eastern Antarctica, during 1998-2001, to study the chemical species. A graphical technique was applied to the INAA data. Results showed that Na, Cl, Mg, Ca, Sr, Br, I, Sr and Rb were marine elements while Al, Sc, Fe and Mn were crustal elements. Compared to marine and crustal elements, five elements (Se, Co, Sb, Zn, Cr) were highly abundant in the aerosols collected at Zhongshan station, which indicated that they might come from the petroleum burning, heating and equipment operation. The presence of pollutant elements suggested that human activities have affected the local environments in Antarctica.

Key words aerosol, Antarctica, anthropogenic activities, heavy metals.

1 Introduction

Since the industrial revolution, the human activities have caused serious changes of environment and have affected the global ecosystem. Environment becomes one of the main issues to keep sustainable development. Due to the remote geographical location and harsh climatic condition, the Antarctic is always regarded as the region with minimum influence from human activities^[1-3]. However, many studies showed that human activities in the Antarctic and other areas all over the world have triggered the local environmental changes in the Antarctic region^[2, 4, 5].

Due to the increasing of scientific explorations, as well as rapid development of all kinds of non-governmental activities and tourisms, in the Antarctic region, human activities have caused impacts on the Antarctic terrestrial ecosystem at various levels^[6]. The use of machine tools has caused physical destruction of surface soil and underlying ever frozen layer. In summer, the previously well-drained area becomes swamp, the frozen layer thaws and the soil shrinks and collapses. Most of the damages are permanent^[4, 7]. Furthermore, the large-scale use of petroleum fuel has polluted the soil with heavy metal and hydrocarbon nearby some Antarctic research stations^[8-11]. More and more evidences showed that natural environment of Antarctica,

the last piece of pure land, was seriously affected by human activities.

Since 1960, the Scientific Committee on Antarctic Research (SCAR) meeting has determined to prevent the organisms and environment in the Antarctic. From then on, the scientific societies and governments have established and implemented a series of environmental protection regulations. Several Antarctic key protection areas such as Special Protected Areas (SPAs) and Sites of Special Scientific Interest (SSSIs) were designated. Furthermore, when the Protocol on Environmental Protection to the Antarctic Treaty and the five attachments inured on January 14, 1998, the corresponding "Committee for Environmental Protection (CEP)" was established as the advisory committee for environment-related issues of Antarctic Treaty Consultative Meeting (ATCM). The CEP is directly responsible to ACTM and requires each Antarctic Treaty Consultative Party to submit "National Antarctic Environmental Report" to the committee every year.

The report of atmospheric environment in Antarctic is one of the important contents of National Environmental Report, reflecting the human activity at scientific station area and the environmental quality nearby. In present study, the elemental compositions of the aerosols collected at Zhongshan Station during 1998-2001 were analyzed by instrumental neutron activation analysis (INAA), and tried to evaluate their sources and environmental impacts.

2 Sampling and methods

2.1 Climate and Environment around Zhongshan Station

Zhongshan Station locates at $69^{\circ}22'24''\text{S}$ $76^{\circ}22'24''\text{E}$, at the north tip of Mirror Peninsula, Larsemann Hills in East Antarctica. Larsemann Hills ($69^{\circ}12'\text{S}\sim 69^{\circ}28'\text{S}$ and $76^{\circ}\text{E}\sim 76^{\circ}30'\text{E}$), which is one of the few ice-free coastal regions in the Antarctic continent in summer. This region, with a total area of about 200 km^2 (terrestrial area 40 km^2), is composed of four large peninsulas (the Stornes Peninsula, the Broknes Peninsula, the Mirror Peninsula and the Little Peninsula) and more than 130 islands. . . The highest point in this area is 180 m above sea-level^[16].

The weather of this region is very complex due to the combined influences of circumpolar cyclone and Antarctic continental high pressure. With 58 polar nights and 52 polar days each year, this region belongs to high-latitude and high-cold permafrost zone. During the period from 1989 to 2000, the annual average temperature is approximately $-10\text{ }^{\circ}\text{C}$, ranged from $-40.4\text{ }^{\circ}\text{C}$ to $9.6\text{ }^{\circ}\text{C}$ ^[17]. The surface soil freezes for more than 300 days each year. From the end of December to the middle February of the next year, the ice melts. The weather is dry and the average relative humidity is 60% during the period from 1989 to 2000. The average number of rainy days is 145. The easterly wind prevails during the year and wind speed in summer is clearly lower than in other seasons. During 1989~2000, the average wind speed is 7.2 m/s , with the maximum wind speed of 50.3 m/s . The number of strong wind days is 171 (wind speed $>17.0\text{ m/s}$)^[17].

There are 15 buildings (totally, 2700 m^2) in Zhongshan Station and 19 transpor-

tation vehicles. It is home for about 60 and 25 scientists during austral summer and winter respectively. Zhongshan Station consumes 200 tons of diesel fuel each year for power generation, heating, water production, and equipment operation.

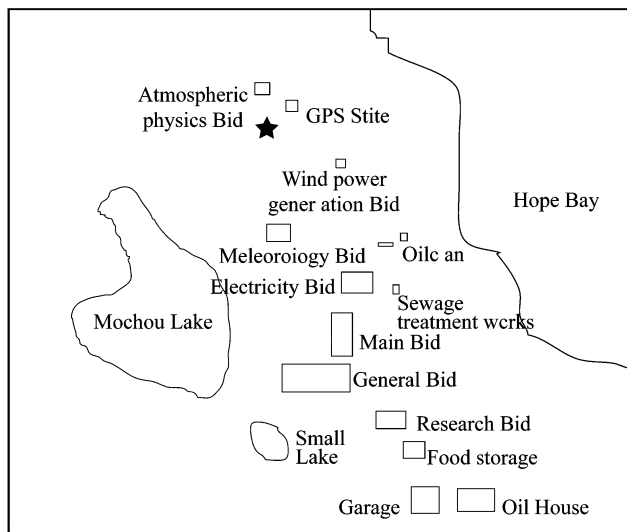


Fig. 1 Map of Zhongshan Station (★ Sampling site).

2.2 Sample Collection and Analysis

The aerosol samples were collected by using a high-volume sampler M400 (USA) deployed on the mountains near the Physics Observatory in Antarctic Zhongshan Station. Bulk samples were collected for 5 or 7 days on 20.3 × 25.4 cm Whatman41 filters (Whatman International Ltd, England). The mean volume of air sampled for each filter was 6000 m³. All samples were stored at 4 °C before subjected to indoor analysis.

Instrumental neutron activation analysis was carried on in China Institute of Atomic Energy. The filters were sealed into sanitary polyethylene bags (for the short irradiation), or packed in precleaned foils (for the long irradiation). Chinese national standard GBW07312 (water sediments) and American NIST SRM 1632a (coal powder) were used as quality control. The amount of each sample was about 100 mg.

Sample irradiation was conducted in the random channels of heavy water reflector of the Heavy Water Research Reactor (HWRR), Chinese Institute of Atomic Energy. Irradiation lasted for 6 hours. Thermal neutron fluency rate was about $3\text{--}5 \times 10^{13} \text{ ncm}^{-2}\text{s}^{-1}$. Activated samples were transferred to the sanitary (without radioactivity) standard measuring boxes.

Seventy-two aerosol samples collected at Zhongshan Station during 1998–2000 were selected for instrumental neutron activation analysis. The concentrations of 27 elements in samples were determined, including Al, Ba, Br, Ca, Ce, Cl, Co, Cr, Cs, Eu, Fe, Hf, I, La, Mg, Mn, Na, Nd, Rb, Sb, Sc, Se, Sr, Ta, Th, V and Zn. Except marine and crustal elements, the concentrations of other elements were relatively low. After experimental error and standard deviation analysis, Ba, Co, Cs, Eu, Hf, La, Nd, Rb, Se, Ta, Th were excluded and 15 elements, Na, Al, Br, Ca, Ce, Cl, Fe, I, Mg, Mn, Rb, Sb, Sc, Sr and Zn were reserved for further analysis.

Table 1. Aerosol samples information from Zhonshan Station

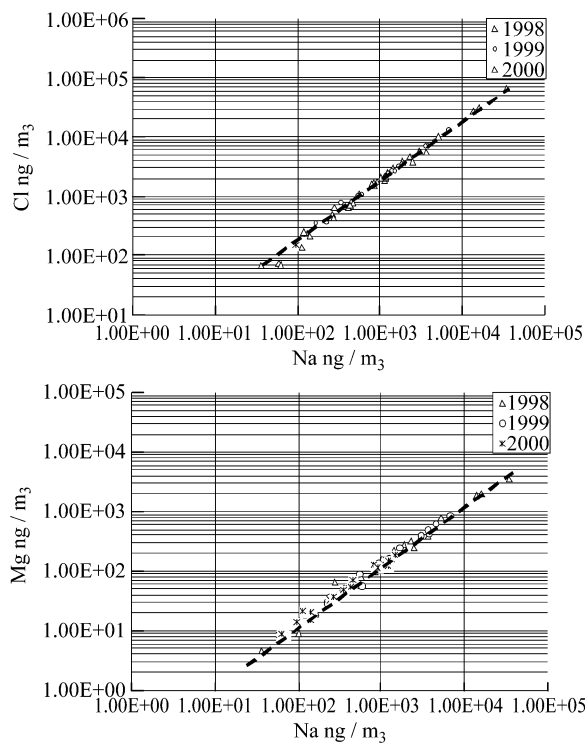
Sampling Time	1998	1999	2000
Number of samples	25	22	25

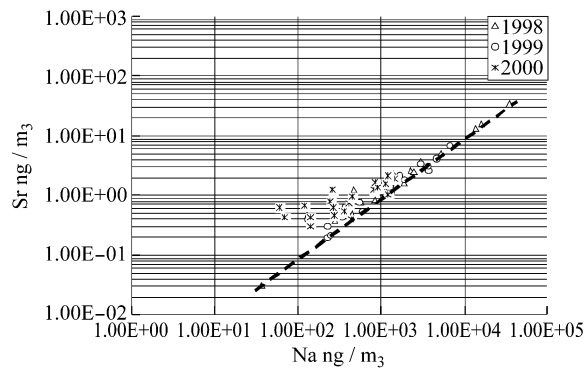
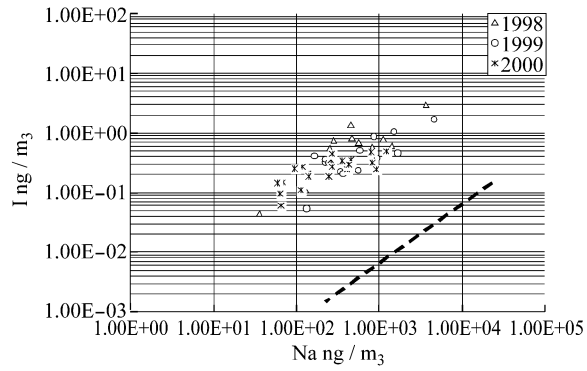
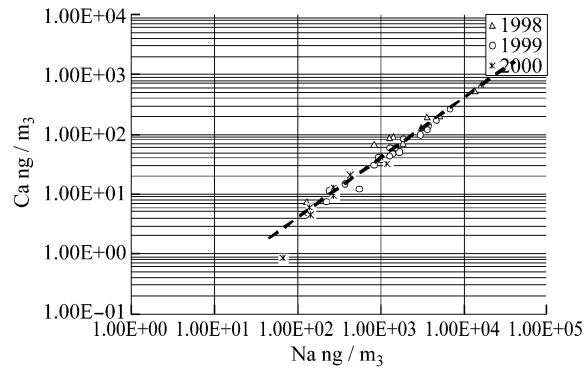
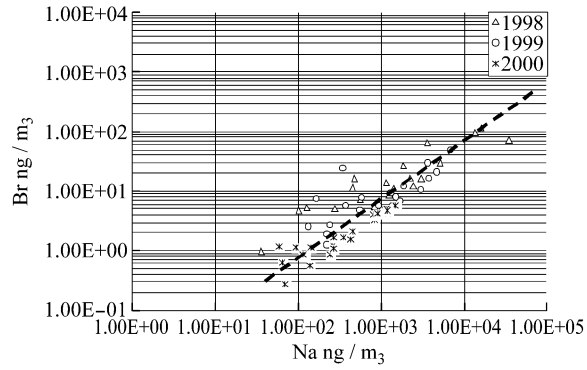
3 Results and discussions

3.1 Application of Graphical Technique for Analysis of Element Composition of Aerosols from Zhongshan Station

Marine Elements Sea salts were the main components of aerosols, as expected, since Zhongshan Station lies along Antarctic coasts. Na and Cl are the main elements. Concentrations of elements Cl, Mg, Ca, Sr, Br, I, Sr and Rb exhibited quite good correlation to that of Na (Fig. 2), indicating they are marine elements. The dotted line in the figure 2 is the ratio of element X to Na in the seawater. The ratios of Cl, Mg, Ca, Sr, Br, Sr, and Rb to Na in the aerosols are nearly identical with those in the seawater. Ratios of I to Na in aerosols are higher than those in the seawater, mainly due to the high volatility of I.

Crustal elements In aerosols over Zhongshan Station, Sc, Fe and Mn correlated well with Al (Fig. 3), an indicator for crustal element. The dotted line in the figure shows the ratios of element X to Al in the soil.





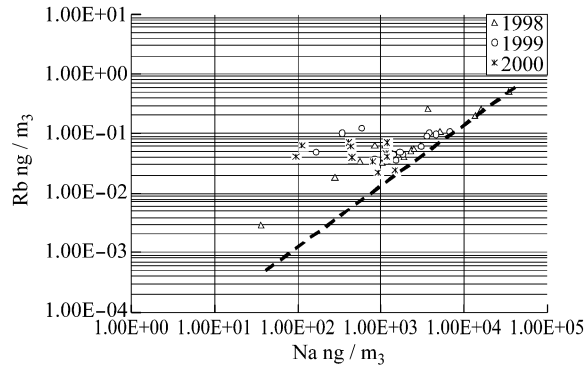


Fig. 2 Marine elements of aerosol in Zhongshan Station.

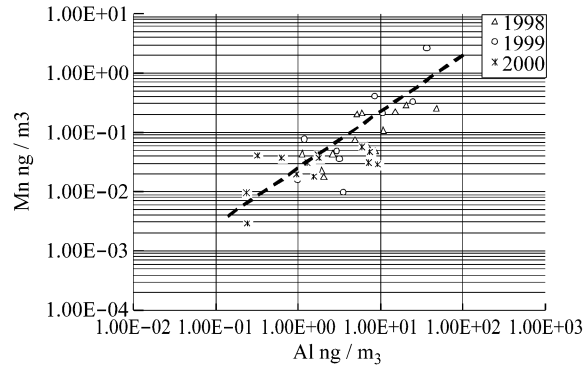


Fig. 3 Crustal elements of aerosol in Zhongshan Station.

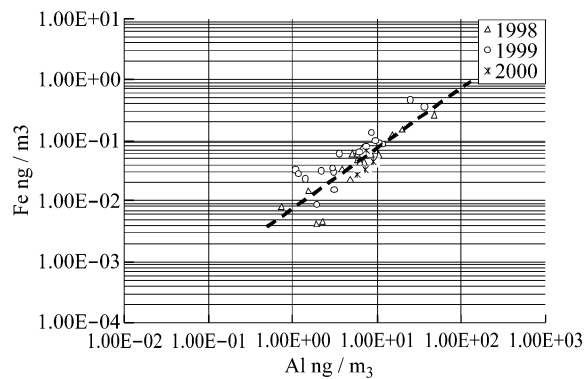
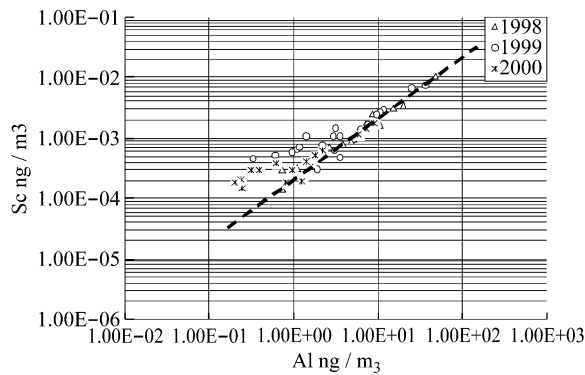


Fig. 3 Crustal elements of aerosol in Zhongshan Station.

Pollutant elements Relative to the marine elements and soil elements, five elements (Se, Co, Sb, Zn and Cr) showed no correlation with Na or Al as seawater and crustal source indicators, respectively. However, their enrichment factors (EF), to marine element Na or crustal element Al, are abnormally high. The highest EF reaches the value more than 1000. In this study, the element analysis were based on graphical technique for analysis of element origin, proposed by Rahn^[18], with Na/Al as abscissa and X/Al as ordinate (X stands for the element to be analyzed).

$$\text{Since } X_{\text{crust}} = \text{Al} \left(\frac{X}{\text{Al}} \right)_{\text{crust}} ; X_{\text{marine}} = \text{Na} \left(\frac{X}{\text{Na}} \right)_{\text{marine}}$$

In Fig. 4 the dotted line 1 shows the ratio X/Na in the seawater and the dotted line 2 shows the ratio X/Al in the soil. Hence, if the analyzed element is a marine element, the dots in the figure shall scatter near the dotted line 1 and if the analyzed element X is a crustal element, the points in the figure shall distribute near the dotted line 2. For example, the ratio of Ca to Na in the seawater is around 0.038 while its ratio to Al in the soil is around 0.504. In Fig. 4, in aerosols from Zhongshan Station, its ratio to Na is mostly nearing with its marine ratio value. So, Ca in the aerosol of Zhongshan Station mainly originates from the seawater.

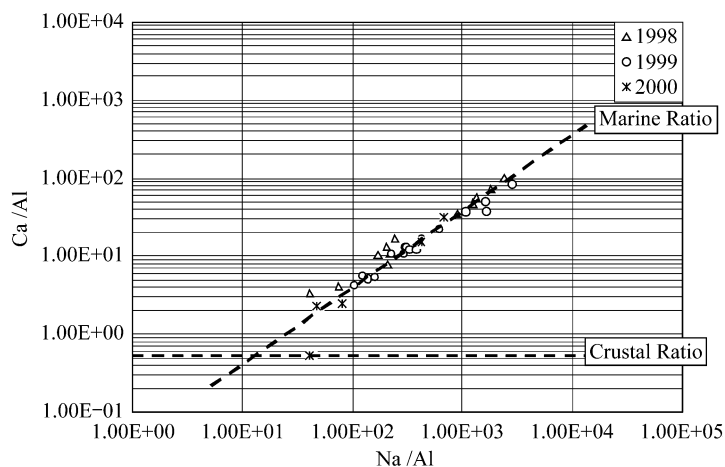
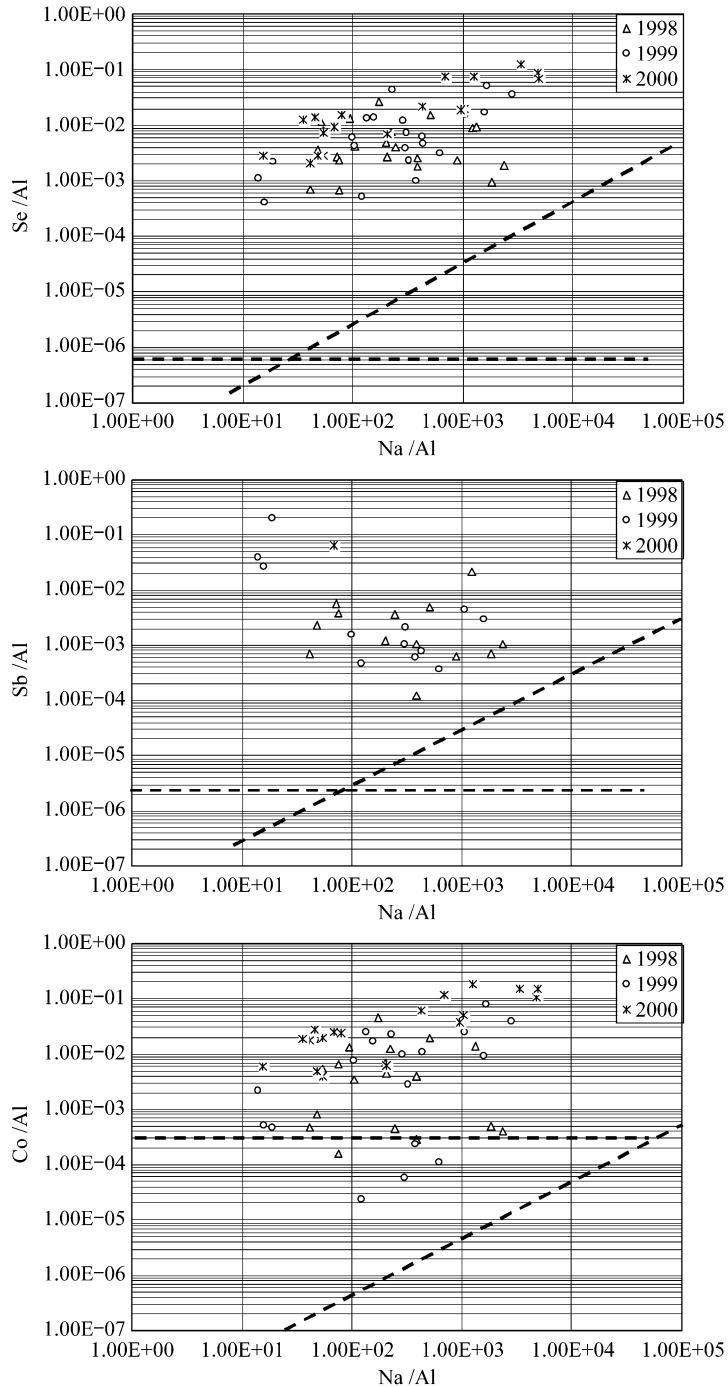


Fig. 4 Application of Graphical techniques for analysis of element Ca.

As for Se, Co, Sb, Zn and Cr, the corresponding dots all locate above lines of marine and crustal ratio in the plots, suggesting that they would be enriched rather than marine Na or than crustal Al (EF can generally be read from the figure). Therefore, they could be classified as pollutant source elements (Fig. 5).

The generally used mathematical statistic methods for aerosol analysis are cluster analysis, chemical mass balance, factor analysis, enrichment factor method, and so on. However, after the complex mathematical treatments, understanding of the results needs sufficient information of the environment and principles behind the analysis. Even so, there are some results still confused. For instance, in the study of aerosol element data obtained from Zhongshan Station in 1998, by Huang *et al.*^[19], factor analysis showed crust factor 1 not only had high load to Fe, Al, Mn and K, but also had certain load to common heavy metal elements such as Cr, V and Zn.

The marine factor 2 contributed to heavy metal elements such as V, Pb and K except its high load to Na, Ca and Mg. But in fact, the enrichment factors of Cr, V, Zn and Pb to marine Na are all greater than 1000, while some even above 10000. The enrichment factors of these elements to crustal Al are all greater than 5, the highest reaching 463. Based on enrichment factors, the four elements Cr, V, Zn and Pb in aerosols in Zhongshan Station in 1998 mainly originate from human's pollution.



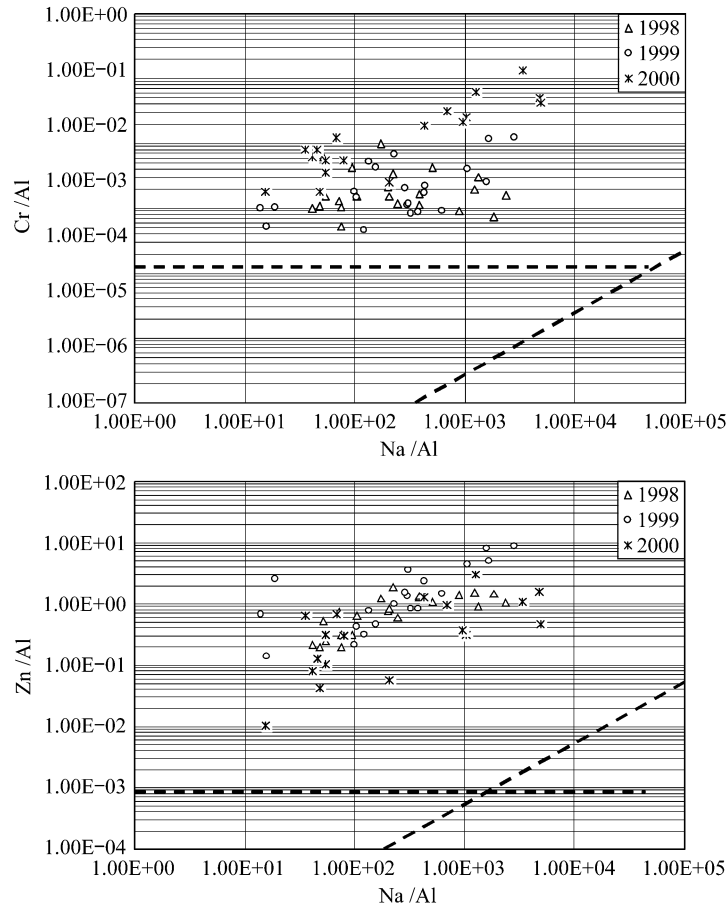


Fig. 5 Pollutant elements of the aerosols in Zhongshan Station.

The similar confusions occurred in aerosol factor analysis in the case of Korean King Sejong Station in Antarctic Peninsula^[20]. Except its contribution in crustal Al and Ba, as well as marine Sr, factor 1 also made contribution to heavy metal elements Co and Cs. Therefore, it is regarded as a mixed factor; factor 3 is taken as a pollutant factor, with high contributions to Bi, Cu, Tl, Zn and Ca, while relatively low contribution to heavy metal Co, Cr and Pb, even negatively correlative to Co.

Combining the correlation and enrichment factor analysis, the graphical technique can concisely and intuitively show the enrichment degree of the analyzed elements, and separate the pollutant elements from marine elements and crustal elements. Therefore, it is an efficient means to analyze the sources of the aerosol elements. By means of the graphical technique, the analysis of samples obtained during three years in Zhongshan Station reveals that heavy metal elements Se, Co, Sb, Zn and Cr are highly enriched in aerosols in the station area, which suggested human's activities have an obvious impact on the atmosphere in the Station.

3.2 Temporal Variation of Heavy Metal Elements

Surrounded by the Southern Ocean, the Antarctic is the most remote continent and the least polluted area by human's activities. In 1981, Cunningham and Zoller^[21] reported the concentrations of 15 chemical elements^[21] in aerosols at the South Pole (90°S). Mazzera *et al.* collected aerosols with a diameter less than 10 μ m (PM₁₀) and study the origin of PM₁₀ and the aerosol sulphate^[22]. Mishra *et al.* analyzed the metal components of aerosols in the Korean Station in Antarctic Peninsula and the variations of the metal and other ions, as well as their origins, by factor analysis^[20]. Limited studies on the aerosol collected in Antarctica have been carried out in China. Li and Cao studied the physicochemical characteristics of aerosols and environmental changes, and summarized the current progress of Antarctic atmospheric aerosols^[23]. With the method of atomic absorption, Huang *et al.* analyzed aerosols collected in 1998-1999 and found that most of heavy metal elements changed subject to the seasons, with a high concentration in spring and summer.^[19] Factor analysis showed that Cu, Pb, Zn, Cd and V originated from human pollution and the crust. However the long-term variation of aerosol elements in the Station still needs further investigation.

The temporal variations of elements in aerosols during 1998-2000 revealed that marine element Na didn't exhibit a strong seasonal variation. Generally it reached a high concentration from May to August in winter of southern hemisphere. Analysis of aerosols in Zhongshan Station in 1998 by Huang *et al.* also shows that high Na concentration occurs in the windy winter^[19]. Most of the studies show that marine elements along Antarctic seashore possess obvious seasonal trends with high concentration in summer, which are mainly related to the retreat of ice in summer and southward advance of coastline. However, in the winter of Zhongshan Station, particularly in windy May to September, the severe wind also increased the concentration of marine aerosol.

Crustal element Al showed certain seasonal variations, with a high concentration from November to February of austral summer, which probably related to ice melting and exposed land surface in summer. The increasing traffic and drifting dust would cause the increase of crustal elements.

Though different chemical analysis methods were applied, the concentration of 5 enriched pollutant elements (Se, Co, Sb, Zn and Cr) of Zhongshan Station were in the same order of magnitude to those of other Antarctic Research Stations, such as American McMurdo Station and Korean King Sejong Station^[19, 20, 24] (Table 2). This indicated that impact of human activities to the atmosphere in Zhongshan Station, was equivalent to other scientific research stations in Antarctica.

Table 2. Concentrations of aerosol heavy metals in Zhongshan Station

	Cr	Co	Zn	Se	Sb
Maxima	0.4972	0.3127	22.0867	0.1143	1.7624
Average	0.1228	0.0304	4.2450	0.0231	0.0575
Minima	0.0081	0.0000	0.0674	0.0026	0.0000

Huang *et al.* hold the opinion that heavy metal elements Cu, Pb, Cd and V in

Zhongshan Station during 1998-1999 reach their highest value in the austral summer^[19]. These elements were not detected or were below detection limit in our samples. Zn and Cr, as well

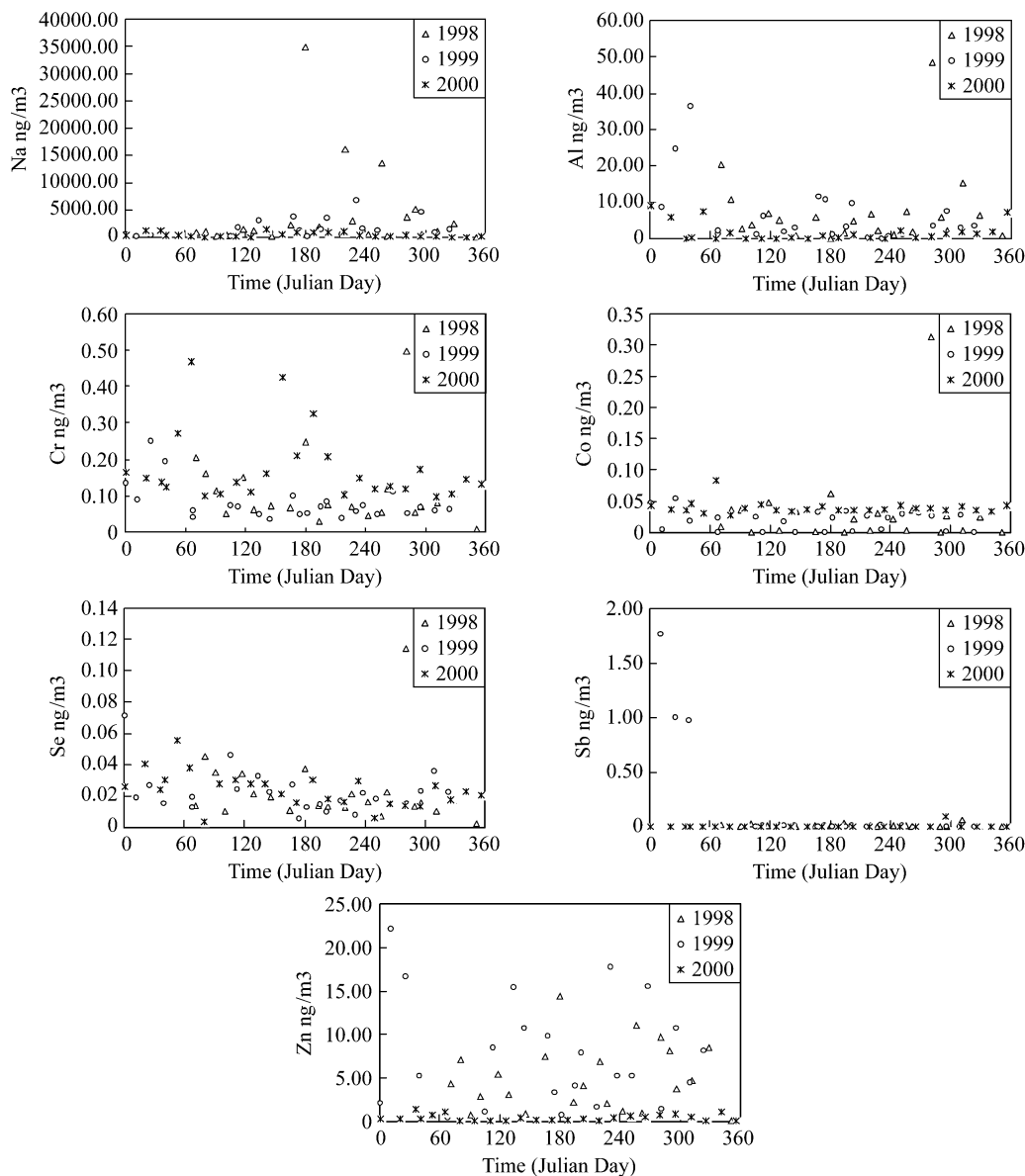


Fig. 6 Temporal variations of 7 elements in Zhongshan Station.

as the other three heavy metal elements Se, Co and Sb, showed no obvious seasonal variations in both studies (Fig. 6). Generally, variations of marine and crustal elements, subject to annual variations of local weather, exhibited seasonal dynamics. However, relatively consistent distribution of heavy metal elements suggested that their origins were not controlled by natural forces, but by the local human activities. Zhongshan Station is home for 25 persons in winter and 60 persons in summer every year. More than 200 tons of polar oil was used for warming, operation of the instruments and waste disposal, and so on. Waste gas released from fuel burning produce harmful heavy metal elements, such as Pb, V and Ni. Meanwhile, discharge of daily

waste water and stack of solid trash also can make pollutant elements like Cr, Pb, Cu and Al enter into soils, water bodies and local organisms.

The five pollutant elements in the aerosols of Zhongshan Station exhibited different temporal variations. This is consistent with previous study of the aerosols in Zhongshan Station in 1998 by Huang *et al.* [19] and the investigation in King Sejong Station by Mishra *et al.* [20]. This suggested different sources of these pollutant elements and should be analyzed further. Mishra *et al.* have also discussed possible import of heavy metals from other continents [20]. It was shown that human activities in other regions released approximately 1 800 kg lead to Antarctica annually, accounting for only 20% of lead content in Antarctic atmosphere [3]. Suttie and Wolff found that the concentration of heavy metals Cd, Cu, Pb and Zn in the surface snow were high near the outlet of a generator, but they decreased to background level at sites 40 m away from the outlet [25]. Since Zhongshan Station locates in East Antarctica and is far from other continents, pollution from other regions is relatively limited. Therefore, it is suggested that the atmospheric heavy metals in Zhongshan Station are mainly from the local human activities.

4 Influence of aerosol heavy metals on the Antarctic ecosystem and significance for environmental protection

The element analysis suggested that, except the marine and crustal aerosols, pollutant aerosol from human activities was a significant part in the aerosols in Zhongshan Station. Human activities in Zhongshan Station, including power generation, warming and instrument operation, have inevitably affected the atmospheric environment around the Station.

It is generally accepted that atmospheric contaminants threaten the terrestrial plants of Antarctic, especially the abundant lichens in the ice-free zone of Antarctic [26]. In Artwoski Station, trace metal contents in the lichens near the incinerating plant of the Station is several times or several dozen times higher than those in the healthy lichens outside the Station [27]. In Casey Station, chlorophyll content in the polluted lichens (*Umbilicaria decussate* and *Usnea spaelata*) was only 1/2 to 1/3 of that in the healthy lichen of the same species in SPAs [28]. In Admiralty Bay, substances arriving soil surface from rain are 2.5 t/km² annually, while those from dusts reach to 12.7 t/km². Therefore, the air pollution is a serious threat to the local environment in Antarctica [29].

As a “natural laboratory of science” and “regulating valve of global climate”, the Antarctic plays an irreplaceable role in the Earth’s ecosystem. The protection of Antarctic environment means far beyond protecting the Antarctic itself, which essentially means protecting the planet we live on [2, 6]. Antarctic is the last natural terrestrial ecosystem in the world. However, Antarctic terrestrial ecosystem is the simplest, as well as the most fragile, ecosystem on the earth. Once destroyed, it will be very difficult or even impossible to recover [8]. Scientists should pay special attention to pollution and negative effects on the Antarctic environment caused by scientific ac-

tivities.

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