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RESEARCH ARTICLE

Levels and distribution of heavy metals in atmospheric particular matters over the northern South China Sea

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Abstract Oceans play a significant role in the cycling of trace metals and persistent organic pollutants. In this study, aerosol samples covering the whole northern South China Sea (SCS) were collected in 2005 and 2007, respectively, for analysis of trace metals and major elements. The levels of trace metals detected ranged from 0.514 to 119 ng/m^3 in 2005 and from 0.130 to 24.2 ng/m^3 in 2007, respectively. Cu, Zn, and Pb were the three predominant metals with high enrichment factors (>10), indicating the strong anthropogenic inputs. The trace metals over SCS were comparable to the values in suburban and background sites of South China, but generally higher than those over other seas and oceans. Considering the fact that they were influenced by their proximity to source regions and air mass origins, the elevated metals in 2005 were probably attributed to the strong wind and long-range atmospheric transport driven by Asian monsoon.

Keywords South China Sea · Heavy metals · Asian monsoon · Atmospheric transport

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Introduction

Atmospheric deposition can act as an important source of trace metals and persistent organic pollutants to oceans due to the fact that atmospheric particulates are closely associated with the consequences of urban and other air pollution sources (Beldowska et al. 2012; Castillo et al. 2013; Wang et al. 2013). Generally, trace metals, such as Cu, Pb, and Zn, often exist in particulate phase in air. According to the International Agency for Research on Cancer (IARC), many trace metals are harmful to human, and some even have carcinogenic effect. Studies revealed that the current level of atmospheric particulate may have serious effects on health, especially in some industrial and urban areas (Hieu and Lee 2010; Karar and Gupta 2006). Consequently, atmospheric metal exposure has caused a wide concern in the last few decades.

A number of studies have found atmospheric transport to be the main transport way of trace metals from continent to ocean, leading to their global distribution via long-range atmospheric transport, even in remote environments such as the Arctic and Antarctica (Chen and Siefert 2004; Magi et al. 2005; Witt et al. 2010). Limited data are available at present regarding the atmospheric distribution of trace metals over ocean areas. Generally, the atmospheric distribution of the metals originating from crustal sources exhibits higher concentrations over the oceans close to, predominantly the Northern Hemisphere, deserts (Prospero 1999; Witt et al. 2010). Other metals (i.e., Pb, Zn, and Cd), originating mainly from anthropogenic sources, have been observed to be more evenly distributed over the northern and southern Atlantic (Witt et al. 2006). The East and North Asia, two densely populated areas in the world, have significant implications for global environmental quality. It is suggested that the largest anthropogenic emission of atmospheric trace metals occur in Asia (Onishi et al. 2012). Previous studies have also demonstrated that aerosol can be transported from Asian region to

other countries and remote ocean areas. A number of studies have been undertaken focusing on the characteristics of aerosols in megacities of this area, including Beijing, Guangzhou, Hong Kong, Tokyo, Taipei, etc. (Duan et al. 2003; Lee et al. 2007; Takahashi et al. 2008). Obviously, coastal atmospheric environments adjacent to large urban regions can be strongly affected by pollution emissions from nearby cities. High trace elements in aerosol particles can result in enhanced air-to-sea deposition fluxes of these elements to coastal waters. Iron (Fe) derived from the aerosols is a key factor influencing the primary productivity in the surface layer of seawater (Martin et al. 1994). Therefore, they will greatly affect the coastal ecosystem and accelerate coastal primary production.

The South China Sea (SCS) is a marginal sea in Southeast Asia, surrounded by fast-developing countries, including China, Vietnam, Thailand, Indonesia, and Philippines. The northern SCS is adjacent to the Pearl River Delta (PRD) region, one of the most developed areas in China with several megacities, such as Hong Kong and Guangzhou. The rapid industrialization and urbanization of the PRD have produced an elevated amount of contaminants and thus affected regional environmental quality. Such environmental settings make the SCS extremely sensitive to the changes of surrounding land ecosystems. However, very few data of heavy metals over SCS are available at present. Therefore, this study was conducted with main objectives being to investigate the occurrence and distribution of typical heavy metals in aerosol samples over SCS and to identify potential sources of those compounds.

Materials and methodology

Sample collection

Total suspended particulates (TSPs) were sampled using a modified Anderson-type Hi-Volume air sampler during two summer cruises, 6–22 September 2005 and 10–28 August 2007, respectively. A total of 65 8-h day/night air samples (32 in 2005 and 33 in 2007) were collected during the Shiyan III cruise which circumnavigated the northern SCS in a zigzag transaction line (Fig. 1). Meteorological data, including air temperature, wind speed and direction, atmospheric pressure, and humidity, were attained from an online detection system. The details of sampling and some meteorological information were shown in the supporting information (Tables S1 and S2, Figs. S1 and S2). A high-volume air sampler was placed windward on the foredeck of the ship. TSP samples were collected on a preheated (12 h at 450 °C) quartz microfiber filter (20.3×25.4 cm, Whatman) by drawing air of approximately 140 m^3 at a flow rate of 17.5 m^3/h. After sampling, loaded filters were wrapped with prebaked aluminum foils and sealed with double layers of polyethylene bags.

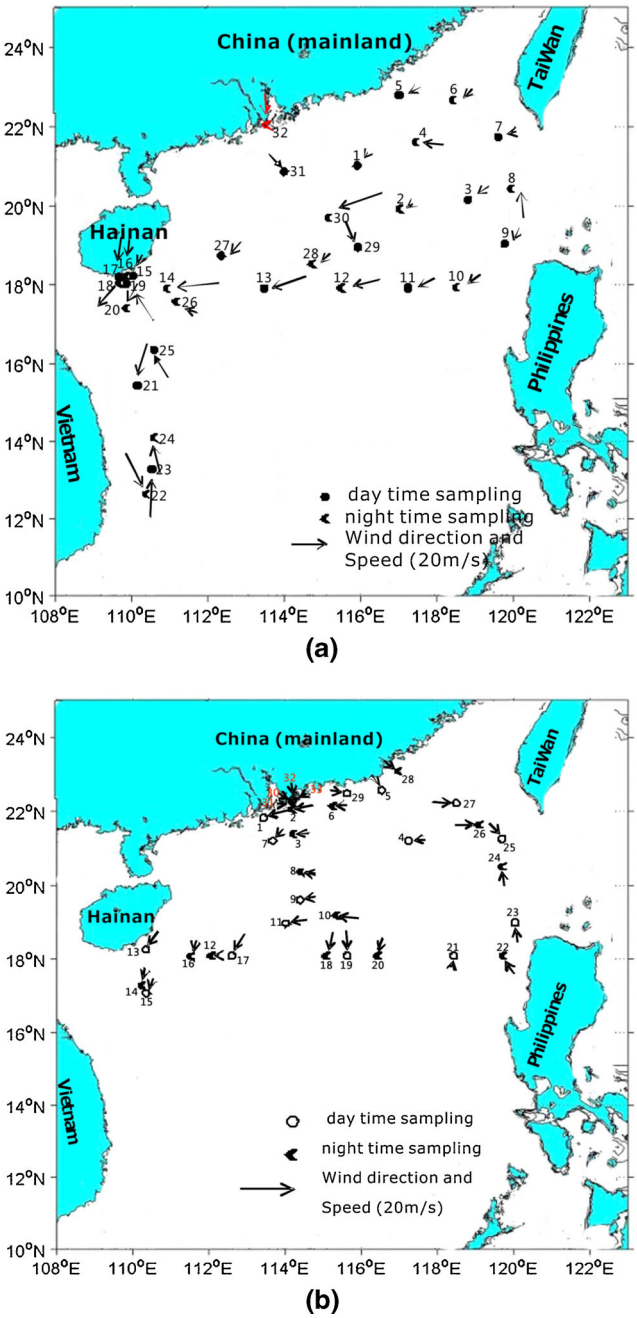

Fig. 1 Map of aerosol sampling locations over the SCS in 2005 (**a**) and 2007 (**b**). Wind direction is a vector average of 8-h sampling; the *arrow length* represents speed of 20 m/s

Analysis of heavy metals

One quarter of the filter was cut using stainless steel scissors and digested with concentrated acids using acid-cleaned Pyrex test tubes (Wong et al. 2003). Briefly, 14 ml of concentrated high-purity HNO_3 and 3.5 ml of concentrated $HClO_4$ were added into tubes to make the filters fully submerged. Sample blanks, standard reference material (NIST SRM 1648, urban particulate matter), and sample replicates were randomly inserted in the sample analysis batch. The mixtures were

gently shaken using a vortex and then heated progressively to 190 °C in an aluminum heating block for 24 h until they are completely dried. Thereafter, the tubes were heated at 70 °C for 1 h after adding 10 ml of 5 % (*v*/*v*) high-purity HNO_3. The supernatant was decanted into an acid-cleaned polyethylene tube after cooling to room temperature.

Metals were determined using inductively coupled plasma atomic emission spectrometry (ICP-AES; Perkin Elmer Optima 3300 DV). Blanks, quality control standards, and standard reference materials were inserted during the analytical measurement. The quality control tests showed that the blanks were <1 % of the mean concentration for all metals. The recovery rates for the standard reference material ranged from 80 to 97 %, and the precision (RSD) was generally lower than 5 %. The recovery rates for Al were around 55 % due to the presence of aluminosilicate minerals.

Back trajectories

Air mass origins were calculated for the cruise samples by the HYSPLIT transport and dispersion model from the NOAA Air Resources Laboratory (http://www.arl.noaa.gov/). Back trajectories were traced for 5 days with 3-h intervals from 00:00 coordinated universal time (UTC) to 09:00 UTC during day sampling and from 12:00 to 21:00 UTC during night sampling at 100, 500, and 1,000 m above sea level.

Results and discussion

Metal concentrations

Descriptive statistics of major elements (Fe, Al, Mg, and Mn) and trace metals (Cd, Cr, Cu, Pb, Ni, and Zn) over SCS are shown in Table 1. Generally, the five major elements over SCS were far lower than those in aerosol from urban and suburban areas in the PRD (Lee et al. 2007), but generally higher than or comparable to those in the remote ocean areas (Witt et al. 2006). Relatively low Fe/Al ratios were found over SCS which might be related to the mixing of Fe-depleted clay dust from ceramic industries in the PRD region.

The trace metals showed a wide range of concentration due probably to the combined effects of climatic conditions and pollution source strength. The concentrations ranged from 0.514 to 119 ng/m^3 in 2005 and from 0.130 to 24.2 ng/m^3 in 2007, respectively. Cu, Zn, and Pb were predominant metals with the maximum concentrations up to 28.7, 27.9, and 119 ng/m^3, respectively. Table 2 shows the comparison of selected trace metals in aerosols from different areas. The metals over SCS were remarkably higher than those in remote marine aerosol samples (remote ocean) while significantly lower than those in urban or suburban areas. These trends probably result from the relatively high atmospheric concentrations in most countries around the SCS caused by tens of millions of tons of electrical and electronic equipment disposal. A number of studies have documented that the aerosol from the high industrialization and urbanization regions has been recognized as the main source of contaminants to marine environments (Favez et al. 2008; Hellebust et al. 2010; Joiris et al. 2001; Kocak et al. 2004).

Table 1 also clearly revealed that trace metals in 2005 were remarkably higher than those in 2007 for both concentration levels and detection frequencies. All metals, except Cd and Cr, were found with 100 % detection rate in 2005. Contrastingly, in 2007, only Cu and Zn were 100 % detected. The average of Pb in 2005 was 6.60 ng/m^3, which is almost twice as that in 2007. Higher concentrations of Cu were also found in 2005 (about three times of mean concentration). Differently, Cr in 2005 is comparable to that in 2007 due to the fact that Cr level was generally low in aerosol samples (Cong et al. 2007; Wu et al. 2009). Further, analytical limitation for Cr element in this

Table 1 Heavy metal concentrations over the SCS in 2005 and 2007

Metals (ng/m^3)	2005 (*n*=32)			2007 (*n*=33)		
	Mean±SD	Concentration range	Detection rate (%)	Mean ±SD	Concentration range	Detection rate (%)
Fe	334±174	136–656	100	176±130	61–522	100
Al	615±189	237–1192	100	395±257	102–889	100
Mg	302±49.0	184–448	100	137±97.4	58.7–583	100
Mn	7.67±2.20	3.15–13.1	100	0.98±1.54	n.d.–7.03	42
Cd	0.242±0.18	n.d.–0.514	47	0.065±0.044	n.d.–0.130	33
Cr	1.71±2.05	n.d.–6.6	47	1.53±1.04	n.d.–3.14	46
Cu	20.6±3.56	9.87–28.7	100	6.94±3.27	1.46–24.2	100
Ni	2.46±1.92	1.06–9.27	100	0.94±0.615	n.d.–1.62	61
Pb	6.60±6.24	1.83–27.9	100	3.70±3.01	n.d.–10.3	49
Zn	50.6±25.8	17.6–119	100	13.3±4.98	6.97–24.1	100

n.d. not detected

Table 2 Examples of the reported values of trace metal concentrations (mean) in remote marine aerosol samples and urban/suburban samples (ng/m^3)

Location	Description	Cr	Zn	Cu	Ni	Pb	References
Northern SCS (in 2005)	Sea	1.71	50.8	20.6	2.46	6.60	Present study
Northern SCS (in 2007)	Sea	1.53	13.3	6.94	0.94	3.70	
Irish Sea	Sea	2.1	36	21	3.0	43	Chester et al. (2000)
Mediterranean	Sea	2.3	22.4	5.9	–	24.9	Kocak et al. (2004)
North Atlantic	Ocean	–		0.60	0.91	0.55	Church et al. (1984)
Indian Ocean	Ocean	–		3.84	0.708	1.08	Witt et al. (2006)
Hong Kong, China	Suburban	12.4	298	30.8	–	53.5	Lee et al. (2007)
Guangzhou, China	Suburban	16.9		65.2	–	219	Lee et al. (2007)
Beijing, China	Urban	19		110	–	430	Okuda et al. (2008)
Tokyo, Japan	Urban	6.09		30.2	–	125	Var et al. (2000)
Ho Chi Minh, Vietnam	Urban	8.63		12.8	–	146	Hien et al. (2001)
Taichung, Taiwan	Urban	29.3		199	–	574	Fang et al. (2003)

– not detected or below LOQs

study may cause the degree of detected accuracy to decrease, especially under the situation of low concentration.

Correlation analysis and potential sources

Correlations between the concentrations of the elements may suggest common sources. The enrichment factor (EF) can be used to distinguish between natural and anthropogenic sources and to quantify the contributions from the natural and anthropogenic inputs (Ragosta et al. 2008). Here, Fe is used as the reference element to calculate EF values (Taylor and McLennan 1995). Fe, Mg, Al, and Mn have EF values of less than 1.5, suggesting a predominant contribution of these elements from soil and dust (Table 3). For mixed source-dominated elements (Cr, Cd, and Ni), EF values were between 2 and 10. Cd comes primarily from vehicle emissions and natural sources such as volcanic and crustal inputs. The slightly elevated Cr and Ni contributed to the industry emission. Pb, Cu, and Zn were found to be high with the value of Zn (mean) up to 95. The results may indicate that these elements mainly originate from anthropogenic activities. The atmospheric Pb, emitted from leaded petrol, is now less significant after the phasing out of leaded gasoline in China, and in other anthropogenic sources of Pb such as mining and smelting operations, coal burning may be becoming more important (Sun et al. 2006; Xu et al. 2012). However, Pb isotopic ratios suggested that vehicular emission still could be one of the sources of Pb in summer from city areas at present in China (Xu et al. 2012). In addition, Pb can be emitted into the atmosphere from the wind-blown dust and soil particles which are known to be highly contaminated due to the historical uses of Pb. Therefore, the high concentration of Pb in atmospheric particles from SCS probably resulted from the inputs of industrial and vehicle emissions and the use of Pb from local ores in the cities/regions around the SCS (Lee et al. 2007). The major sources of Cu in atmospheric particles from the globe are the combustion of fossil fuels, industrial metallurgical process, and waste incineration (Nriagu and Pacyna 1988). For Zn, smelting and incinerating operations could be the predominant source.

Al and Fe showed a good correlation indicating the absence of mineralogical differentiation or biological addition for the elements determined (Fig. 2). It also suggests the domination of dust origin for aerosol Fe. The Fe/Al ratios (mean) were 0.84 and 0.74 (1-year period) in Guangzhou and 1.0 and 0.67 in Hong Kong, respectively (Lee et al. 2007). Compared with

Table 3 EF values for trace metals in aerosol samples at different parts of the world

Site	Pb	Cu	Zn	Ni	Cr	References
Northern SCS (in 2005)[a]	82	41	95	3.5	1.4	This study
Northern SCS (in 2007)[a]	87	26	47	2.5	2.3	This study
East China Sea[b]	200	220	100	2.8	380	Hsu et al. (2010)
Mt. Muztagata, China	20	16	14	20	58	Makra et al. (2002)
Aksu, China	23	12	2	30	–	Zhang et al. (2003)
Zhenbeitai, China	40	11	4.2	25	–	Zhang et al. (2003)
Beijing, China	104	12	39	12.1	609	Sun et al. (2005)
Indian Ocean	17–104	120–328	98–1104	11–142	11–24	Witt et al. (2010)

Superscript letters a and b indicate mean values; – not available or not detected

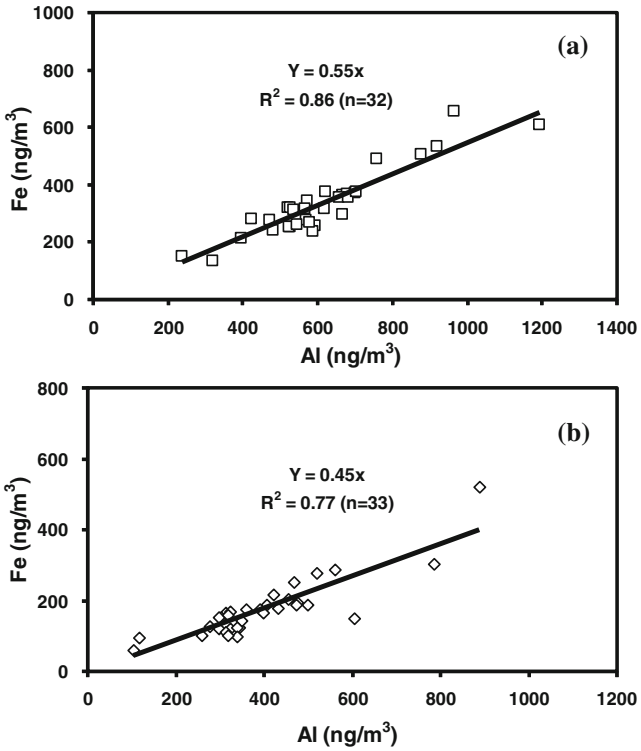

Fig. 2 Correlation of Al and Fe from the aerosol samples in 2005 (**a**) and 2007 (**b**)

urban and suburban areas, the figures were generally lower in remote regions and oceans (Cong et al. 2007). The mean Fe/Al ratios over SCS were 0.55 in 2005 and 0.40 in 2007, respectively. Obviously, the figures detected in 2005 were consistent with those measured in suburban and urban areas around SCS. Hence, more contaminants were transferred from continents to SCS during the sampling period in 2005 due to different climate conditions, such as wind direction and speed, temperature, and monsoons (see further discussion in "Effects of meteorological parameters" section).

Spatial variations and implication to source regions

Figure 3 exhibits the variations of trace metal concentration in the aerosols during the two sampling periods. Generally, the total concentrations of the five metals exhibited two peaks, and both appeared in samples close to the continent. For example, the samples 3–5 and 31–33 in 2005 were close to mainland China, which might imply some Southeast Asian countries as a major source. Similar results were also found for volatile organic pollutants within this region, such as pesticides, polychlorinated biphenyls (PCBs), and polychlorinated naphthalenes (Li et al. 2012; Zhang et al. 2007). In addition, the samples from the harbor (samples 15, 17, and 18 in 2005 and sample 13 in 2007) exhibited distinctly high concentrations, possibly providing evidence for a strong local source.

The spatial distributions of trace metals in the coastal and marine atmosphere were primarily influenced by surrounding land sources and atmospheric transportation. To investigate the possible outflows from different Asian source regions, air mass origins were determined for the cruise samples. In general, back trajectory analysis reveals that during sampling time, major air masses flowed from the East China Sea and Western Pacific along the coast or passed over Taiwan Island and the Philippines (Fig. S3). For example, at the beginning of the sampling in 2005, back trajectory analysis revealed the air mass stemmed from southeast of China and Taiwan island (type I). At that time, elevated concentrations of metals were observed both close to mainland China and over the open sea. It revealed that industrial smelting operations, coal burning, and vehicular emissions from northern inland areas of China and the PRD region could contribute to the enrichment of trace metals through the long-range transport of air pollutants via northerly winds. During the period from 13 to 21 September 2005, there were two types of air masses prevailing over the SCS (types II and III). Relatively high Zn and Cu were found in those samples when the air parcels near some Southeast Asian countries (type II). This result implies that some Southeast Asian countries might be one of the sources, mainly from ship dismantling industry. On the contrary, the marine air mass originated from North Pacific contained the relatively low concentrations of trace metal during the type III period (samples 20–22) possibly due to the dilution/mixing effects. At the end of sampling (samples 31 and 32), the air masses travelled through the southern coast of China, and high metals were observed whose air masses were from the northeast of the South China coastal region (type IV). Differently, sample 30 possesses relatively low levels whose air masses were from southwest of the South China coast. Similar phenomena were also observed on 27 August 2007, when elevated concentrations of Cu and Pb were found in sample 31. As suggested by backward air trajectory analysis, the high concentrations of trace metals were mainly related to air mass passing over the neighboring cities around the SCS.

Effects of meteorological parameters

Meteorological parameters such as wind speed and direction, air temperature, or annual precipitation have significant effects on the transportation of atmospheric pollutants. Due to the influences of monsoon and several tropical cyclones and typhoons, the mean wind speed during the sampling in 2005 was about two times higher than that in 2007. The highest wind speed reached to 30 m/s in the period of sampling due to the strong typhoons. Hence, it is reasonable to conclude that more atmospheric pollutants could be brought by strong wind from continental regions to SCS, which can partly explain the higher metals found over SCS in 2005. Except for wind speed, wind direction can also greatly affect the level of metals.

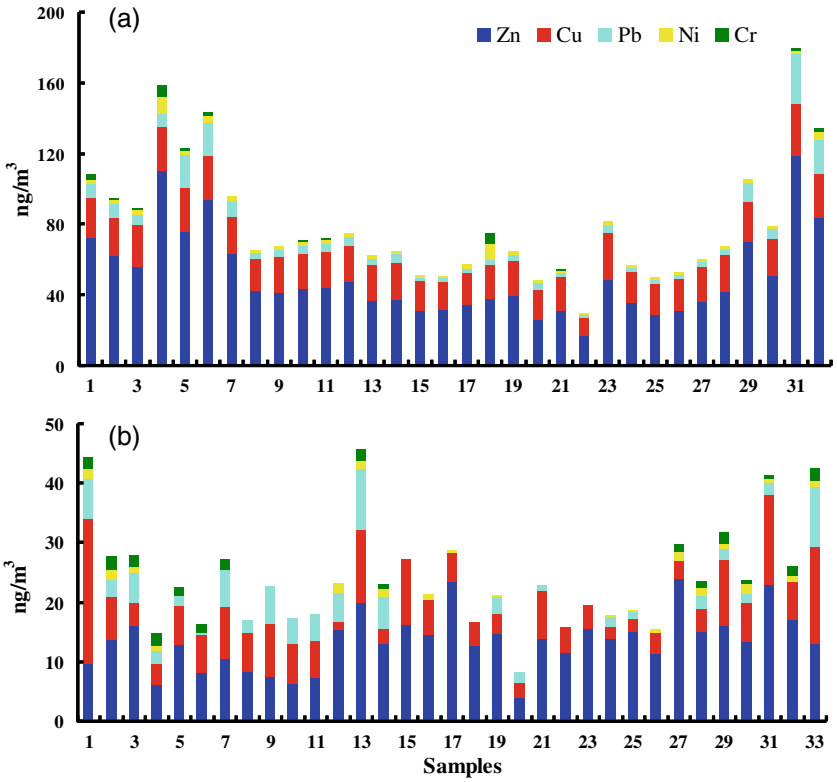

Fig. 3 Concentrations of trace metals over the SCS in 2005 (**a**) and 2007 (**b**)

Evidently, north and northeast wind can bring pollutants from the continent to SCS. On the contrary, south and southwest wind from West Pacific is relatively "clean." This can be shown by the samples 22 and 23 collected almost in the same area in 2005. However, noteworthy difference in concentration was found due to the adverse direction. Temperature and humidity are two more significant parameters to atmospheric pollutants, but mainly to volatile pollutants, such as POPs. In addition, temperature and humidity displayed inconspicuous changes during sampling periods in both 2005 and 2007. Therefore, they were not discussed here.

Except for local emissions, it has been well documented that aerosol can be transported from Asian region to other countries and remote ocean areas driven by Asian monsoons (Hsu et al. 2010; Kang et al. 2007; Nair 2006). Asia is the world's most important monsoon region dominated by two monsoon circulations: southwest monsoon (summer monsoon, from June to August) and northeast monsoons (winter monsoon, from September to March). Besides, the conversion period between the two monsoons is very short (Fig. S4). It should be noted that the aerosol sampling campaign of 2007 started at the beginning of August. Correspondingly, in 2005, the sampling campaign did not end until the end of September. Particularly, high level of trace metals in the aerosol can be transported from northern continents to the SCS by winter monsoon. Five years of investigation during 2003–2008 in the southern East Sea demonstrated that higher metals were also detected in winter and spring due to Asian dust driven by Asian monsoon (Kang et al. 2007). Further, the same seasonal pattern of volatile contaminants, such as pesticides, polycyclic aromatic hydrocarbons (PAHs), etc., were also found around the SCS regions due to the winter monsoon. However, during the summer monsoon, surface winds over the northern SCS originate in the south Indian Ocean, whereas winds aloft (northwesterlies) are mainly from the ocean area. Previous study revealed that the aerosols from long-range transport own lower Cd/Pb and Zn/Pb ratios (Hsu et al. 2005). In comparison to 2007, evidently lower Cd/Pb (mean 0.018) and Zn/Pb (mean 3.6) ratios were obtained in 2005, suggesting that the concentration variations may be partially attributed to the long-range transport of these pollutants.

Conclusions

A comprehensive survey of heavy metals in the atmosphere of northern SCS has been conducted. On the whole, the levels of trace metals detected in this study were generally higher than those in other open seas and oceans of the world and comparable with the values measured in suburban and background sites. Pb, Cu, and Zn were found to be highly enriched in the aerosol samples, indicating the main input from anthropogenic activities. The measured metals were influenced by their proximity to source regions and air mass origins. The potential sources of Pb, Cu, and Zn might be the local industrial and vehicular emissions. The back trajectory analysis showed that

high concentrations of trace metals were, to a large extent, related to the air mass from the surrounding regions of SCS, such as China, Taiwan, and the Philippines, reflecting an influence of the long-range air transport of metal contaminants from northern inland areas to the South China coast.

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