Aircraft Measurements of Ionic and Elemental Components in PM$_{2.5}$ over Eastern Coastal Area of China

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ABSTRACT

To understand the ionic and elemental components in PM$_{2.5}$ over eastern coastal areas in China, aircraft measurements were carried out from 25 December, 2002 to 6 January, 2003. PM$_{2.5}$ filter samples were collected and analyzed for mass concentrations, nine ionic components and 15 trace elements. The highest concentrations of PM$_{2.5}$ were observed at the lowest altitudes, indicating the influence of ground-level sources. Sulfate, nitrate and ammonium were the main water-soluble components, and the sum of their concentrations accounted for 60–70% of the total ionic mass. Cl$^-$ depletion was observed to an extent of 3–26%. Anthropogenic ally derived particles contributed about 60% of the total measured ionic components, and natural sources such as sea salts and soil dusts contributed another 40%. Different to many earlier studies, Ca^{2+} was found to mainly originate from anthropogenic sources. PM$_{2.5}$ showed an acidic nature, with a neutralization potential/acidic potential ratio of less than 1.0. NH$_4^+$ was the major neutralizer of aerosol acidity. A good correlation was observed between the concentrations of NO$_3^-$ and nssCa^{2+}, suggesting that photochemically produced HNO$_3$ was partly absorbed by mineral particles. S had the highest concentrations among the 15 elements. The enrichment factor values of the observed elements were all over 1, indicating that all of them were influenced by anthropogenic sources. The enrichment factors of Pb, S, As, Cu and Zn were over 100, and suggesting that they were greatly enriched.

Keywords: Aircraft measurements; Enrichment factor; Source analysis; Neutralization potential/Acidic potential ratio.

INTRODUCTION

Atmospheric aerosols play important roles in many environment problems, including visibility, earth radiation budget and human health effects. Aerosols can modify both short and long wave radiation by scattering the visible and absorbing the infrared of the spectrum (Charlson *et al.*, 1987; Andreae, 1996). Fine particulate matters can be breathed into human lungs and cause cardiovascular and respiratory illness (Samet *et al.*, 2000; Peters *et al.*, 2001). Atmospheric aerosols can also atmospheric haze and thus impair the visibility (Chiang *et al.*, 2007; Schäfer *et al.*, 2008; Zhang, 2010).

In the last decade, there has been a strong attention to the study of atmospheric aerosol in Asian. Many Asian countries (including China) have been experiencing increased energy consumption and rapid economic growth; emissions related to man-made activities are expected to have risen significantly. The increased emissions may have modified chemical compositions of atmospheric aerosols on regional to global scales and need to be monitored to evaluate their current effects and to predict potential future changes (Zhang *et al.*, 2009). In addition to emission sources related to man-made activities, natural sources of aerosol particles and their precursors also need to be understood. Since 1990s, considerable research investments in Asia and the western North Pacific region have brought together many field experiments at national and international scales, e.g., INDOEX, ACE-Asia, TRACE-P, APEX, PEACE, to study precursor gases, aerosol characteristics, and radiative forcing in the south and east Asian regions (Ramanathan *et al.*,

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2001; Huebert *et al.*, 2003; Jacob *et al.*, 2003; Nakajima *et al.*, 2003; Kondo *et al.*, 2004).

The eastern coastal region of China is located in the west rim of northern pacific and has undergone intensified industrial activities. Some studies have been conducted in this region to understand the chemical and physical properties of particulate matters (Zhang *et al.*, 2005; Wang *et al.*, 2006b; Zhang *et al.*, 2008a, b; Li *et al.*, 2011). However, all field studies were undertaken at surface level and knowledge of aerosol properties at upper levels is still limited. The purpose of the present study is to conduct aerosol composition measurements at upper levels using aircraft.

METHODS

Sampling Description

The aircraft measurements were conducted from 25

December of 2002 to 6 January of 2003 using a single engine Chinese Yun-5B light duty aerotransport. The flight region covered the coastal area from Wenzhou, Zhejiang province to Dalian, Liaoning Province (Fig. 1). Detailed flight information was shown in Table 1. Generally, flights 1 to 6 were conducted over the southern areas of China, and flights 7 to 10 were performed over the northern regions.

The instrumentation aboard the aircraft was designed to measure both physical and chemical properties of aerosols, as well as to provide a set of gaseous pollutants measurements such as O_3, SO_2 and nitrogen oxides. Size distribution measurement was performed using an aerodynamic particle sizer (APS 3310, TSI Inc, Minnesota). O_3 was monitored with an absorption ozone analyzer (Model 49C, TECO). SO_2 was monitored with a UV pulse fluorescence SO_2 analyzer (Model 43C-TL, TECO). Nitrogen oxides were measured with an ozone-chemiluminescence
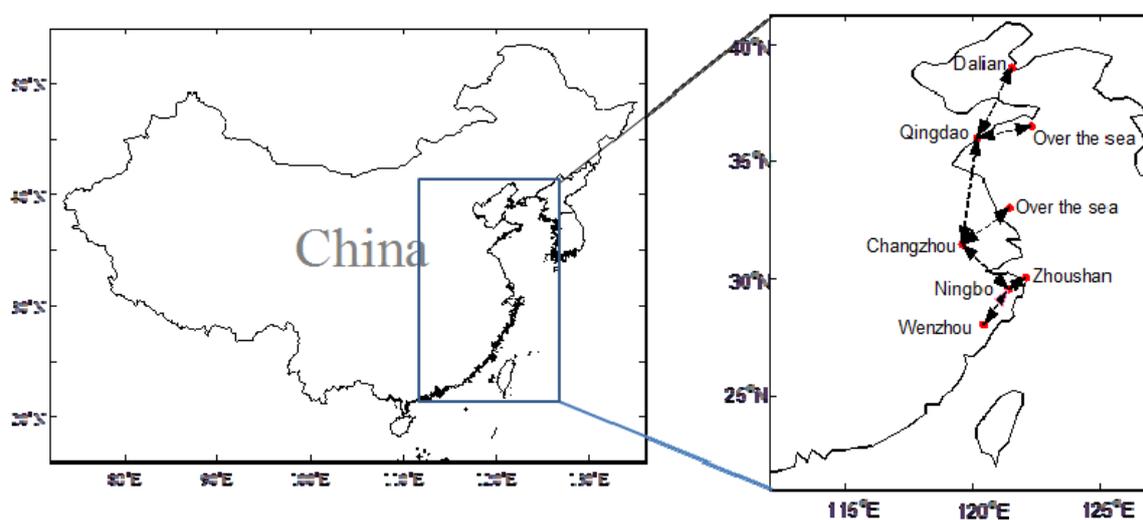

Fig. 1. Flight routes.

Table 1. Flight information.

Flight date	Sampling duration[*]	Flight NO.	Flight area	Flight altitude(m)
25 Dec., 2002	11:31–13:42	Flight 1a	Changzhou-over sea	2000
	14:22–15:51	Flight 1b	over sea-Changzhou	3000
26 Dec., 2002	10:15–12:36	Flight 2a	Changzhou-over sea	1000
	12:41–15:08	Flight 2b	over sea-Changzhou	500
27 Dec., 2002	9:14–10:27	Flight 3	Changzhou-Ningbo	1000
28 Dec., 2002	10:08–12:05	Flight 4a	Ningbo-Wenzhou	1000
	12:09–14:07	Flight 4b	Wenzhou-Ningbo	500
31 Dec., 2002	9:20–11:44	Flight 5a	Ningbo-Zhoushan	2210
	11:49–13:57	Flight 5b	Zhoushan-Ningbo	725
1 Jan., 2003	9:34–11:20	Flight 6a	Ningbo-Wenzhou	2000
	11:24–13:01	Flight 6b	Wenzhou- Ningbo	3000
3 Jan., 2003	12:09–15:16	Flight 7	Changzhou-Qingdao	2000
4 Jan., 2003	9:07–10:31	Flight 8a	Qingdao-Dalian	2000
	10:36–11:45	Flight 8b	Dalian-Qingdao	1200
5 Jan., 2003	10:59–12:29	Flight 9a	Qingdao-Over sea	2000
	12:33–14:03	Flight 9b	Over sea-Qingdao	500
6 Jan., 2003	12:00–15:15	Flight 10	Qingdao-Changzhou	500

[*] Excluding the ascent/decent data.

NO–NO$_x$ analyzer (Model 42C-TL, TECO). Filter samples were used to determine aerosol chemical composition. In this paper, only filter samples of PM$_{2.5}$ are presented.

Ambient air was introduced into the cabin of the aircraft through a stainless steel tube using an isokinetic inlet. The schematic diagram of the sampling systems was described in elsewhere (Wang *et al.*, 2006a).The collection efficiency of PM$_{2.5}$ was over 90%, which was calculated using the methods reported in earlier studies (Pui *et al.*, 1987; Muyshondt *et al.*, 1996).

A total of 16 PM$_{2.5}$ filter samples were obtained in the 16 flights. The PM$_{2.5}$ samples were collected by the sampler with the flow rate of 78 L/min made by China Geologic Device Factory (2nd Type). The filter for sample collection was the PF020 PTFE filter (ADVENTEC Company, Japan). The filter samples were kept chilling by using refrigerator during the transportation from the observation sites to the lab, and then were stored in a specific refrigerator at less than –24°C before analysis.

Analytical Techniques

One-quarter of the sample filter was extracted with 20mL of deionization water for 40 min and then the extract solution was filtered by 0.47 μm microfilm made by Beijing Chemical Technology School. The cations and anions in extract solution, including Na$^+$, NH$_4^+$, K$^+$, Ca^{2+}, Mg^{2+}, F$^-$, Cl$^-$, NO$_3^-$ and SO$_4^{2-}$, were analyzed using DX500 Ionic Chromatography (DIONEX, USA). Another area of the filter was extracted using an acid blended by HNO$_3$ and HF and HClO$_4$, then the residue solution was used to analyze the 25 elements like Al, Fe, Ca, Na, K, etc. using Inductively Coupled Plasma Emission Spectrometer (GE Company, USA), in which only the 15 main elements were discussed in this study. The two blanks filters were obtained for each flight. The blanks filters were quite steady with mass concentration of each detected chemical component being less than 5% of the mass concentration of PM samples.

Quality Control of Data

The ion balance technique was widely used to check the quality of the data. Using this check, the ratio of the sum of cations to the sum of anions was estimated. During our flight campaigns, a good correlation coefficient (0.99) with the slope of 1 was observed between the sum of cations and the sum of anions which indicated the good ionic balance.

The Backward Trajectory of Air Mass

To track sources of long-range transported particle, the 72-hour air mass backward trajectories were calculated using the HYSPLIT4 (Hybrid Single Particle Lagranian Integrated Trajectory, version 4) model (Draxler and Hess 2004) with FNL meteorological data developed by National Oceanic and Atmospheric Administration.

RESULTS AND DISCUSSION

Mass Concentration

Vertical variations of mass concentrations with flight altitude over southern and northern region were shown in

Fig. 2. It can be seen that the mass concentrations in the northern regions were higher than those in southern areas. The higher mass concentrations in the northern regions should be caused by coal burning for hearting, which emitted a large amount of particulate matters. In general, mass concentrations increased with the decreasing of altitude. The maximum concentration of 0.27 mg/m^3 was observed at 500 m suggesting the dominance of surface sources. Concentrations of PM$_{2.5}$ were smaller between 1000 and 4000 m. However, the mass concentrations at 1000 m in the southern region were conspicuously low, which might be related to air mass origins. Air mass trajectories arriving at these sites were plotted in Fig. 3. It was observed that the air masses at 500 m and 1500 m were predominantly from Mongolia and then travelled over ocean, which resulted in lower mass concentrations of PM$_{2.5}$.

Ionic Species

The ion concentrations of PM$_{2.5}$ during the whole flight campaign were shown in Fig. 4. All ion concentrations were shown as neq/m^3. Among the total anions, SO$_4^{2-}$ was the largest fraction (about 69%) with concentrations of 29.02–406.00 neq/m^3 followed by NO$_3^-$. Among the total cations, NH$_4^+$ was the largest contributor (47%) with concentrations of 17.11–331.75 neq/m^3 followed by Ca^{2+}. The sum of SO$_4^{2-}$, NO$_3^-$ and NH$_4^+$ accounted for 60–70% of the total ions mass concentrations, indicating the significant contributions of anthropogenic emissions to atmospheric aerosols.

Sources for Ionic Components

Using the computed crustal and marine fractions, we have categorized the ions into three main sources, ie. (i) crustal or soil originated, (ii) marine or sea salt, and (iii) anthropogenic or man-made sources. Na$^+$ was known to
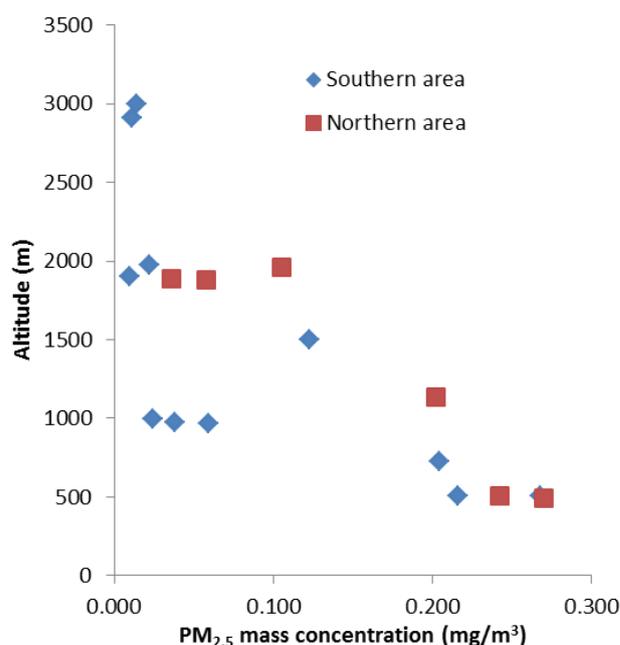

Fig. 2. Vertical distribution of mass concentrations of PM$_{2.5}$ over southern and northern area.
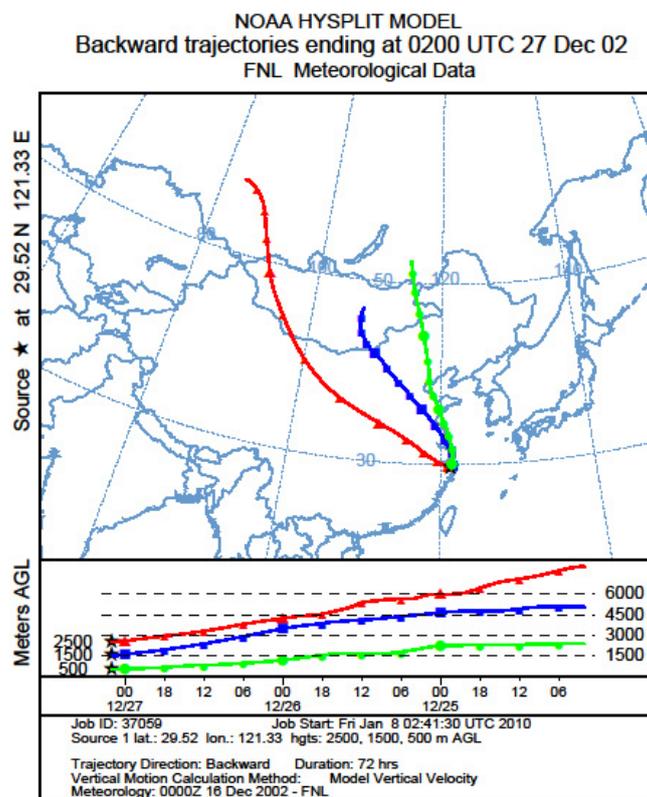
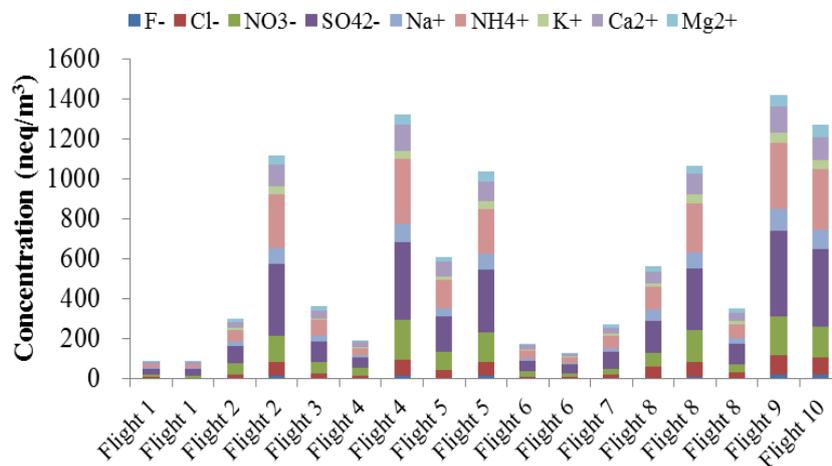**Fig. 3.** 72-hour air mass backward trajectories ending at Ningbo in Dec. 27, 2002.

Fig. 4. Ionic species in $PM_{2.5}$.

completely from sea salt; while F^-, Cl^-, NO_3^- and NH_4^+ were assumed to have no crustal origin. The marine contribution could be calculated by using the formula: $[X]_{marine} = ([X]/[Na^+])_{seawater} \times [Na^+]_{aerosol}$, in which X denotes the ion measured. If assuming Mg^{2+} was from marine and crustal sources only, the crustal portion of Ca^{2+} and K^+ could be estimated by the equivalent $(Ca^{2+}/Mg^{2+})_{crustal}$ and $(K^+/Mg^{2+})_{crustal}$ ratios of 1.87 and 0.48 (Huang *et al.*, 2008).

Assuming SO_4^{2-} from the crustal source was supplied by gypsum, it could be calculated using the formula $[SO_4^{2-}]_{crustal} = 0.47[Ca^{2+}]_{crustal}$ (Huang *et al.*, 2008); then the anthropogenic sulfate could be calculated using the following

formula: $[SO_4^{2-}]_{anthropogenic} = [SO_4^{2-}]_{total} - [SO_4^{2-}]_{marine} - [SO_4^{2-}]_{crustal}$. Using this method, 73–84% of SO_4^{2-} was estimated to be from anthropogenic sources, 14–24% from crustal sources, and 2.1–4.4% from marine sources. This suggests that the contribution from gypsiferrous ($CaSO_4$) particles was not negligible, consistent with the results from an earlier study (Guo *et al.*, 2010).

NO_3^- was completely from anthropogenic sources. $NssSO_4^{2-}$ and NO_3^- had a good correlation, since they were both produced from fossil-fuel combustion and subsequent photo-oxidations. The ratio of $nssSO_4^{2-}$ to NO_3^- can be used to identify the relative contributions of coal and gas or

oil burning to airborne particulate matters (Yao *et al.*, 2002). In general, the higher ratio suggested that airborne particulate matter mainly comes from the emission of coal burning, whereas the lower values indicated the more emissions from gas or oil combustion. During the flight experiments, the mean equivalent ratio of $nssSO_4^{2-}/NO_3^-$ was about 2.04, which was higher than the value reported in some developed countries such as U.S.A., indicating more coal combustion emissions in China. However, Compared with the results observed in Beijing in the winter of 1994 (4.58) and of 1995 (3.28) (Zhou *et al.*, 1998), our value was lower, which might be related to the increased emissions from mobile sources in recent years in China.

NH_4^+ was entirely from the anthropogenic sources with crustal and marine sources showing no contribution during all the flights. Major sources of NH_4^+ in China were probably the use of fertilizers in agricultural fields and human and animal excretions. Emissions from the burning of fossil fuels in various industries and in vehicular exhausts could also be sources. NH_4^+ is formed from the reactions between gaseous NH_3 and acidic species (e.g., H_2SO_4, HNO_3, and HCl). NH_4^+ showed a good correlation with both $nssSO_4^{2-}$ and NO_3^- (R^2 being 0.99 and 0.97, respectively), and also with the sum of NO_3^- and $nssSO_4^{2-}$, indicating these species were formed together as ammonium sulphate and ammonium nitrate. The slope of NH_4^+ to $nssSO_4^{2-}$ was 1.26. It is known that the equivalent ratio of NH_4^+ to $nssSO_4^{2-}$ for NH_4HSO_4 and $(NH_4)_2SO_4$ were 0.5 and 1.0, respectively. The slope obtained in winter 2002 was over 1.0, indicating that $(NH_4)_2SO_4$ was formed during these observations. The regression slope between NH_4^+ and the sum of $nssSO_4^{2-}$ and NO_3^- was 0.56 in $PM_{2.5}$. It indicates that there was about 44% of deficit in NH_4^+ compared to $nssSO_4^{2-}$ and NO_3^-. Sulphate was completely neutralized but this was not the case for all the nitrate.

Cl^- and Na^+ were mostly from sea-salt aerosols and there was no contribution from the crustal source. The ratio of Cl^- to Na^+ in seawater is 1.174 (Keene, 1986). From Fig. 5, it can be seen that Na^+ and Cl^- had a good correlation ($R^2 > 0.99$) with the slope of 0.87, which was lower than the seawater ratio of 1.174. The lower ratio indicated that the depletion of Cl^- had occurred. The percentage of Cl^- depletion can be calculated as:

$$Cl_{depletion}\ (\%) = (1.174[Na^+]_{measured} - [Cl^-]_{measured})/$$
$$1.174[Na^+]_{measured} \times 100 \qquad (1)$$

Fig. 6 showed the extent of Cl^- depletion with altitude. The extent of Cl^- depletion decreased with the increase of altitude over both ocean and land. At the low altitudes, the Cl^- depletion was relatively high, which was related to the high emissions of SO_2 and NO_x near the ground surface. The Cl^- depletion in aerosols was thought to be caused by various chemical processes involving sea-salt particles (Zhuang *et al.*, 1999).

The crustal contributions for Mg^{2+} varied from 54% to 75% whereas marine contributions varied from 25% to 46%. Most earlier studies suggest that crustal sources were the dominant sources for Ca^{2+}. Surprisingly, the present study identified that the dominant contribution for Ca^{2+} in $PM_{2.5}$ was from anthropogenic (48%–74%) with crustal sources only contributed about 23%–48%. The anthropogenic sources of Ca^{2+} could be attributed to the construction work. A good correlation was seen between the concentrations of $nssCa^{2+}$ and NO_3^-. This suggests that photochemically produced HNO_3 was mostly absorbed by mineral particles and that mineral particles acted as an important carrier for NO_3^- in this region. Similar results have also been found in previous observation in East Asia (Carmichael *et al.*, 1997). The molar ratio of $NO_3^-/nssCa^{2+}$ was about 1.58, suggesting
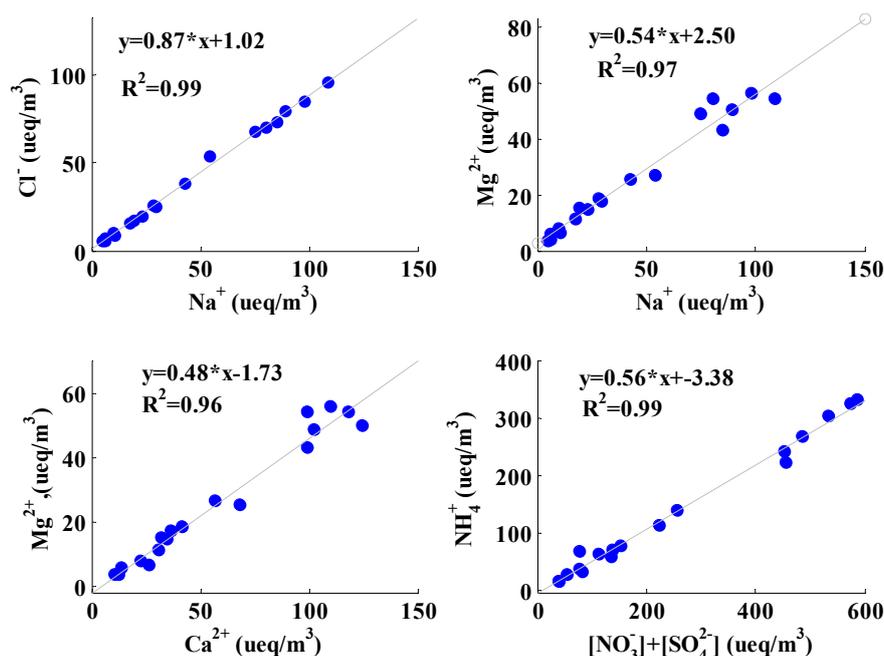

Fig. 5. Correlation between aerosol ionic species in $PM_{2.5}$. The solid lines represent the least squares fits.
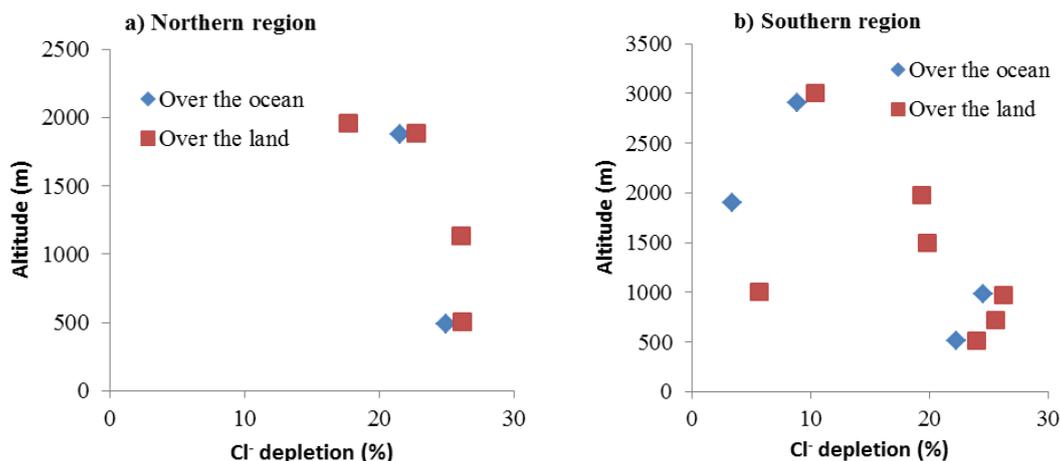
Fig. 6. Variation of Cl⁻ depletion with altitude over northern and southern regions.

that almost all soluble nssCa²⁺ was in the form of Ca(NO₃)₂. K⁺ was predominantly from the crustal source (over 50%), whereas anthropogenic sources contributed 30%–40%, and marine source contributed 3%–7%.

Finally, we quantified the respective contributions of these three sources (crustal, marine and anthropogenic) to the total measured ionic components of PM$_{2.5}$ (Fig. 7). During the whole flight campaign, anthropogenic sources were the dominant contributors to PM$_{2.5}$ (about 60%) while marine and crustal sources each contributed 20%. Both local emissions and long-rang transport of anthropogenic pollutants contributed to the observed PM$_{2.5}$ mass concentrations. During the flight observation campaign, wind speed was slow and air masses arriving at the eastern coastal areas were mostly originated from Mongolia and north-western region of china (Fig. 3).

Neutralization of Aerosol Acidity

The neutralization Factor (Nf) of major cations (Ca²⁺, NH₄⁺, Mg²⁺ and K⁺) were calculated to investigate their acid neutralizing capacity. In this calculation, NO₃⁻ and SO₄²⁻ were considered to be the major acidifying anions.

The role of Cl⁻ in the acidity as well as the role of Na⁺ in the alkalinity were negligible since they originated mostly from sea in the form of seasalts that were neutral. The seasalts portions of Ca²⁺, Mg²⁺ and SO₄²⁻ were also originated from sea in the form of neutral salt and were also excluded in the calculation of Nf. Thus, the calculation of Nf was according to:

$$NfX = NssX/[NO_3^- + NssSO_4^{2-}] \tag{2}$$

where X is NH₄⁺, NssCa, NssMg and NssK. Fig. 8 shows the neutralization factors for different cations in PM$_{2.5}$ at different altitudes. It can be observed that NH₄⁺ ion served as the major neutralizing components. Its average neutralization factor was 0.5. In general, the order of neutralization capacity was NH₄⁺ > Ca²⁺ > Mg²⁺ > K⁺.

To assess the balance between acidity and alkalinity, the ratio of Neutralization Potential (NP) to the Acidic Potential (NP/AP) was computed as follows:

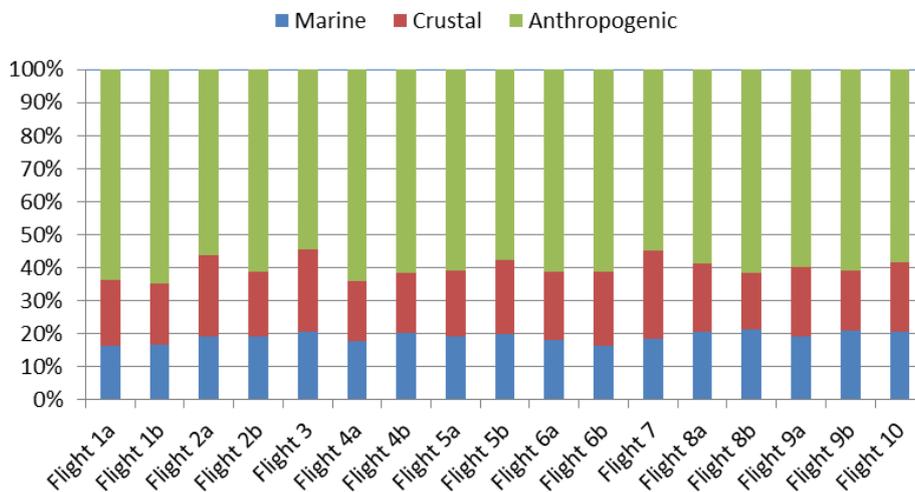$$NP/AP = ([NH_4^+] + [NssCa^{2+}] + [NssMg^{2+}] + [NssK^+])/([NssSO_4^{2-}] + [NO_3^-]) \tag{3}$$

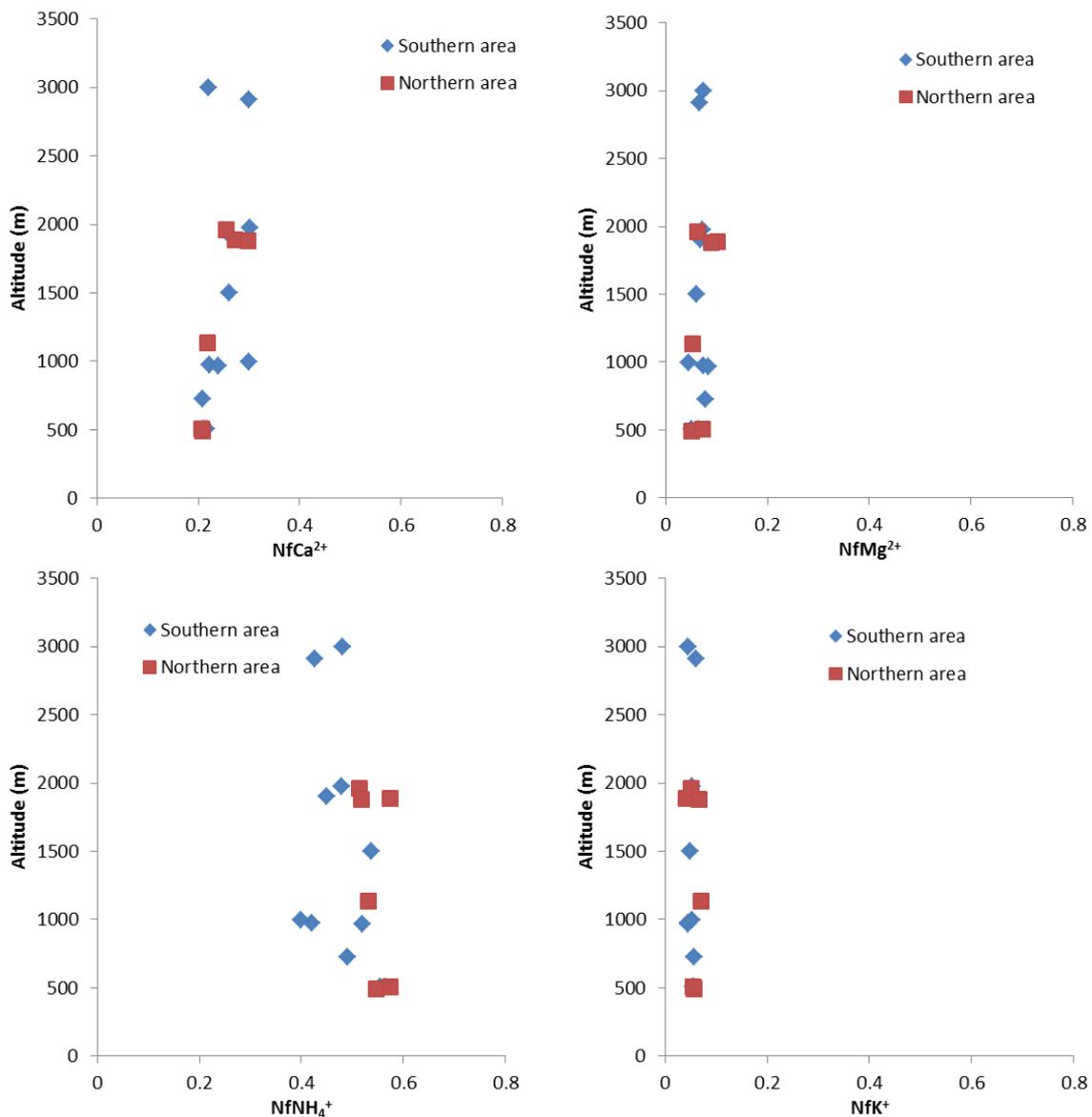**Fig. 7.** Contributions of different sources to the ionic components.

Fig. 8. The neutralization factors for different cations.

It can be observed that the ratio was less than one during the whole flight observation campaign indicating the AP dominated the NP and PM$_{2.5}$ showed acidic nature.

Elemental Composition

Element concentrations for 15 species in PM$_{2.5}$ were analyzed (Fig. 9). The element concentrations followed the order of S > Ca > Fe > Na > Al > K > Mg > Pb > P. S had the highest concentration (over 3.3 μg/m^3) and accounted for 31.44% of the total elements, indicating the effects of coal-burning activities.

Enrichment factor (EF) is one method for judging pollution characteristics and origins of particulate matter (PM). In general, PM coming from soil and dust contains more crustal elements, of which the enrichment factors are usually small. Thus, the elements with EF values smaller than 1 are commonly assumed to have relatively small influence from anthropogenic activities. Similarly, the elements having

EF values between 1 and 10 are influenced by both natural sources and anthropogenic origins, and the elements having EF larger than 10 are believed to be greatly enriched due to human activities. If we could identify the sources of the elements with high enrichment factors, we could then evaluate the influence of such pollution events on the characteristics of PM. Using Al as the reference element, the enrichment factor was calculated as following:

$$EF = (Cx/Al)_{aerosol}/(Cx/Al)_{crust} \tag{4}$$

where "$(Cx/Al)_{aerosol}$" refers to the ratio of the concentration of the studied element Cx to that of the reference element Al in aerosol and $(Cx/Al)_{crust}$ was the same ratio in the crustal (Taylor 1964), respectively.

As shown in Fig. 10, the EF values of the observed elements were over 1, indicating that all of them were partly influenced by anthropogenic sources. The enrichment
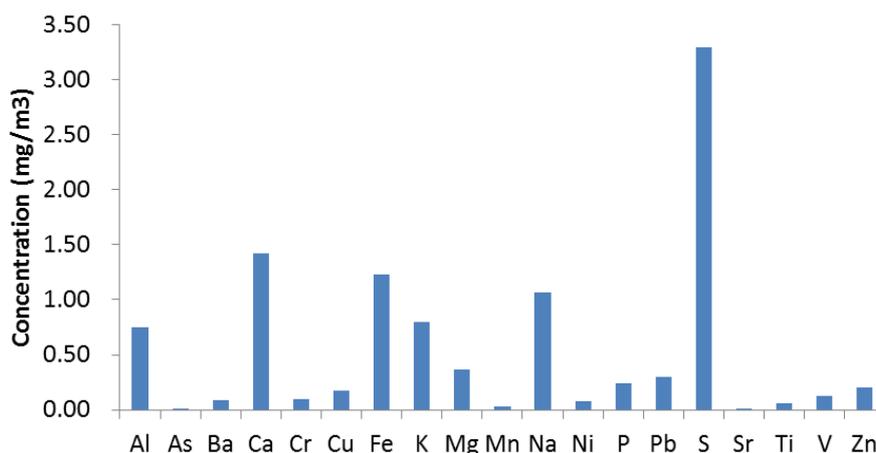
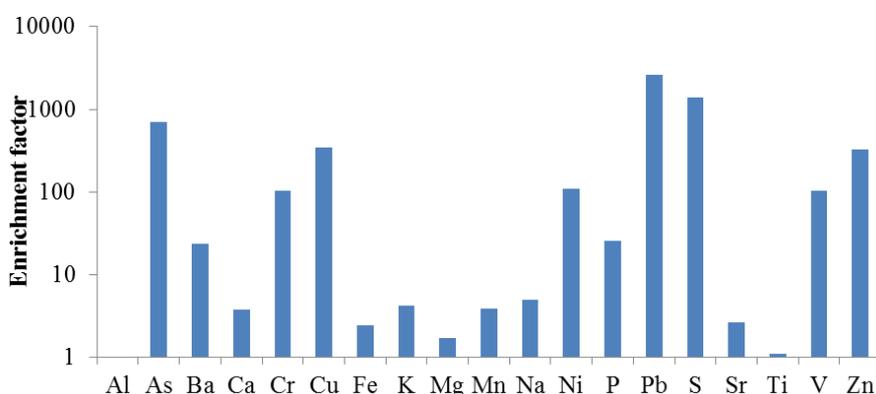**Fig. 9.** Concentrations of elemental fractions.

Fig. 10. Enrichment factors for elemental fractions.

factors of Pb and S were more than 1000, and those of As, Cu and Zn were over 100, indicating that these elements were greatly derived from anthropogenic pollution sources. The enrichments of As and S was possibly caused by coal-combustion. Pb could come from industrial processes or low-tech coal combustion (Li *et al.*, 2008; Li *et al.*, 2010; Widory *et al.*, 2010). The enrichments of Zn and Cu were possibly caused by smelting industry. The enrichment factors of Ca, Fe, K, Mg, Mn, Na, Sr and Ti were between 1 and 10, indicating that they were influenced by both anthropogenic emissions and natural source or soil particles.

CONCLUSIONS

PM$_{2.5}$ was measured by aircraft over eastern coastal areas in winter (12/25/2002–1/6/2003). Mass concentration, nine ionic species and 15 elements were analyzed. The highest concentrations of PM$_{2.5}$ were observed at the lowest altitude, indicating the influence of ground-level sources. Sulfate, nitrate and ammonium were the main water-soluble components with the sum of their concentrations accounted for 60–70% of the total ions mass. Part of Cl$^-$ was depleted in measured aerosols. SO$_4^{2-}$ was predominantly from anthropogenic sources and with negligible crustal sources. Mean equivalent ratio of nssSO$_4^{2-}$/NO$_3^-$ was about 2.04, indicating coal combustion emissions were the main sources

of particulate matter in China. Anthropogenically derived particles contributed about 60% of the total measured ionic components. Natural sources like sea salt and soil dust contributed about 40%. Ca^{2+} was mainly from anthropogenic sources. Crustal sources were the dominant contributor for Mg^{2+}. NH$_4^+$ was the major neutralizer of aerosol acidity. Neutralization potential/Acidic potential ratio was less than one indicating that PM$_{2.5}$ was acidic. A good correlation was observed between the concentrations of NO$_3^-$ and nssCa^{2+}, showing that photochemically produced HNO$_3$ was partly absorbed by mineral particles. S had the highest concentrations in fifteen elements, indicating the effect of coal-burning activities. The enrichment factor values of observed elements were all over 1, indicating that all of them were influenced by anthropogenic sources. High enrichment factors of Pb, S, As, Cu and Zn indicated that they were largely derived from anthropogenic pollution sources. Pb could come from industrial processes or low-tech coal combustions.

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