



Source Apportionment of PM₁₀ in Four Cities of Northeastern China

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ABSTRACT

Ambient particulate matter with the aerodynamics diameter less than 10 μm (PM₁₀) was sampled in four northeastern Chinese cities (Shenyang, Anshan, Fushun and Huludao) from August 2001 to August 2005. Chemical compositions including 20 elements, SO₄²⁻, organic and total carbon were determined. In addition, chemical source profiles consisting of the same particulate components were obtained from a number of naturally occurring geological sources (soil dust from exposed lands and marine salt) and sources of atmospheric particulates resulting from human activities (construction derived dust, coal combustion fly ash, iron and steel manufacturing dust, zinc dust, vehicle exhaust, and sulfate). Chemical mass balance modeling (CMB) was applied to determine the particulate matter (PM) sources and their contributions to PM₁₀ in these four cities. The results showed that soil dust and coal fly ash were the major sources of ambient PM₁₀ in all four cities. The construction derived dust, iron and steel manufacturing dust also was the major source of PM₁₀ in Huludao, Anshan, respectively. Higher contribution of iron and steel manufacturing dust (33.3 μg/m³, 20.9%) in Anshan was observed in this study, compared with previous studies. The zinc dust was a special PM source of Huludao, with a contribution of 19.1 μg/m³ (7.0%) to PM₁₀.

Keywords: Source apportionment; PM₁₀; Chemical mass balance; Cities of northeastern China.

INTRODUCTION

Particulate matter (PM) has been implicated in negative respiratory and cardiovascular health outcomes, including effects leading to premature mortality (Ostro *et al.*, 2001; Pope *et al.*, 2002; Chen *et al.*, 2005; Oftedal *et al.*, 2008; Gan *et al.*, 2011). PM also have a pronounced effect on atmospheric chemistry and air quality including cloud formation, visibility, solar radiation and acid deposition (Khoder, 2002; Watson, 2002; Celis *et al.*, 2004; Tsai and Chen, 2006). Total suspended particulates (TSP) and PM with the aerodynamics diameter less than 10 μm (PM₁₀) were considered as the potential reason of heavy air pollution in China, especially in northern China. According to the Report on the State of the Environment in China (State Environmental Protection Administration (SEPA), 2002–2007), PM₁₀ were found to be the principal air pollutants of China's urban areas. In 2004, the TSP and PM₁₀ concentrations of 210 cities in China (61.40% of total cities being monitored) did not meet the annual average concentration of National Ambient Air Quality Standard (NAAQS), with 200 μg/m³ for TSP and 100 μg/m³ for

PM₁₀ (SEPA, 2005), and the TSP and PM₁₀ annual average concentrations of the four cities (Shenyang, Anshan, Fushun and Huludao) of Liaoning province (northeastern China) examined in this study all exceeded the NAAQS. Therefore, it is important to understand which emission sources contribute to the high PM₁₀ levels for developing effective control strategies to reduce the PM₁₀ concentration for the public health. Source apportionment techniques for airborne particulate matter are the method that quantifies the contribution of different sources to airborne particulate matter concentrations at receptor locations in the atmosphere. Source apportionment techniques are valuable tools that aid in the design of effective emissions control programs to reduce particulate air pollution (Gupta *et al.*, 2007). Receptor model is the application of multivariate statistical methods addressed to the identification and quantitative apportionment of air pollutants to their sources (Callén *et al.*, 2009), mainly including chemical mass balance (CMB) (Watson and Chow, 2001; Samara *et al.*, 2003; Gupta *et al.*, 2007; Stone *et al.*, 2010), principal component analysis (PCA) (Querol *et al.*, 2001; Pandolfi *et al.*, 2008; Moreno *et al.*, 2009), and positive matrix factorization (PMF) (Yatkin and Bayram, 2008; Pandolfi *et al.*, 2010; Yubero *et al.*, 2010). Source apportionment studies of urban ambient PM₁₀ using CMB have also been carried out in some cities of China in recent years (Senlin *et al.*, 2007; Wu *et al.*, 2009; Kong *et al.*, 2010). Bi *et al.* (2007) summarized the results

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of source apportionment for ambient PM₁₀ in six cities of northern China using CMB and concluded that urban resuspended dust was a major contributor for ambient PM₁₀ in all six cities. And significant contributions from coal combustion fly ash were also found in the six cities, especially during winter. However, in the northeastern China, available data or researches on the PM₁₀ sources and contributions to ambient PM₁₀ were limited. Northeastern China is a traditional heavy industry base, covering a wide range of industries, such as machinery, electronics, metal refining, petroleum, chemical industries, construction materials, coal, and so on. Most of these industries are high energy demand and heavy pollution emission sections.

Liaoning province was one of the most heavy air pollution provinces in northeastern China, and the cities in the province had been suffering from high levels of PM pollution (Department of Environmental Protection of Liaoning Province (DEPLP), 2002–2006). The four cities (Shenyang, Anshan, Fushun and Huludao) of Liaoning province examined in this study were all experiencing high concentrations of PM₁₀, therefore, it is important to identify the major potential contributors and estimate their contributions to ambient PM₁₀. This study conducted ambient PM₁₀ sampling in the four cities from 2001 to 2005, and constructed PM₁₀ source and receptor profiles. The source identification and apportionment of PM₁₀ in urban areas of the four northeastern Chinese cities were determined via a CMB receptor model.

METHODOLOGY

Study Area

Shenyang (41°50'N, 123°24'E), Anshan (41°7'N, 122°59'E) and Fushun (41°52'N, 123°55'E) are located in the middle of Liaoning province, while Huludao (40°56'N, 120°38'E) is a coastal city in the southwest of Liaoning Province. The locations of four cities are provided in Fig. 1. All four cities experience a continental monsoonal climate, with similar characteristics of hot, humid summers, and dry, cold winters. The mean annual precipitation of Shenyang, Anshan, Fushun and Huludao are 619 mm, 720 mm, 700 mm, 638 mm, respectively and nearly half of the annual rainfall occurs in July and August. The domestic heating period in four cities is generally between November and March due to the cold winter and non-heating period is between April and October. During domestic heating period, numerous coal was consumed and large amount of pollutant was released to atmosphere. Moreover, the four cities are all traditional industrial cities in China, with coal as the predominant energy. Therefore, coal combustion fly ash and industrial emission were considered as potential sources in the four cities. In addition, the large and intense population (7,000, 000 in Shenyang, 3,584,000 in Anshan, 2,268, 100 in Fushun and 2,709,000 in Huludao) in the four cities made the traffic emission should not be ignored.

Ambient Sampling

The descriptions of sampling time of the four cities are described in Table 1. Ambient PM₁₀ samples were collected

in two weeks (a heating period week and a non-heating period week), in each city of different years, that represent heating and non-heating period, respectively at sites that represent different functional areas (city downtown areas, industrial areas, residential areas and heavy traffic areas). Collection of samples began at 8:00 local time and continued for 24 h. Ambient PM₁₀ samples were collected using medium-volume samplers (Model TH-150S, manufactured by Tianhong Instrument Co., Ltd. Wuhan, China) operating at a flow rate of 100 L/min with a 10 μm cut-point impactor. The flow rate of each sampler, equipped with a mass flow controller, was calibrated automatically with bubble flow meters. Flow rate variations throughout each 24 h sampling period were within ± 5% (Kong *et al.*, 2010). Ambient air particulate matters were continuously collected on polypropylene-fiber filters (Ø = 90 mm, Pallflex2500 QAT-UP) for elemental analysis, and on quartz-fiber filters (Ø = 90 mm, Pall Gelman Laboratory, Ann Arbor, MI) for ionic/carbon component analysis and determination of gravimetric values of PM₁₀.

Polypropylene filters were calcined at 60°C for 0.5 h and quartz fiber filters at 800°C for 2 h to remove any organic compounds that may be present on the filters before sampling. After sampling, the filters were removed from the inlet and folded in half and wrapped in a laminar-flow clean hood until use (Kong *et al.*, 2010). Filters were weighed before and after sampling by a microbalance (Meter Toledo M5) with balance sensitivity ± 0.010 mg. All filters were equilibrated at room temperature for 48 h in a desiccator before weighing commenced. After weighing, the loaded filters were stored in a freezer under conditions of about – 4°C until chemical analysis. A total of 525 available ambient samples were collected in the four cities, and 91 invalid ambient samples were rejected due to the rain, sampler's malfunction, filter's fracture, or other unexpected accidents.

Source Sampling

Source emissions samples including soil dust, coal fly ash, iron and steel manufacturing dust, construction derived dust and zinc dust were collected in the four cities. Soil dust was collected from exposed lands within 15 km from the urban areas with the sampling sites located in the different direction (east, south, west, north and prevailing wind direction) of the four cities. In every site, topsoil and the soil 20 cm below the surface were collected (1–2 kg) and mixed with the ratio of 1:1. Sampling from industrial stacks was not possible due to their big height (Samara *et al.*, 2003), therefore coal fly ash were collected from the air pollution control devices (electrostatic precipitators, cyclone dust separators or wet scrubbers). Construction derived dust was collected from cement floors, windowsills and stairs of the buildings under construction, and also from production lines of nearby cement factories in each city. Smelting dust including iron and steel manufacturing dust from production lines of steel factories (Shenyang, Anshan and Fushun) and zinc dust emitted from production lines of Huludao Nonferrous Metals Group Co. were collected. The collected source samples were stored in labeled polyethylene bags, then dried in a dark room with ventilation devices and

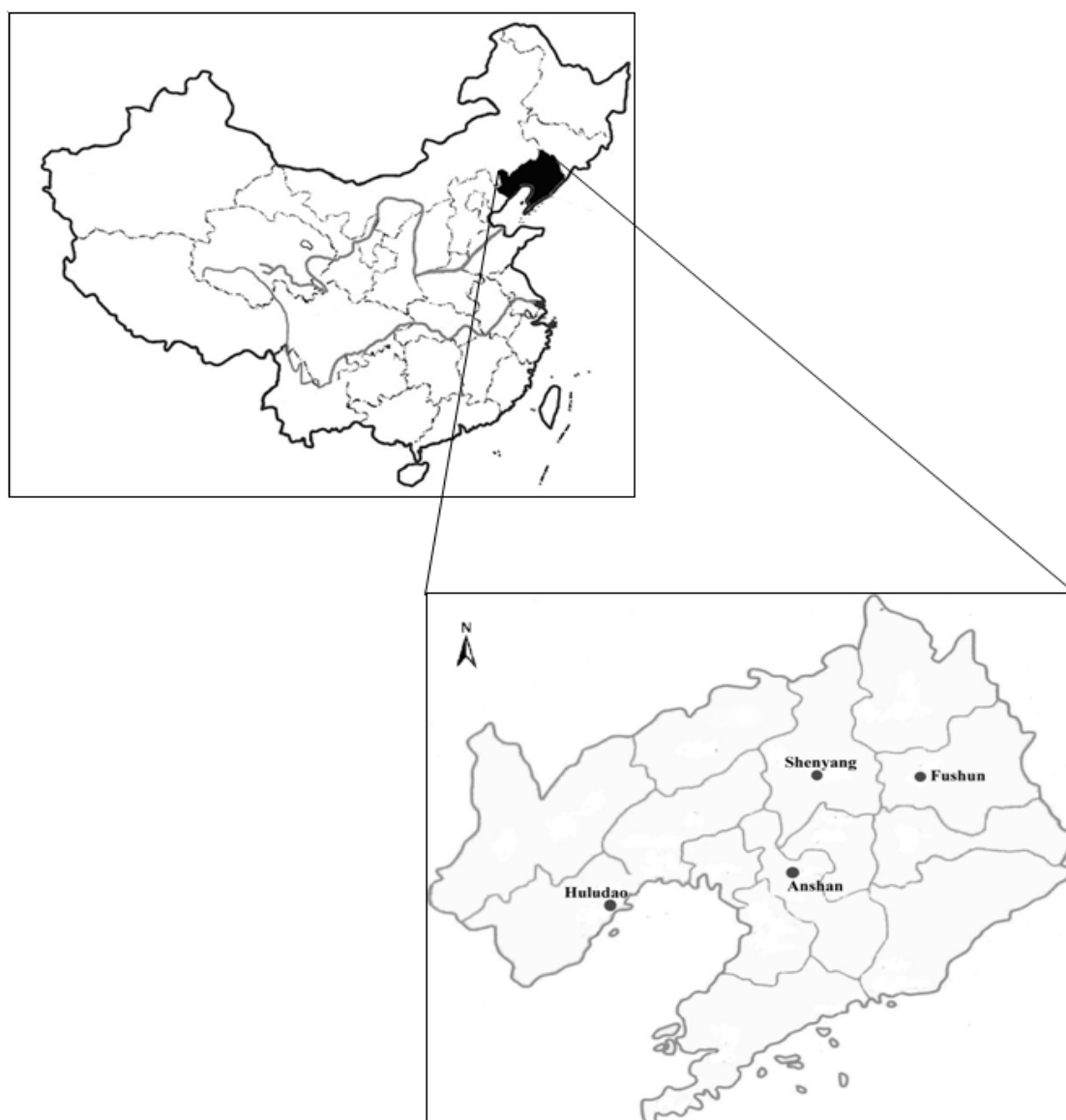


Fig. 1. Locations of the four cities in northeastern China.

Table 1. Descriptions of sampling time of the four cities.

City	Non-heating period	Heating period
Shenyang	August 2001	January 2002
Anshan	August 2005	March 2005
Fushun	July 2002	January 2002
Huludao	May 2003	January 2004

sieved through 150 mesh nylon sieves. The sieved samples from different locations within a given source type were thoroughly mixed, then suspended in a chamber and sampled through size selective inlets onto filters to obtain the PM₁₀ samples used for analyses (Chow *et al.*, 1994, Vega *et al.*, 2001).

Chemical Analysis

Inductively coupled plasma-atomic emission spectrometer (Baldwin *et al.*, 1994) (ICP–AES, IRIS Intrepid II, Thermo Electron) and inductively coupled plasma-mass spectroscopy

(Chio, *et al.*, 2004; Han, *et al.*, 2009; Kong, *et al.*, 2011) (ICP-MS) (Agilent 7500a, Agilent Co. USA) analysis were employed for the determination of Na, Mg, Al, Si, P, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Pb, As, Ba, Sr in samples collected on polypropylene-fiber filters and from dust samples. Water soluble ions (SO₄²⁻) collected with quartz-fiber filters and from dust samples were measured using ion chromatography (Chow and Watson, 1999) (DX-120, Dionex Ltd., USA) after sample extraction with deionized water. Total carbon (TC) and organic carbon (OC) were determined by improve thermal/optical reflectance (TOR) method with DRI Model 2001 Thermal/Optical Carbon Analyzer (Louie *et al.*, 2005). Blanks (including filters) and duplicate sample analyses were performed for approximately 10% of all the samples. Blank filters were processed simultaneously with field samples. In this study, the method detection limits (MDLs) of an analysis was defined as the value of standard deviation in the replicate analysis of blanks, multiplied by a conservative factor of

three (Morales *et al.*, 1996; Chan, *et al.*, 1999). Certified reference materials (CRM) were used to ensure good accuracy and precision (National Research Center of CRM, China).

CMB Receptor Model

The chemical mass balance (CMB8.2) receptor model (Watson *et al.*, 1997) from the US Environmental Protection Agency (US EPA) was applied to quantify the source contributions to PM₁₀ in the four cities. The CMB consists of a least-squares solution to a set of linear equations which expresses each ambient concentration of a chemical species as a linear sum of products of the corresponding source profile species and source contribution estimates (SEC) (Mazzera *et al.*, 2001). Uncertainties of ambient concentrations and source profile species are used both to weight the least-squares regression and to estimate the SEC uncertainties (Watson *et al.*, 1984). The details of the CMB model are previously described (USEPA, 2006).

RESULTS AND DISCUSSION

Ambient PM₁₀ Measured Concentrations

The measured ambient PM₁₀ concentrations of the four northeastern Chinese cities (Shenyang, Anshan, Fushun and Huludao) are given in Table 2. The annual average concentrations of PM₁₀ were calculated using Eq. (1) (Bi *et al.*, 2007; Wu *et al.*, 2009). The results were shown in Table 2.

$$C \text{ (Shenyang, Anshan, Fushun, Huludao)} \\ = ([\text{Non-heating}] \times 215 + [\text{Heating}] \times 150) / 365 \quad (1)$$

where C (*), annual average PM₁₀ concentration; [Non-heating], the average PM₁₀ concentration in non-heating period; [Heating], the average PM₁₀ concentration in heating period.

None of the annual average PM₁₀ concentration in the four cities met the NAAQS of 100 µg/m³. Huludao exhibited the highest measured annual ambient PM₁₀ concentration. Shenyang and Anshan had annual ambient PM₁₀ concentrations similar to those of Fushun. In this study, we found that the pattern of pollution in Shenyang, Anshan and Fushun was in accordance with that reported for northern China: Heating period exhibited higher PM₁₀ concentrations than non-heating period (attributed to increased coal combustion during heating period). During non-heating period, PM₁₀ concentration in Huludao was

much higher than that in Shenyang Anshan and Fushun. In the present study, Huludao PM₁₀ samples were collected in May while Shenyang, Anshan and Fushun PM₁₀ samples were collected in July/August during non-heating period (Table 1). It's windy and dusty with the wind speed up to 4.18 m/s in May in Huludao while the weather is relatively calm with abundant precipitation in July/August in other three cities. This may lead to relatively higher concentration of PM₁₀ in Huludao in non-heating period.

The chemical composition of ambient PM₁₀ (inorganic elements, SO₄²⁻ and carbon component) of the four cities is summarized in Table 3.

Characterizations of Source Profiles

The source profiles are the fractional abundances of chemical species in the source emissions (Mazzera *et al.*, 2001). The logograms of each source, corresponding by city, are listed in Table 4. The source profiles, including soil dust, coal fly ash, construction derived dust, iron and steel manufacturing dust, zinc dust and vehicle exhaust dust, are presented (mean ± SD) in Table 5–8. The uncertainties were the standard deviation of several samples with the same source type collected in the same city.

Table 5 shows that Si, Al and Ca are abundant constituents in soil dust from four cities (Si in soil dust from Fushun was not measured) with low variability in the range of 21.66%–28.20%, 6.45%–10.71% and 1.27%–2.47%, respectively. In all four cities, HLSD (soil dust of Huludao) profile contained the most abundant Si, which was 24.8% higher than the corresponding abundance in Tianjin profile; The most abundant Al was also found in HLSD profile, which was 19.3% higher than that from Taiyuan; ASSD (soil dust of Anshan) profile contained the most Ca, which was much lower than that in Tianjin and Taiyuan profiles (Bi *et al.*, 2007). Fe, TC and OC were found in abundance in soil dust in four cities with high variability in the range of 2.03%–6.95%, 1.05%–4.92% and 0.76%–3.13%, respectively. The abundance of Fe in ASSD profile was much higher than that observed in SYSD (soil dust of Shenyang), FSSD (soil dust of Fushun) and HLSD profiles. TC and OC in HLSD profile were significantly lower than those observed in SYSD, ASSD, FSSD profiles. The OC/TC ratios in these four soil dust profiles ranged from 0.48 to 0.72. The ratios are lower than Imperial and Mexicali Valleys soil profile (Watson and Chow, 2001) and Hong Kong urban soil profile (Ho *et al.*, 2003).

Coal fly ash, mainly emitted from various coal combustion sources, is a very important source category of

Table 2. Measured ambient PM₁₀ concentrations of four cities in northeastern China^a

City	Number of sampling sites	Heating period (µg/m ³)	Non-heating period (µg/m ³)	Annual average concentration (µg/m ³)
Shenyang (114 ^a)	5	247.0	147.0	188.1
Anshan (186 ^a)	8	252.7	94.2	159.4
Fushun (123 ^a)	5	195.9	136.8	161.1
Huludao (102 ^a)	4	270.8	278.6	275.4
average		241.6	164.1	196.0

^aNumber of valid ambient samples.

Table 3. Summary of chemical composition of ambient PM₁₀ samples of the four cities in this study (annual average, µg/m³)^a.

Species	Shenyang	Anshan	Fushun	Huludao
Na	2.88 ± 1.86	2.12 ± 0.89	0.22 ± 0.07	6.38 ± 1.88
Mg	3.00 ± 1.58	0.95 ± 0.42	0.33 ± 0.12	3.47 ± 1.48
Al	8.35 ± 4.18	2.18 ± 1.79	1.59 ± 0.43	14.18 ± 3.23
Si	22.33 ± 14.28	3.55 ± 1.52	-	37.68 ± 10.66
P	1.30 ± 0.47	-	-	-
K	4.63 ± 2.41	7.75 ± 1.97	1.80 ± 0.52	9.01 ± 2.19
Ca	8.22 ± 3.44	2.84 ± 0.76	1.98 ± 0.70	22.43 ± 13.03
Ti	0.68 ± 0.44	17.99 ± 11.50	0.22 ± 0.07	1.23 ± 0.40
V	0.03 ± 0.01	0.29 ± 0.17	0.01 ± 0.01	0.05 ± 0.01
Cr	0.09 ± 0.08	0.00 ± 0.00	0.16 ± 0.05	0.05 ± 0.02
Mn	0.38 ± 0.17	0.02 ± 0.02	0.15 ± 0.07	0.67 ± 0.19
Fe	6.04 ± 3.99	0.13 ± 0.10	3.74 ± 1.71	12.60 ± 3.19
Co	0.03 ± 0.00	6.15 ± 7.47	-	0.06 ± 0.03
Ni	0.04 ± 0.02	0.01 ± 0.01	0.02 ± 0.02	0.06 ± 0.02
Cu	0.23 ± 0.13	0.01 ± 0.01	0.03 ± 0.01	0.22 ± 0.10
Zn	1.52 ± 0.91	0.04 ± 0.03	0.85 ± 0.45	12.90 ± 10.66
Pb	0.36 ± 0.19	0.00 ± 0.00	0.22 ± 0.09	1.96 ± 1.02
As	-	0.56 ± 0.33	0.01 ± 0.01	-
Ba	0.13 ± 0.07	-	0.06 ± 0.02	-
Sr	-	-	0.03 ± 0.03	-
TC	53.94 ± 23.60	33.71 ± 16.50	33.04 ± 10.81	37.64 ± 2.87
OC	43.49 ± 22.86	25.80 ± 13.92	29.47 ± 9.89	22.51 ± 1.85
SO ₄ ²⁻	-	0.95 ± 0.35	-	27.04 ± 8.40

^aHyphens refer to the unanalyzed species.

Table 4. Logograms of each source corresponding to city^a.

	soil dust	coal fly ash	iron and steel manufacturing dust	zinc dust	construction derived dust
Shenyang	SYSD (26 ^b)	SYCFA (10 ^b)	SYISD (10 ^b)	-	SYCD (16 ^b)
Anshan	ASSD (16 ^b)	ASCFA (22 ^b)	ASISD (26 ^b)	-	ASCD (8 ^b)
Fushun	FSSD (24 ^b)	FSCFA (16 ^b)	FSISD (10 ^b)	-	FSCD (8 ^b)
Huludao	HLSD (16 ^b)	HLDCFA (12 ^b)	-	HLDZD (8 ^b)	HLDCD (8 ^b)

^aHyphens refer to the sources without samples.

^bNumber of samples.

ambient PM₁₀. The chemical profile of coal fly ash in the four cities was found to have abundant species of TC, OC, Si and Al (Table 6). The proportions of TC, OC, Si and Al are in the range of 13.60–27.70%, 8.10–25.60%, 7.85–17.99%, 8.59–14.82% for the four cities, respectively. The coal fly ash was also enriched in Fe (1.73–6.79%) and Ca (1.05–4.28%). For comparison, a PM₁₀ coal fly ash profile reported by Chow *et al.* (2004) contained 1.06% TC, 0.41% OC, 13.4% Si, 10.7% Al, 3.7% Fe and 19.8% Ca. Si is the most abundant constituent in HLDCFA (coal fly ash of Huludao) profile while TC is the constituent with greatest mass percent in coal fly ash profiles of the other three cities. The OC/TC ratio in ASCFA (coal fly ash of Anshan) profile (0.60) is much lower than that in SYCFA (coal fly ash of Shenyang), FSCFA (coal fly ash of Fushun) and HLDCFA profiles (0.90–0.94). The OC/TC ratios of coal fly ash in the four cities are similar to those in northern China cities (Bi *et al.*, 2007). However, the ratios in this study are higher when compared with the coal combustion source profile in Texas (Chow *et al.*, 2004).

Construction derived dust is caused by the construction activities and cement production factories. The four construction derived dust profiles (Table 7) obtained in this study were quite similar between each other. All were characterized by high abundances of Ca (32.69%–49.02%) followed by Si (7.11%–13.08%), Al (3.36%–6.07%) and TC, K, Fe. The most abundant Ca and Al were found in FSCD (construction derived dust of Fushun) profile, which contained approximately 49.02% calcium, 6.07% Al, by weight, respectively. HLDCD (construction derived dust of Huludao) profile contained the most abundant Si, with approximately 13.08% Si by weight. The PM₁₀ construction derived dust profiles obtained in this study are quite similar to the PM₁₀ cement profile reported by Wu *et al.* (2009).

Fe is the most abundant among species in SYISD (iron and steel manufacturing dust of Shenyang), ASISD (iron and steel manufacturing dust of Anshan) and FSISD (iron and steel manufacturing dust of Fushun) profiles with concentrations in the range of 25.16–35.34% followed by Ca, TC and OC with levels in the range of 5.91–14.16%,

Table 5. Profiles of soil dust in four cities for PM₁₀ (weight percent by mass)^a.

Species	SYSD	ASSD	FSSD	HLDSD
Na	1.16 ± 0.12	0.34 ± 0.25	1.09 ± 0.11	1.07 ± 0.12
Mg	0.60 ± 0.01	0.95 ± 0.05	0.75 ± 0.12	0.45 ± 0.12
Al	8.24 ± 0.08	8.29 ± 3.57	6.45 ± 2.24	10.71 ± 1.31
Si	27.81 ± 1.12	21.66 ± 3.82	-	28.20 ± 0.65
P	0.10 ± 0.04	-	-	-
K	1.60 ± 0.28	0.84 ± 0.43	2.39 ± 0.31	0.69 ± 0.11
Ca	2.05 ± 0.42	2.47 ± 0.57	1.63 ± 1.18	1.27 ± 0.58
Ti	0.97 ± 0.02	0.45 ± 0.21	0.47 ± 0.04	0.22 ± 0.08
V	0.00 ± 0.01	0.00 ± 0.01	0.01 ± 0.01	0.02 ± 0.01
Cr	0.02 ± 0.01	0.01 ± 0.01	0.01 ± 0.01	0.02 ± 0.01
Mn	0.04 ± 0.01	0.11 ± 0.03	0.09 ± 0.02	0.04 ± 0.01
Fe	2.03 ± 0.19	6.95 ± 1.07	3.74 ± 1.59	3.68 ± 0.24
Co	0.01 ± 0.01	0.00 ± 0.01	-	0.00 ± 0.01
Ni	0.01 ± 0.01	0.01 ± 0.01	0.00 ± 0.01	0.01 ± 0.01
Cu	0.01 ± 0.01	0.00 ± 0.01	0.01 ± 0.01	0.00 ± 0.01
Zn	0.02 ± 0.01	0.03 ± 0.01	0.02 ± 0.01	0.03 ± 0.03
Pb	0.00 ± 0.01	0.00 ± 0.01	0.00 ± 0.01	0.00 ± 0.01
As	-	0.00 ± 0.00	0.00 ± 0.00	-
Ba	0.05 ± 0.01	-	0.08 ± 0.01	-
Sr	-	-	0.03 ± 0.01	-
TC	4.38 ± 1.90	4.92 ± 0.40	4.12 ± 3.93	1.05 ± 0.50
OC	2.10 ± 1.02	2.72 ± 0.32	3.13 ± 2.62	0.76 ± 0.43
SO ₄ ²⁻	-	0.45 ± 0.05	-	0.01 ± 0.01

^a Hyphens refer to the unanalyzed species.

Table 6. Profiles of coal fly ash in four cities for PM₁₀ (weight percent by mass).

Species	SYCFA	ASCFA	FSCFA	HLDCFA
Na	0.22 ± 0.01	0.55 ± 0.31	0.38 ± 0.07	0.22 ± 0.01
Mg	0.29 ± 0.01	0.17 ± 0.16	0.47 ± 0.15	0.40 ± 0.32
Al	9.28 ± 0.84	8.59 ± 7.46	9.57 ± 7.96	14.82 ± 3.92
Si	12.36 ± 0.92	7.85 ± 6.52	-	17.99 ± 3.99
P	0.08 ± 0.06	-	-	-
K	0.56 ± 0.24	0.43 ± 0.39	1.26 ± 0.38	0.90 ± 0.25
Ca	2.78 ± 1.35	4.28 ± 2.91	1.05 ± 0.64	1.93 ± 0.85
Ti	0.68 ± 0.05	0.17 ± 0.13	0.83 ± 0.11	0.71 ± 0.04
V	0.01 ± 0.01	0.00 ± 0.01	0.03 ± 0.01	0.01 ± 0.01
Cr	0.01 ± 0.01	0.02 ± 0.01	0.02 ± 0.01	0.01 ± 0.01
Mn	0.03 ± 0.01	0.01 ± 0.01	0.07 ± 0.03	0.04 ± 0.02
Fe	1.73 ± 0.13	1.85 ± 1.25	6.79 ± 1.66	5.07 ± 2.72
Co	0.00 ± 0.01	0.00 ± 0.01	-	0.00 ± 0.01
Ni	0.00 ± 0.01	0.03 ± 0.02	0.01 ± 0.01	0.00 ± 0.01
Cu	0.01 ± 0.01	0.01 ± 0.01	0.01 ± 0.01	0.01 ± 0.01
Zn	0.03 ± 0.01	0.03 ± 0.02	0.02 ± 0.01	0.04 ± 0.02
Pb	0.01 ± 0.01	0.01 ± 0.01	0.00 ± 0.01	0.03 ± 0.04
As	-	0.00 ± 0.00	0.00 ± 0.00	-
Ba	0.03 ± 0.01	-	0.09 ± 0.03	-
Sr	-	-	0.05 ± 0.03	-
TC	27.70 ± 2.63	13.60 ± 9.94	14.43 ± 13.96	16.04 ± 9.10
OC	25.60 ± 3.99	8.10 ± 7.30	12.98 ± 1.31	15.05 ± 8.52
SO ₄ ²⁻	-	3.67 ± 0.65	-	0.72 ± 0.57

5.31–8.33%, 1.90–3.65%, respectively (Table 8). The highest concentration of Fe is in SYISD profile with level of 35.34%, while the highest abundance of Ca is in ASISD profile with a value of 14.16%. The Zn abundances differ substantially

among the three profiles, with 0.04% in ASISD profile, 0.31% in SYISD profile, and 2.08% in FSISD profile. The HLDZD (zinc dust of Huludao) highly enriched with Zn and Pb. HLDZD contained 63.18% Zn and 11.66% Pb. However,

the other elements in HLDZD profile are below 0.70%.

The vehicle exhaust (VE) profile reported by USEPA (Table 8) was adopted in the study. And the same chemical

profile for vehicle exhaust was used for CMB modeling, considering the similar composition among the vehicle exhaust profiles of the four cities.

Table 7. Profiles of construction derived dust in four cities for PM₁₀ (weight percent by mass).

Species	SYCD	ASCD	FSCD	HLDZD
Na	0.93 ± 0.08	0.79 ± 0.08	0.18 ± 0.08	0.32 ± 0.11
Mg	1.96 ± 0.35	1.91 ± 0.19	1.91 ± 0.67	3.00 ± 1.03
Al	4.18 ± 1.80	3.36 ± 0.34	6.07 ± 1.28	3.48 ± 0.43
Si	9.14 ± 0.50	7.11 ± 0.71	-	13.08 ± 1.93
P	0.02 ± 0.01	-	-	-
K	2.76 ± 0.55	1.05 ± 0.11	2.63 ± 4.64	1.11 ± 0.09
Ca	37.81 ± 6.65	35.35 ± 3.54	49.02 ± 8.68	32.69 ± 2.26
Ti	0.66 ± 0.32	0.41 ± 0.04	0.24 ± 0.06	0.11 ± 0.02
V	0.00 ± 0.01	0.00 ± 0.01	0.00 ± 0.01	0.00 ± 0.01
Cr	0.01 ± 0.01	0.00 ± 0.01	0.00 ± 0.01	0.01 ± 0.01
Mn	0.02 ± 0.01	0.06 ± 0.01	0.13 ± 0.08	0.02 ± 0.01
Fe	1.44 ± 0.19	2.60 ± 0.26	1.91 ± 0.39	0.97 ± 0.22
Co	0.00 ± 0.01	0.00 ± 0.01	-	0.00 ± 0.01
Ni	0.01 ± 0.00	0.00 ± 0.01	0.00 ± 0.01	0.00 ± 0.01
Cu	0.01 ± 0.00	0.00 ± 0.01	0.00 ± 0.01	0.00 ± 0.01
Zn	0.01 ± 0.01	0.02 ± 0.01	0.08 ± 0.03	0.00 ± 0.01
Pb	0.00 ± 0.01	0.01 ± 0.01	0.03 ± 0.05	0.00 ± 0.01
As	-	0.00 ± 0.00	0.00 ± 0.00	-
Ba	0.05 ± 0.02	-	0.04 ± 0.01	-
Sr	-	-	0.14 ± 0.06	-
TC	3.16 ± 0.21	3.47 ± 0.35	1.13 ± 1.24	3.21 ± 1.29
OC	0.75 ± 0.14	1.81 ± 0.18	0.20 ± 0.08	0.80 ± 0.40
SO ₄ ²⁻	-	2.90 ± 0.29	-	1.70 ± 0.28

Table 8. Profiles of iron and steel manufacturing dust/ zinc dust and vehicle exhaust in four cities for PM₁₀ (weight percent by mass).

Species	SYISD	ASISD	FSISD	HLDZD	VE ^a
Na	0.23 ± 0.01	0.34 ± 0.01	0.26 ± 0.11	0.00 ± 0.01	0.30 ± 0.28
Mg	3.10 ± 2.22	0.42 ± 0.12	1.65 ± 1.81	0.00 ± 0.01	0.22 ± 0.30
Al	1.32 ± 0.47	2.87 ± 1.28	1.73 ± 0.55	0.42 ± 0.03	0.27 ± 0.15
Si	3.05 ± 1.30	2.53 ± 0.42	-	0.57 ± 0.04	0.69 ± 0.48
P	0.05 ± 0.03	-	-	-	0.00 ± 0.01
K	0.80 ± 0.73	0.11 ± 0.15	0.95 ± 0.44	0.62 ± 0.04	0.23 ± 0.20
Ca	5.91 ± 3.87	14.16 ± 9.28	8.60 ± 3.45	0.23 ± 0.02	0.60 ± 0.76
Ti	0.09 ± 0.05	0.58 ± 0.57	0.06 ± 0.02	0.03 ± 0.01	0.10 ± 0.08
V	0.00 ± 0.01	0.00 ± 0.01	0.00 ± 0.01	0.01 ± 0.01	0.03 ± 0.02
Cr	0.09 ± 0.10	0.00 ± 0.01	0.74 ± 1.26	0.02 ± 0.01	0.01 ± 0.02
Mn	0.32 ± 0.27	0.04 ± 0.03	0.82 ± 1.12	0.02 ± 0.01	0.02 ± 0.02
Fe	35.34 ± 18.71	25.16 ± 20.27	31.81 ± 7.21	0.19 ± 0.01	1.18 ± 0.62
Co	0.00 ± 0.01	0.00 ± 0.01	-	0.00 ± 0.01	0.00 ± 0.01
Ni	0.03 ± 0.05	0.01 ± 0.01	0.14 ± 0.25	0.11 ± 0.01	0.01 ± 0.01
Cu	0.01 ± 0.02	0.00 ± 0.01	0.04 ± 0.05	0.25 ± 0.02	0.08 ± 0.02
Zn	0.31 ± 0.35	0.04 ± 0.04	2.08 ± 3.43	63.18 ± 4.29	0.22 ± 0.03
Pb	0.15 ± 0.18	0.00 ± 0.01	0.20 ± 0.32	11.66 ± 0.79	0.03 ± 0.01
As	-	0.00 ± 0.00	0.00 ± 0.00	-	0.00 ± 0.00
Ba	0.03 ± 0.03	-	0.02 ± 0.01	-	-
Sr	-	-	0.02 ± 0.00	-	-
TC	5.31 ± 4.51	5.85 ± 2.19	8.33 ± 9.69	0.00 ± 0.01	89.87 ± 8.99
OC	3.65 ± 3.85	3.04 ± 1.58	1.90 ± 1.38	0.00 ± 0.01	51.68 ± 5.68
SO ₄ ²⁻	-	3.16 ± 0.32	-	0.00 ± 0.01	3.87 ± 3.01

^aVE-vehicle exhaust.

Results of Source Apportionment

The average source contributions to ambient PM₁₀ are presented in Figs. 2–4. Soil dust and coal fly ash were found to be the major sources of PM₁₀ in all four cities as shown in Fig. 2. The soil dust was the largest contributor of PM₁₀ in Fushun and Huludao, with the concentration of 60.5 µg/m³ (37.5%) and 79.3 µg/m³ (28.8%), respectively (Fig. 2). The coal fly ash turned out to be the largest contributor of PM₁₀ in Shenyang and Anshan, with the concentration of 67.0 µg/m³ (35.6%) and 39.6 µg/m³ (24.9%), respectively. FSSD had the greatest annual average contribution (37.5%) in all four cities (Fig. 2). However, the highest annual average concentration of soil dust was found in Huludao (79.3 µg/m³). During non-heating period, concentration of HLSD (91.9 µg/m³) is much higher than that found in other three cities (Fig. 4). The higher soil dust concentration in Huludao during non-heating period can be attributed to more exposed lands and the higher wind speed during the sampling period (May) compared to other three cities. The sampling period of Huludao was during its windy period with the wind speed up to 4.18 m/s which was much higher than other three cities (2.7 m/s, Shenyang; 2.0 m/s, Anshan; 2.4 m/s, Fushun). SYCFA had greatest annual average concentration (67.0 µg/m³) in all four cities, and the corresponding contribution was 35.6% (Fig. 2). The coal fly ash contribution in Shenyang was much higher than that in Beijing (13.3%) (Wang *et al.*, 2008) and Kaifeng (21.0%)

(Wu *et al.*, 2009) but lower than that in Kolkata (47%) (Gupta *et al.*, 2007). The coal fly ash concentrations in the four cities are much higher in heating-period (Fig. 3) than non-heating period (Fig. 4) due to increasing coal combustion for heating and temperature inversion in winter. The coal fly ash concentration in heating period in the four cities was 2.2–8.1 times those calculated in non-heating period. The largest concentration of coal fly ash (116.0 µg/m³) was found in Anshan in heating period, and the corresponding contribution was 45.9% (Fig. 3).

The construction derived dust was the second largest contributor in Huludao, with a concentration of 57.4 µg/m³ (20.8%) (Fig. 2). It was much higher than that in Shenyang (11.2 µg/m³, 5.9%), Anshan (6.3 µg/m³, 4.0%) and Fushun (4.9 µg/m³, 3.0%). The higher construction derived dust concentration can be attributed to the abundance of construction activities in Huludao in the study period. The reason also may be that there is a big cement plant within the urban area in Huludao. Moreover, the big cement plant is located in northern Huludao, and the wind is blowing from north direction most of the time in winter. The construction derived dust concentration in the four cities was higher in non-heating period (Fig. 4) than heating period (Fig. 3). The probable reason was that the number of construction sites increased greatly in non-heating period, and the cold weather in heating period in northeastern China was unfavorable to construction activities.

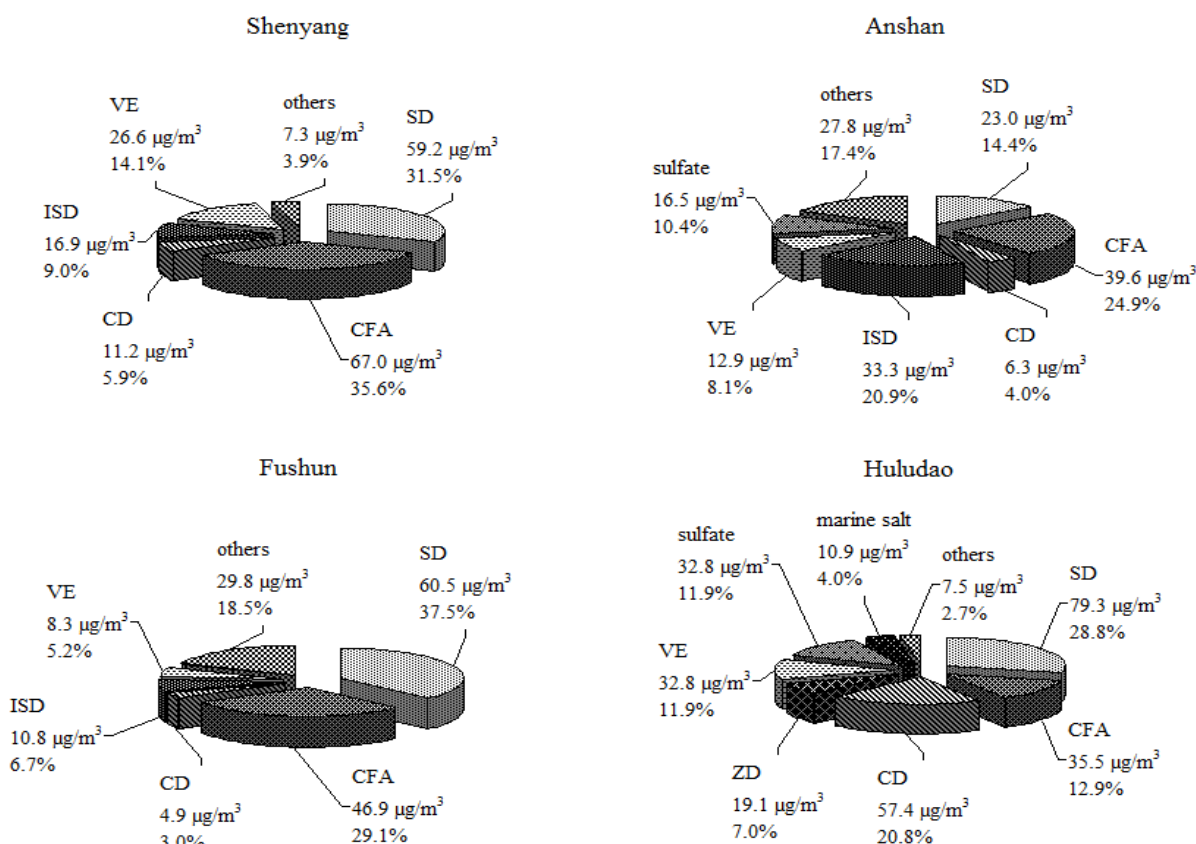


Fig. 2. Chemical mass balance source apportionment of ambient PM₁₀ in four cities of northeastern China (annual average result). SD-soil dust, CFA-coal fly ash, CD-construction derived dust, ISD-iron and steel manufacturing dust, ZD-zinc dust, VE-vehicle exhaust dust.

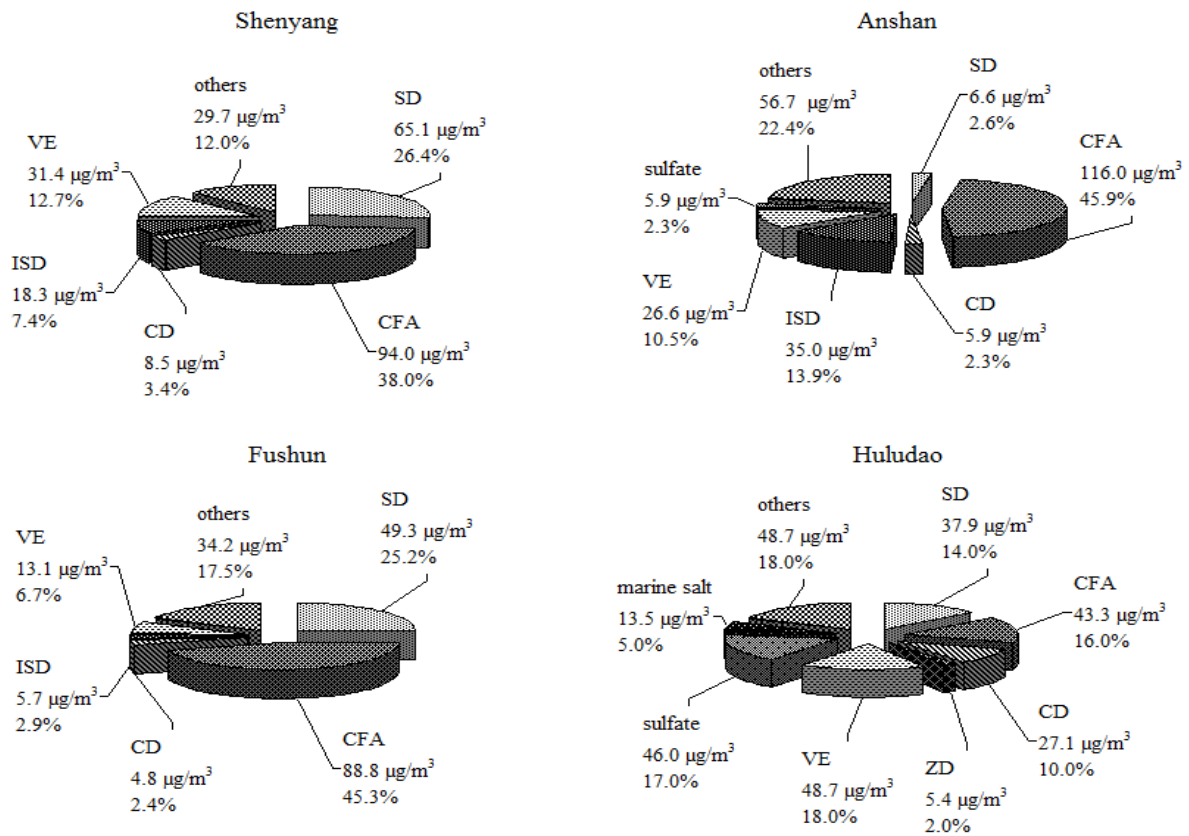


Fig. 3. Chemical mass balance source apportionment of ambient PM_{10} in four cities of northeastern China in heating period.

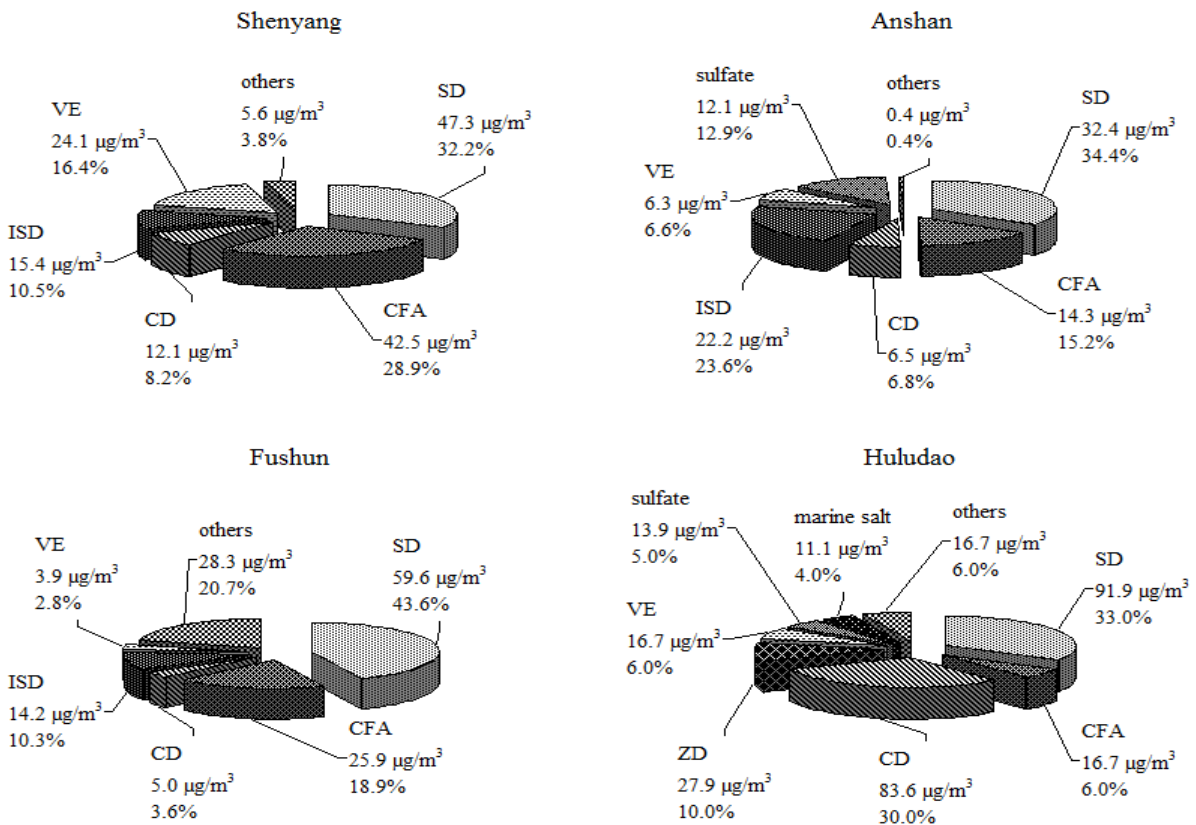


Fig. 4. Chemical mass balance source apportionment of ambient PM_{10} in four cities of northeastern China in non-heating period.

The zinc dust, a special PM source of Huludao, contributed $19.1 \mu\text{g}/\text{m}^3$ (7.0%) to PM_{10} (Fig. 2). Huludao Nonferrous Metals Group Corporation (HNMGC) is located in eastern Huludao, and discharge abundance of smoke dust containing zinc every year. The process of zinc smelting in HNMGC was the dominant source of zinc dust in Huludao atmosphere. Huludao is the only coastal city in the four cities, and marine salt as another special PM source in Huludao, contributed $10.9 \mu\text{g}/\text{m}^3$ (4.0%) to PM_{10} (Fig. 2) much lower than marine salt contribution to PM_{10} (18–23%) in California's central coast (Chow *et al.*, 1996). Vehicle exhaust and sulfate had the same contribution of $32.8 \mu\text{g}/\text{m}^3$ (11.9%) to PM_{10} in Huludao (Fig. 2).

Iron and steel manufacturing dust was the second largest contributor in Anshan, with a concentration of $33.3 \mu\text{g}/\text{m}^3$ (Fig. 2). It was higher than the soil dust concentration in the city ($23.0 \mu\text{g}/\text{m}^3$) and also much higher than iron and steel manufacturing dust concentration in Shenyang ($16.9 \mu\text{g}/\text{m}^3$) and Fushun ($10.8 \mu\text{g}/\text{m}^3$). The reason for the high concentration of ASISD might be the great portion of iron and steel industries and large amount of iron and steel manufacturing dust emission in the city. Under the control of northwestern flow in winter (heating period), the pollutants from Anshan Iron and Steel Corporation (one of the biggest steel makers in China, the biggest atmospheric pollution source in Anshan), which is located in the northwest of Anshan would make the air of the whole urban area more polluted. Therefore, concentration of ASISD was higher in heating period ($35.0 \mu\text{g}/\text{m}^3$) (Fig. 3) than non-heating period ($22.2 \mu\text{g}/\text{m}^3$) (Fig. 4). The iron and steel manufacturing dust contribution to PM_{10} in Anshan (20.9%) was slightly higher than that found in Panzhihua (19.5%) (Xue, 2010). And it was 13.3, 4.2 and 1.7 times that observed in Wuxi (Han *et al.*, 2009), Detroit (Gildemeister *et al.*, 2007) and Taiyuan (Zeng, 2010).

CONCLUSIONS

In this study, ambient PM_{10} measurements were obtained for the urban areas of four cities in northeastern China. Chemical source profiles were conducted for particulate emissions from several urban, industrial, and geological sources. Quantificational contributions of the sources conducted by CMB showed that soil dust and coal fly ash were the major sources of PM_{10} in all four cities. The coal fly ash concentrations in the four cities are much higher in heating-period than non-heating period due to enhanced usage amount of coal for heating. The construction derived dust also was the major source of PM_{10} in Huludao with a concentration of $57.4 \mu\text{g}/\text{m}^3$ (20.8%). The zinc dust was a special PM source of Huludao with a contribution of $19.1 \mu\text{g}/\text{m}^3$ (7.0%) to PM_{10} , which mainly emitted from the process of zinc smelting in HNMGC. In addition, the iron and steel manufacturing dust was also the major source of PM_{10} in Anshan, and its contribution to the ambient PM_{10} was much higher than the results in previous studies, which might be caused by the great amount of iron and steel manufacturing dust emission in Anshan.

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