

## CONTENTS

**Aquatic environment**

- Speciation of organic phosphorus in a sediment profile of Lake Taihu I: Chemical forms and their transformation  
Di Xu, Shiming Ding, Bin Li, Xiuling Bai, Chengxin Fan, Chaosheng Zhang ..... 637
- Flow field and dissolved oxygen distributions in the outer channel of the Orbal oxidation ditch by monitor and CFD simulation  
Xuesong Guo, Xin Zhou, Qiuwen Chen, Junxin Liu ..... 645
- Removal of Cu(II) from acidic electroplating effluent by biochars generated from crop straws  
Xuejiao Tong, Renkou Xu ..... 652
- Optimisation of chemical purification conditions for direct application of solid metal salt coagulants:  
Treatment of peatland-derived diffuse runoff  
Elisangela Heiderscheidt, Jaakko Saukkoriipi, Anna-Kaisa Ronkanen, Bjørn Kløve ..... 659
- Removal of nitrogen from wastewater with perennial ryegrass/artificial aquatic mats biofilm combined system  
Chongjun Chen, Rui Zhang, Liang Wang, Weixiang Wu, Yingxu Chen ..... 670
- Microbial community characterization, activity analysis and purifying efficiency in a biofilter process  
Hong Xiang, Xiwu Lu, Lihong Yin, Fei Yang, Guangcan Zhu, Wuping Liu ..... 677
- Performance of a completely autotrophic nitrogen removal over nitrite process for treating wastewater with different substrates at ambient temperature  
Xiaoyan Chang, Dong Li, Yuhai Liang, Zhuo Yang, Shaoming Cui, Tao Liu, Huiping Zeng, Jie Zhang ..... 688
- Performance study and kinetic modeling of hybrid bioreactor for treatment of bi-substrate mixture of phenol-*m*-cresol in wastewater: Process optimization with response surface methodology  
Sudipta Dey, Somnath Mukherjee ..... 698
- Analysis of aerobic granular sludge formation based on grey system theory  
Cuiya Zhang, Hanmin Zhang ..... 710
- Ethyl thiosemicarbazide intercalated organophilic calcined hydrotalcite as a potential sorbent for the removal of uranium(VI) and thorium(IV) ions from aqueous solutions  
T. S. Anirudhan, S. Jalajamony ..... 717

**Atmospheric environment**

- Observed levels and trends of gaseous SO<sub>2</sub> and HNO<sub>3</sub> at Mt. Waliguan, China: Results from 1997 to 2009  
Weili Lin, Xiaobin Xu, Xiaolan Yu, Xiaochun Zhang, Jianqing Huang ..... 726
- Influence of SO<sub>2</sub> in incineration flue gas on the sequestration of CO<sub>2</sub> by municipal solid waste incinerator fly ash  
Jianguo Jiang, Sicong Tian, Chang Zhang ..... 735
- Seasonal variation and source apportionment of organic and inorganic compounds in PM<sub>2.5</sub> and PM<sub>10</sub> particulates in Beijing, China  
Xingru Li, Yuesi Wang, Xueqing Guo, Yingfeng Wang ..... 741
- Emissions of particulate matter and associated polycyclic aromatic hydrocarbons from agricultural diesel engine fueled with degummed, deacidified mixed crude palm oil blends  
Khamphe Phoungthong, Surajit Tekasakul, Perapong Tekasakul, Gumpon Prateepchaikul, Naret Jindapetch, Masami Furuuchi, Mitsuhiko Hata ..... 751
- Ground-high altitude joint detection of ozone and nitrogen oxides in urban areas of Beijing  
Pengfei Chen, Qiang Zhang, Jiannong Quan, Yang Gao, Delong Zhao, Junwang Meng ..... 758

**Environmental biology**

- Characterization of *Methylocystis* strain JTA1 isolated from aged refuse and its tolerance to chloroform  
Tiantao Zhao, Lijie Zhang, Yunru Zhang, Zhilin Xing, Xuya Peng ..... 770
- Allelopathic effects of gallic acid from *Aegiceras corniculatum* on *Cyclotella caspia*  
Yu Liu, Fei Li, Qixin Huang ..... 776

**Environmental health and toxicology**

Toxicity detection of sodium nitrite, borax and aluminum potassium sulfate using electrochemical method

Dengbin Yu, Daming Yong, Shaojun Dong ..... 785

**Environmental catalysis and materials**A comparative study of Mn/CeO<sub>2</sub>, Mn/ZrO<sub>2</sub> and Mn/Ce-ZrO<sub>2</sub> for low temperature selective catalytic reduction of NO with NH<sub>3</sub> in the presence of SO<sub>2</sub> and H<sub>2</sub>O (**Cover story**)

Boxiong Shen, Xiaopeng Zhang, Hongqing Ma, Yan Yao, Ting Liu ..... 791

Removal of benzotriazole by heterogeneous photoelectro-Fenton like process using ZnFe<sub>2</sub>O<sub>4</sub> nanoparticles as catalyst

Junfeng Wu, Wenhong Pu, Changzhu Yang, Man Zhang, Jingdong Zhang ..... 801

Metal loaded zeolite adsorbents for hydrogen cyanide removal

Ping Ning, Juan Qiu, Xueqian Wang, Wei Liu, Wei Chen ..... 808

Preparation and evaluation of Zr-β-FeOOH for efficient arsenic removal

Xiaofei Sun, Chun Hu, Jiuhui Qu ..... 815

Application of red mud as a basic catalyst for biodiesel production

Qiang Liu, Ruirui Xin, Chengcheng Li, Chunli Xu, Jun Yang ..... 823

Amino-functionalized core-shell magnetic mesoporous composite microspheres for Pb(II) and Cd(II) removal

Yulin Tang, Song Liang, Juntao Wang, Shuili Yu, Yilong Wang ..... 830

Electrochemical detection and degradation of ibuprofen from water on multi-walled carbon nanotubes-epoxy composite electrode

Sorina Motoc, Adriana Remes, Aniela Pop, Florica Manea, Joop Schoonman ..... 838

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## Seasonal variation and source apportionment of organic and inorganic compounds in PM<sub>2.5</sub> and PM<sub>10</sub> particulates in Beijing, China

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### Abstract

The distribution and source of the solvent-extractable organic and inorganic components in PM<sub>2.5</sub> (aerodynamics equivalent diameter below 2.5 microns), and PM<sub>10</sub> (aerodynamics equivalent diameter below 10 microns) fractions of airborne particles were studied weekly from September 2006 to August 2007 in Beijing. The extracted organic and inorganic compounds identified in both particle size ranges consisted of *n*-alkanes, PAHs (polycyclic aromatic hydrocarbons), fatty acids and water soluble ions. The potential emission sources of these organic compounds were reconciled by combining the values of *n*-alkane carbon preference index (CPI), %waxC<sub>n</sub>, selected diagnostic ratios of PAHs and principal component analysis in both size ranges. The mean cumulative concentrations of *n*-alkanes reached 1128.65 ng/m<sup>3</sup> in Beijing, 74% of which (i.e., 831.7 ng/m<sup>3</sup>) was in the PM<sub>2.5</sub> fraction, PAHs reached 136.45 ng/m<sup>3</sup> (113.44 ng/m<sup>3</sup> or 83% in PM<sub>2.5</sub>), and fatty acids reached 436.99 ng/m<sup>3</sup> (324.41 ng/m<sup>3</sup> or 74% in PM<sub>2.5</sub>), which resulted in overall enrichment in the fine particles. The average concentrations of SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup> were 21.3 ± 15.2, 6.1 ± 1.8, 12.5 ± 6.1 μg/m<sup>3</sup> in PM<sub>2.5</sub>, and 25.8 ± 15.5, 8.9 ± 2.6, 16.9 ± 9.5 μg/m<sup>3</sup> in PM<sub>10</sub>, respectively. These three secondary ions primarily existed as ammonium sulfate ((NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>), ammonium bisulfate (NH<sub>4</sub>HSO<sub>4</sub>) and ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>). The characteristic ratios of PAHs revealed that the primary sources of PAHs were coal combustion, followed by gasoline combustion. The ratios of stearic/palmitic acid indicated the major contribution of vehicle emissions to fatty acids in airborne particles. The major alkane sources were biogenic sources and fossil fuel combustion. The major sources of PAHs were vehicular emission and coal combustion.

**Key words:** airborne particles; organic compounds; size distribution; source

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### Introduction

Recently, a great deal of attention has been paid to air pollution in urban settings, especially atmospheric particulates, because they play important roles in many environmental processes, including indirect and direct effects on climate (Admas et al., 2001; Jacobson, 2001; Martin et al., 2004), various biogeochemical cycles, the formation of photochemical smog and haze, and visibility degradation by scattering and absorption of electromagnetic radiation in visible wavelengths (Elias et al., 2009). Atmospheric particulates can include a complex mixture of inorganic substances (metal ions, sulfate, ammonium and nitrate) and hundreds of different organic substances. In general, inorganic species comprise 25%–50% of the aerosol mass, predominantly in the form of sulfates, ammonium salts

and nitrates (Gray et al., 1986). Organic compounds are the main contributors to fine aerosols in the atmosphere of highly industrialized urban areas (Xu et al., 2005; Hou et al., 2006; Rushdi et al., 2006). In Beijing, approximately 40% of the fine aerosol mass is composed of organic material (He et al., 2001; Dan et al., 2004). Many of these organic materials, including PAHs and PCBs, are carcinogenic and mutagenic (Perera et al., 2005; Tsai et al., 2001; Wu et al., 2005). Atmospheric particles with these toxic characteristics can readily penetrate into the lungs and, therefore, are more likely to have short-term and long-term adverse effects, including worsening of respiratory symptoms and the development of diseases (André, 2005; Zheng et al., 2000).

The main sources of atmospheric particles are natural and anthropogenic sources such as fossil fuel combustion, industrial processes and non-industrial fugitive sources (Li et al., 2006; Pio et al., 2001; Duan et al., 2010). In recent

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years, automobile exhaust fumes in urban areas have been important contributors to air pollution in Beijing (Hao et al., 2001; Hao and Wang, 2005; Westerdahl et al., 2009). The total vehicle population in Beijing has almost tripled in the last 10 years to more than 3 million vehicles and the increasing trend toward private vehicle ownership is expected to continue (Liu et al., 2007).

In this study, aerosol samples were collected from September 2006 to August 2007 in Beijing and analyzed for their levels of *n*-alkanes, PAHs, fatty acids and water soluble ions. The aims of this study were to understand the aerosol composition and to characterize the seasonal variations in aerosol composition. The sources of these compounds were also discussed.

## 1 Materials and methods

### 1.1 Site description and aerosol sampling

Aerosol samples were collected with quartz fiber filters (Whatman Company, UK) for 24 hr for each sample using a high-volume air sampler (Anderson, USA) for PM<sub>2.5</sub> and PM<sub>10</sub> weekly from September 2006 to August 2007, on the rooftop of a two-story building in the courtyard of the Institute of Atmospheric Physics (39°58'N, 116°22'E). Before sampling, the quartz fiber filters were wrapped in aluminum foil and pre-heated to 500°C for 4 hr to remove all organic material and conditioned in a constant humidity desiccator (temperature: 25°C; humidity: 50%) for 24 hr and weighed. After sampling, the filters were returned to the desiccator (temperature: 25°C; humidity: 50%) for a minimum of 24 hr. After being weighed, the filters were stored in a freezer (−20°C) until subsequent analysis.

### 1.2 Chemical analysis and quality control

#### 1.2.1 Organic compounds

The analytical procedure used for organic compounds was described previously (Zheng et al., 2000). Briefly, a quarter of each filter was ultrasonically extracted three times, 20 min each time, using 25 mL dichloromethane (DCM) (HPLC grade, Supelco, USA). The extracts were combined and filtered through a glass fiber filter and then concentrated to approximately 5 mL under reduced pressure at 35°C by a rotary evaporator (Buchi, Sweden), followed by drying in a gentle nitrogen stream. Then, 10 mL *n*-hexane was added to the tube and the extracts were dried again in a gentle nitrogen stream. This extraction was transferred into a silica chromatography column (30 cm × 1 cm diameter) filled with 5 g silica gel (80–100 mesh, activated at 180°C for 12 hr) and 5 g alundum (100–200 mesh, activated at 250°C for approximately 12 hr) to separate aliphatic hydrocarbons and PAHs. The column was sequentially eluted with 70 mL *n*-hexane and 70 mL of a mixture of hexane and DCM (1:1, V/V). The two eluted fractions were evaporated using a rotary

evaporator, and the final volume was adjusted to 1 mL under a gentle nitrogen stream for gas chromatography-mass spectrometry (GC-MS) analysis.

Another quarter of each filter was ultrasonically extracted three times 20 min each with 25 mL dichloromethane and extracted twice with ultrapure water 20 min each time. The extracts were combined, filtered with a glass fiber filter, concentrated to 1 mL in a rotary evaporator under reduced pressure, and dried almost to completion under a gentle nitrogen stream. A solution of 14% BF<sub>3</sub> in methanol (approximately 0.5 mL) was added to the reactor, which was then sealed with Teflon tape. The samples were reacted at 100°C for 30 min to convert carboxyl groups to methyl ester groups. Then, 3 mL of pure water and 0.3 mL of acetonitrile were added to the reaction products, and the mixture was extracted with 5 mL of *n*-hexane. The *n*-hexane layer was washed twice with pure water and concentrated to 1 mL in *n*-hexane for GC-MS analysis. The purities of DCM, *n*-hexane, methanol, and acetonitrile exceeded 99.8%. The trace contaminants did not interfere with the identification or quantification of the compounds of interest.

The hydrocarbons, PAHs and acid derivatives were identified and quantified with a trace GC-MS spectrometer (Thermo DSQ Finnigan, USA) and an HP-5MS capillary column (30-m-long, 0.25-mm-diameter, 0.25-μm-film thickness). High-purity helium was used as the carrier gas at a constant flow rate of 1.0 mL/min. A total of 1 μL of sample was injected into the GC in splitless mode. For the aliphatic hydrocarbons and PAH fractions, the GC temperature was programmed as follows: starting at 50°C (5 min), increased to 180°C at 150°C/min and held for 5 min, then ramped to 280°C at 5°C/min and held at 280°C for 15 min. For fatty acid methyl ester analysis, the temperature was held at 50°C for 5 min, ramped to 280°C at 5°C/min and held at 280°C for 20 min. The mass spectrometer was operated in electron impact (EI) mode at 70 eV and fully scanned, ranging from 50 to 550 amu. The identification of organic compounds was performed using *m/z* mass chromatography, and the mass spectra were compared with standards from the National Institute of Standards and Technology 2005.

All analytical procedures were monitored using strict quality assurance and control measures. Lab blanks, field blanks and solvent blanks were used to determine background contamination; they showed no detectable PAHs. The purities of DCM, *n*-hexane, methanol, and acetonitrile exceeded 99.8%. Phthalate esters were the main contaminants found in the blanks. These trace contaminants did not interfere with the identification or quantification of the compounds of interest. The proposed method showed satisfactory precision and accuracy. Due to its volatility, most naphthalene was distributed mainly in the gas phase; however, naphthalene was not considered in this study.

### 1.2.2 Water-soluble ions

The analysis of water-soluble ions in aerosol samples was carried out by an Ion Chromatography System (ICS-1600, Dionex, USA) at the State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry, Institute of Atmospheric Physics, Chinese Academy of Sciences, following the procedures in Gao and Zhao (2008). Briefly, a quarter of the mixed cellulose ester filter was cut and put into an acid-cleaned test tube, and ultrasonically extracted for about 0.5 hr with 50 mL deionized water (conductivity: 18.2 MΩ·cm). The extract was filtered before ion chromatography (ICS-1100, Dionex, USA) analysis. An AS11 analytical column (2 × 250 mm<sup>2</sup>, Dionex) was employed for the determination of selected anions (chloride, nitrite nitrate, and sulfate). Selected cations (sodium, potassium, ammonium, magnesium and calcium) were determined by the same IC system with a CS12A analytical column (2 × 250 mm<sup>2</sup>, Dionex), using methane sulfuric acid (MSA) (EGC II MSA, Dionex) as the eluent. The lowest detection limit for all the above ions was 0.3 μg/m<sup>3</sup>.

## 2 Results and discussion

Summary data concerning the total concentrations of organic pollutants (C<sub>18</sub>–C<sub>35</sub> *n*-alkanes, 16 PAHs, and 25 fatty acids) and water soluble inorganic ions determined in the sampling periods are presented in **Table 1**. The seasons were defined based on the temperatures and the fuel consumption characteristics. December to February was considered to be winter. June to August was defined to be summer; September to November to be fall; and March to May to be spring. Higher concentrations were found in winter than in the other seasons. This finding may be explained by the fact that in winter, lower ambient temperature, decreased rainfall and stable atmosphere lead to easier conversion from the gas phase to the particle phase and allow concentrations to build up in a shallow

boundary layer.

### 2.1 *n*-Alkanes

A total of 18 *n*-alkanes (ranging from C<sub>18</sub> to C<sub>35</sub>) were detected. The total *n*-alkane concentrations ranged from 106.2 to 4368.4 ng/m<sup>3</sup> in PM<sub>10</sub> and from 70.9 to 2750.3 ng/m<sup>3</sup> in PM<sub>2.5</sub>. The total concentrations of *n*-alkanes in Beijing were higher than those found in the other Chinese cities of Qingdao and Guangzhou (Guo et al., 2003b). These alkanes were mainly enriched in the PM<sub>2.5</sub> samples (about 74%). For C<sub>18</sub>–C<sub>23</sub>, about 86% was found in PM<sub>2.5</sub> particles and 69% of C<sub>24</sub>–C<sub>33</sub> was found in PM<sub>2.5</sub> particles, indicating that lower carbon number alkanes were readily enriched in fine particles.

**Figure 1** shows the seasonal distribution diagrams of the individual alkanes, with no odd-even carbon number predominance in the C<sub>18</sub>–C<sub>23</sub> range and a slight odd carbon number preference in the C<sub>24</sub>–C<sub>33</sub> range, which was characteristic of higher plant wax resources. The carbon preference index (CPI), which considers the relative abundance of compounds containing odd and even numbers of carbon atoms, is used to distinguish the two main sources of *n*-alkanes, biogenic and anthropogenic activities (e.g., fossil fuel and biomass combustion) (de Azevedo et al., 1999; Cheng et al., 2006; Feng et al., 2006; Rogge et al., 1993). The carbon number profiles of *n*-alkanes derived from natural sources (primarily cuticular waxes of higher plants) show high CPI values (greater than 3), whereas a CPI value of approximately 1 implies anthropogenic influences (fossil fuel combustion) (Schneider et al., 1983; Simoneit, 1984). The mean CPI were 1.28, 1.25, 1.25, and 1.04 in PM<sub>2.5</sub> and 1.43, 1.11, 1.32, and 1.14 in PM<sub>10</sub> for spring, summer, autumn and winter, respectively. These findings indicate a dominance of anthropogenic *n*-alkanes from automobile exhaust and coal combustion for heating (Kalaitzoglou et al., 2004).

The wax results (**Table 1**) indicate that the mean contributions of biogenic wax *n*-alkanes were 25.9, 39.9, 34.8,

**Table 1** Total concentration ranges and molecular diagnostic parameters of *n*-alkane, PAHs, and fatty acids in PM<sub>2.5</sub> and PM<sub>10</sub> particles

	Spring		Summer		Autumn		Winter	
	PM <sub>2.5</sub>	PM <sub>10</sub>	PM <sub>2.5</sub>	PM <sub>10</sub>	PM <sub>2.5</sub>	PM <sub>10</sub>	PM <sub>2.5</sub>	PM <sub>10</sub>
Σ <i>n</i> -Alkane (ng/m <sup>3</sup> )	851.7	1072.3	543.9	703.0	631.8	971.0	1299.7	1734.7
C <sub>max</sub>	25	27	25	25	27	29	23	25
CPI <sup>a</sup>	1.28	1.43	1.25	1.11	1.25	1.32	1.04	1.14
%Wax <sup>b</sup>	20.3	25.9	29.3	39.9	28.3	34.8	16.0	21.2
ΣPAHs (ng/m <sup>3</sup> )	52.7	73.5	30.3	45.0	50.1	66.1	278.6	315.2
ΣFatty acids (ng/m <sup>3</sup> )	431.7	625.1	194.4	199.9	207.8	263.0	463.6	659.8
Oleic acid/stearic acid	0.16	0.15	0.11	0.11	0.26	0.26	0.32	0.33
ΣWater soluble ions (μg/m <sup>3</sup> )	35.4	59.3	52.9	82.4	49.1	74.2	64.1	87.8
SO <sub>4</sub> <sup>2-</sup> (μg/m <sup>3</sup> )	10.8	16.4	12.6	16.3	18.2	22.0	43.7	48.8
NO <sub>3</sub> <sup>-</sup> (μg/m <sup>3</sup> )	4.8	6.4	5.3	8.4	8.7	12.5	5.3	8.5
NH <sub>4</sub> <sup>+</sup> (μg/m <sup>3</sup> )	11.7	15.9	21.4	30.5	9.6	12.8	7.4	8.6

<sup>a</sup> CPI: carbon preference index, alkane:  $CPI = \frac{\sum(\text{odd carbon number alkanes})}{\sum(\text{even carbon number alkanes})}$

<sup>b</sup> %Wax: percentage of leaf wax-organic compounds in the total fraction of organic compounds,

%wax:  $C_n = \sum(C_n - 0.5(C_{n-1} + C_{n+1})) / \sum C_n\text{-alkanes} \times 100\%$ . Negative values of  $C_n$  were taken as zero (Bi et al., 2005).

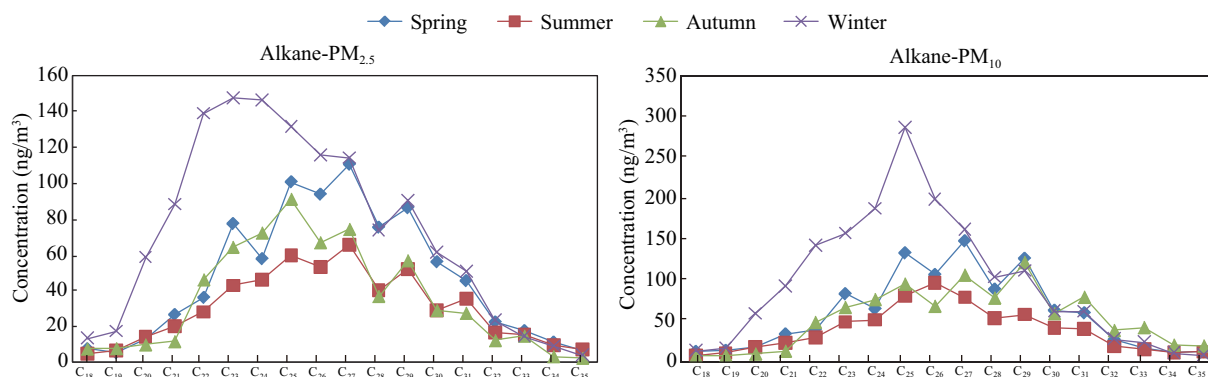


Fig. 1 Typical seasonal distribution diagrams of *n*-alkanes in different size particles.

and 21.2 in  $PM_{10}$  and 20.3, 29.3, 28.3, and 16.0 in  $PM_{2.5}$  for spring, summer, autumn and winter, respectively. Plants flourish in the summer and autumn seasons, and when winter arrives, almost of the plants are brown and withered. The *n*-alkane contributions from biogenic sources were relatively smaller in the winter than in the summer and autumn. These results also indicate the significant contribution of fossil fuel combustion in the winter to air pollution in Beijing.

## 2.2 PAHs

A total of 16 PAHs were quantified including naphthalene (NA); acenaphthylene (ACY); acenaphthene (ACE); fluorene (FL); phenanthrene (PHEN); anthracene (AN); fluoranthene (FIUR); pyrene (PY); benzo[a]anthracene (BaA); chrysene (ChRY); benzo[b] fluoranthene (BbF); benzo[k]fluoranthene (BkF); benzo[a]pyrene (BaP); indeno[1,2,3-cd]pyrene (IcdP); dibenzo[a,h]anthracene (dBAh); and benzo[ghi]perylene (Bpe), with total concentrations of 22.4–1014.3  $ng/m^3$  in  $PM_{10}$  and 9.2–923.2  $ng/m^3$  in  $PM_{2.5}$  (Fig. 2). These PAHs were mainly enriched in the  $PM_{2.5}$  samples (about 83%). Compared with other cities, the total concentrations of PAHs in this study were higher than those in Hong Kong (Bi et al.,

2005; Guo et al., 2003a), Houston (Fraser et al., 2002), Rome (Menichini et al., 1999), Nanjing (Wang et al., 2006), and Qingdao (Guo et al., 2003b), but were lower than those in Tianjin (Wu et al., 2005) and New Delhi (Sharma et al., 2007), indicating that the air pollution from PAHs in Beijing is very high. Similar to the alkanes, a clear seasonal pattern was found for PAHs, with much higher concentrations in the winter, followed by the autumn and spring seasons; the lowest concentrations were found in the summer. These results were consistent with those reported by other authors (Guo et al., 2003b; Pietrogrande et al., 2011; Pindado et al., 2009). The percentage of individual ring PAHs differed between the  $PM_{10}$  and  $PM_{2.5}$  samples. For example, in the spring, the main components had 5 rings in  $PM_{10}$ , while in  $PM_{2.5}$ ; they had 4 rings. Components having four or more fused aromatic rings contributed most strongly to the total concentrations of PAHs (above 87%).

In general, most PAHs are of anthropogenic origin, and there are two main sources of PAHs: mobile (vehicular) and stationary (residential heating and power plants) (Zheng et al., 2000). The molecular composition profiles and characteristic ratios of PAHs have been used to distinguish among different sources (Guo et al., 2003a;

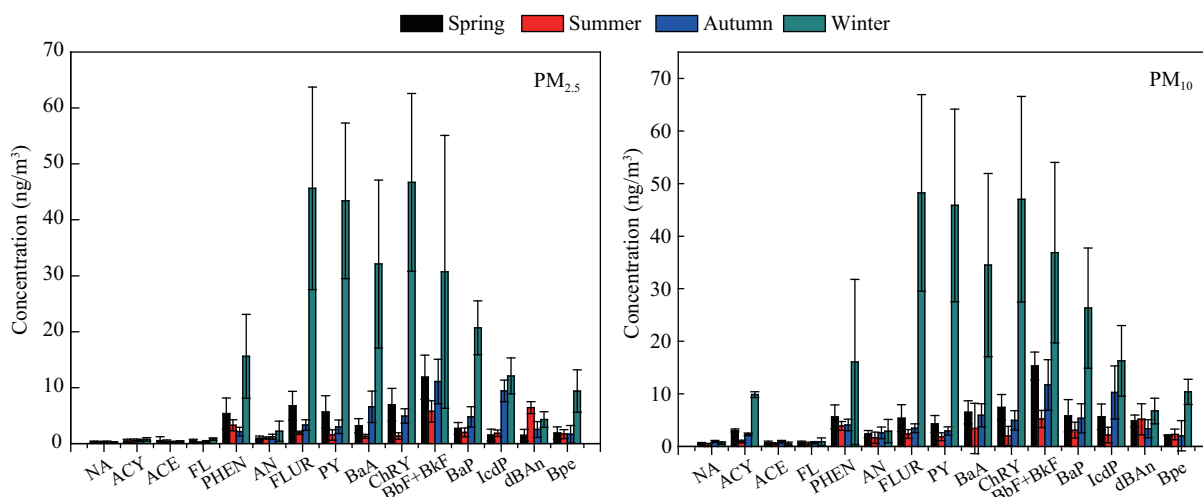
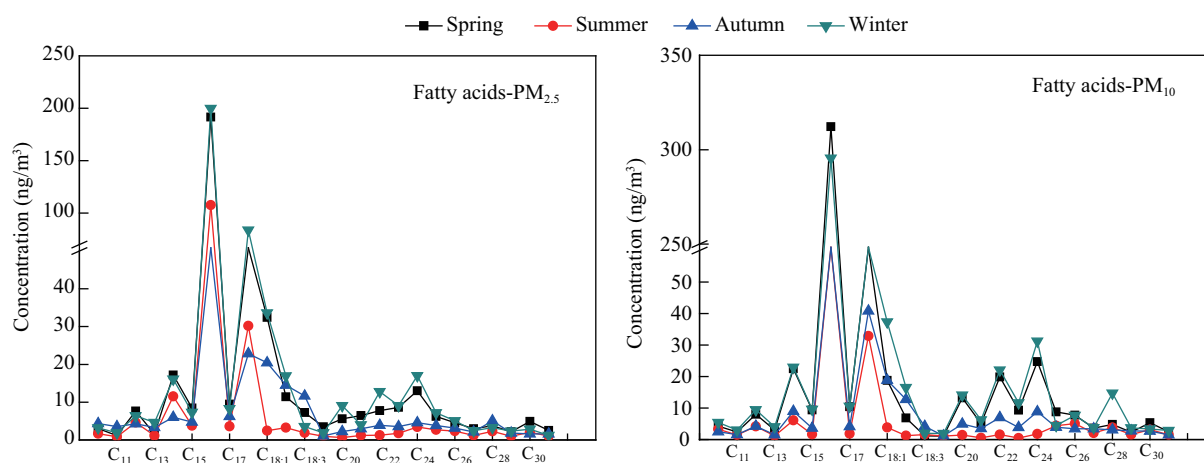


Fig. 2 Seasonal average concentrations of 16 PAHs in atmospheric aerosols in Beijing.

Ladji et al., 2009b). The characteristic ratios of individual PAHs from earlier studies and the average ratios of these individual PAHs in the aerosols collected in Beijing are listed in **Table 2**. The average ratios of AN/(AN+PHEN) for all seasons were above 0.1, indicating that burning of fossil fuels was the major source of PAHs in Beijing. The average ratios of BaP/Bpe were close to those for coal combustion emissions in the autumn and winter, indicating that the discharge gas of coal combustion had seriously polluted the atmosphere in Beijing, while in the summer, the average ratios were close to those for gasoline or diesel combustion. The ratios of BaA/ChRY indicated that the combustion of biomass or coal might be a primary source of PAHs. Ratios of FLUR/(PY+FLUR) that are close to 0.4, 0.6–0.7, and > 0.7 are characteristic of petroleum, exhaust emission, and combustion of grass, wood or coal, respectively (Yunker et al., 2002). In this study, the ratios of FLUR/(PY+FL) were in the range of 0.52–0.56 (mean: 0.55), indicating that with the rapid increase in the numbers of private cars, the level of exhaust emissions has increased dramatically in Beijing. Through the above analysis, we concluded that the primary source of PAHs in aerosols was coal combustion, followed by gasoline combustion.

### 2.3 Fatty acids

A total of 25 individual fatty acids (C<sub>10</sub>–C<sub>31</sub>) were identified and quantified in the PM<sub>2.5</sub> and PM<sub>10</sub> samples of Beijing, including three unsaturated fatty acids (oleic acid (C<sub>18:1</sub>), linoleic acid (C<sub>18:2</sub>) and eleostearic acid (C<sub>18:3</sub>)) (**Fig. 3**). The major compound classes included fatty acids, with total concentrations of 45.5–992.1 ng/m<sup>3</sup> for PM<sub>2.5</sub>, and 61.7–1652.3 ng/m<sup>3</sup> for PM<sub>10</sub>. **Figure 3** indicates that fatty acids were mostly enriched in PM<sub>2.5</sub>, especially in the summer (90%). Palmitic acid (C<sub>16</sub>) was the most abundant acid, followed by stearic acid (C<sub>18</sub>). A predominance of acids with even-numbered carbons of less than 20 carbons was found, which is considered to be derived in part from microbial activities and cooking meat (Oliveira et al., 2007; Rogge et al., 1991; Simoneit, 1986), while homologs > 22 carbons come from vascular plant wax (Feng et al., 2006; Simoneit and Mazurek, 1982). In addition to saturated fatty acids, three unsaturated fatty acids, oleic acid (C<sub>18:1</sub>), linoleic acid (C<sub>18:2</sub>), and eleostearic acid (C<sub>18:3</sub>), were also abundant in Beijing aerosol samples. Linoleic acid and oleic acid are unstable and can be rapidly degraded in the atmosphere (Kawamura and Kaplan,



**Fig. 3** Average concentrations of individual fatty acids in PM<sub>10</sub> and PM<sub>2.5</sub>.

**Table 2** Value of molecular diagnostic ratios of PAHs for source identification

		An/(An+PHEN)	BaP/Bpe	BaA/ChRY	FLUY/(FL+PYR)
PM <sub>2.5</sub>	Spring	0.15	0.91	0.43	0.54
	Summer	0.25	0.30	0.97	0.56
	Autumn	0.41	0.80	1.24	0.56
	Winter	0.18	1.23	1.27	0.54
PM <sub>10</sub>	Spring	0.31	0.62	1.12	0.56
	Summer	0.28	0.64	1.08	0.56
	Autumn	0.38	0.82	1.38	0.54
	Winter	0.19	0.75	1.73	0.52
Coal combustion		–	0.7–6.6	1.0–1.2	–
Petroleum pollution		< 0.1	–	–	0.4
Gasoline combustion		–	0.3–0.4	0.28–1.2	0.6–0.7
Diesel combustion		> 0.1	0.46–0.81	0.17–0.36	–
Biomass burning		–	–	0.93	> 0.7
Dust		–	–	–	–

–: No practical importance.



1987). Thus, the oleic acid to stearic acid ratio is often used as an indicator of aerosol aging. In  $PM_{10}$  samples, the average ratios were 0.16, 0.11, 0.26, and 0.32, and in  $PM_{2.5}$  samples, the average ratios were 0.15, 0.11, 0.26, and 0.33 in the spring, summer, autumn, and winter, respectively. The ratio of  $C_{18:0}/C_{16}$  has been used as a qualitative tool for source assessment (Ladji et al., 2009a; Oliveira et al., 2007; Zhao et al., 2006). Ratios lower than 0.25, 0.25–

0.5, and 0.5–1.0 can be attributed to foliar vegetation combustion and wood smoke, car and diesel truck exhaust, and paved and unpaved road dust and charbroiling, respectively (Rogge et al., 2006). In this study, the ratios were 0.38, 0.29, 0.37, and 0.39 for  $PM_{10}$  and 0.36, 0.28, 0.33, and 0.42 for  $PM_{2.5}$  in the spring, summer, autumn and winter, respectively, indicating the strong contribution of vehicle emissions to the concentrations of fatty acids in

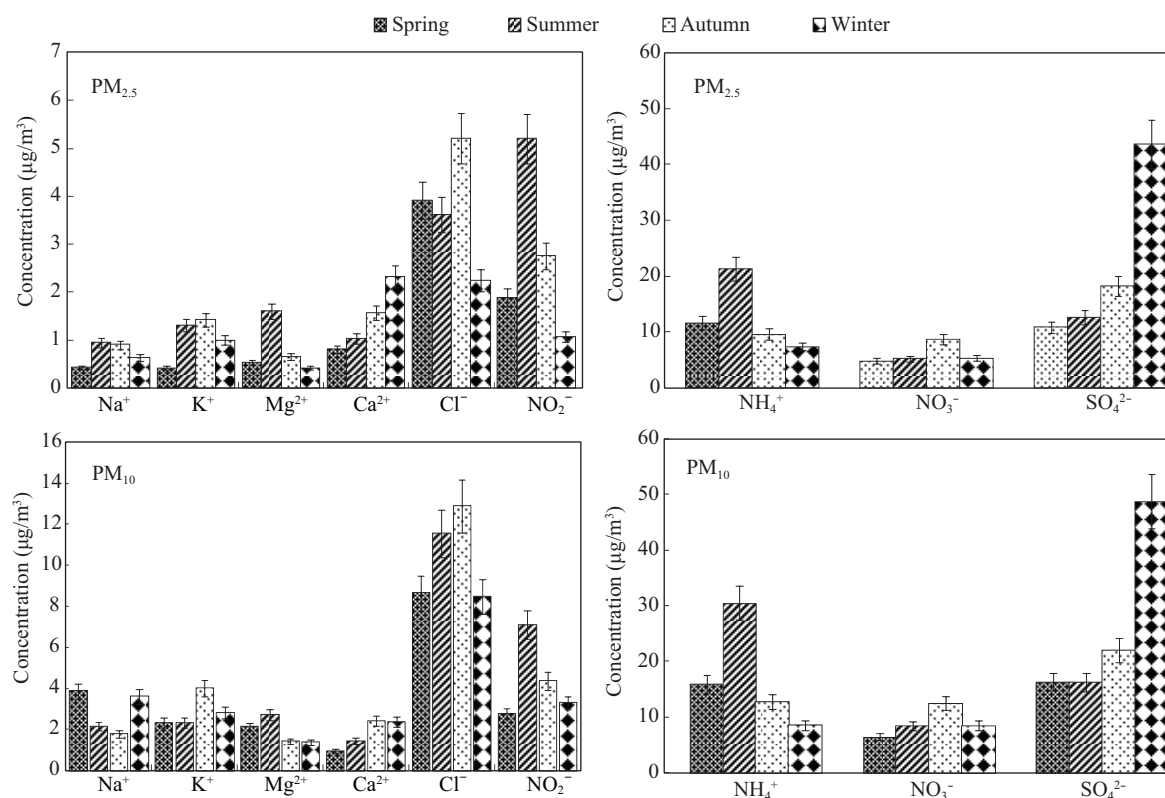


Fig. 4 Average concentrations of individual water soluble ion in  $PM_{10}$  and  $PM_{2.5}$ .

Table 3 Factors analysis of main *n*-alkanes and PAHs

Compound	Factor 1		Factor 2		Compound	Factor 1		Factor 2	
	$PM_{2.5}$	$PM_{10}$	$PM_{2.5}$	$PM_{10}$		$PM_{2.5}$	$PM_{10}$	$PM_{2.5}$	$PM_{10}$
$C_{17}$			0.641	0.517	FL	0.635		0.496	0.917
$C_{18}$			0.828	0.749	PHEN	0.838	0.665	0.333	0.677
$C_{19}$			0.929	0.912	AN	0.367		0.669	0.909
$C_{20}$			0.916	0.945	FLUR	0.924	0.834		0.507
$C_{21}$			0.912	0.990	PY	0.933	0.836		0.507
$C_{22}$	0.427	0.519	0.859	0.950	BaA	0.908	0.883	0.338	0.440
$C_{23}$	0.482	0.640	0.820	0.907	ChRY	0.963	0.895		0.413
$C_{24}$	0.537	0.887	0.770		BbF + BkF	0.916	0.906	0.344	
$C_{25}$	0.648	0.918	0.692		BaP	0.674	0.887	0.768	0.448
$C_{26}$	0.811	0.963	0.470		dBA	0.707	0.639	0.476	
$C_{27}$	0.885	0.974	0.414		IcdP	0.873	0.716	0.625	-0.349
$C_{28}$	0.904	0.948			Bpe	0.903	0.891		0.351
$C_{29}$	0.912	0.928							
$C_{30}$	0.956	0.892							
$C_{31}$	0.932	0.732							
$C_{32}$	0.929	0.601							
Eigen value	7.0	7.7	6.7	6.4		8.8	8.7	0.963	1.56
Variance (%)	68.8	52.3	18.1	29.6		59.3	73.2	22.1	13.0
Cumulative (%)	43.8	48.0	42.0	43.8		59.3	56.9	21.1	20.0
Possible sources	Biogenic source		Fossil fuel combustion			Vehicular emission		Coal combustion	

atmospheric particles.

## 2.4 Water-soluble inorganic ions

In this study, nine major water-soluble inorganic ions were quantified (Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, Ca<sup>2+</sup>, Cl<sup>-</sup>, NO<sub>2</sub><sup>-</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>), the result for the main ions is listed in Fig. 4. The secondary inorganic ions of SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup> were the major components of water soluble ions, accounting for 78% and 63% of the total water soluble ions in PM<sub>2.5</sub> and in PM<sub>10</sub>, and the average concentrations of SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup> were (21.3 ± 15.2), (6.1 ± 1.8), (12.5 ± 6.1) μg/m<sup>3</sup> in PM<sub>2.5</sub>, and (25.8 ± 15.5), (8.9 ± 2.6), (16.9 ± 9.5) μg/m<sup>3</sup> in PM<sub>10</sub>, respectively. The highest concentration of SO<sub>4</sub><sup>2-</sup> was found in winter. The increased use of fuel coal for heating was one of the important reasons for the enhanced SO<sub>4</sub><sup>2-</sup> concentration during this period. Nitrate in autumn was higher than that in the other seasons. The NH<sub>4</sub><sup>+</sup> concentration was highest in summer. We calculated linear regression correlations between different chemical species in PM<sub>2.5</sub> and PM<sub>10</sub>. In this study, NH<sub>4</sub><sup>+</sup>-NO<sub>3</sub><sup>-</sup> concentrations presented a strong correlation not only in PM<sub>2.5</sub> ([NO<sub>3</sub><sup>-</sup>] = 0.43[NH<sub>4</sub><sup>+</sup>] + 1.82, R<sup>2</sup> = 0.83) but also in PM<sub>10</sub> ([NO<sub>3</sub><sup>-</sup>] = 0.41[NH<sub>4</sub><sup>+</sup>] + 0.001, R<sup>2</sup> = 0.73). NH<sub>4</sub><sup>+</sup> and SO<sub>4</sub><sup>2-</sup> concentrations showed a weak correlation both in PM<sub>2.5</sub> (R<sup>2</sup> = 0.50, slope = 1.64) and in PM<sub>10</sub> (R<sup>2</sup> = 0.39, slope = 0.39), suggesting that these three major ions primarily existed as ammonium sulfate ((NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>), ammonium bisulfate (NH<sub>4</sub>HSO<sub>4</sub>) and ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>).

## 2.5 Source apportionment by principal component analysis

Source apportionment for PM<sub>2.5</sub> and PM<sub>10</sub> constituents were identified by means of Principal Component Factor Analysis (PCA). PCA was executed by the varimax rotated factor matrix method, based on the orthogonal rotation criterion which maximizes the variance of the squared elements in the column of a factor matrix, using statistical software (SPSS v19). Factor loadings indicate the correlation of each pollutant species with each component and were related to the source emission composition (Almeida et al., 2006). To examine the suitability of these data for factor analysis, Kaiser-Meyer-Olkin (KMO) and Bartlett's tests were performed. KMO is a measure of sampling adequacy that indicates the proportion of variance which is common variance. A high value (close to 1) generally indicates that factor analysis may be useful. If the KMO test value is less than 0.5, factor analysis will not be useful (Parinet et al., 2004). In this study, KMO < 0.5 for water-soluble ions and fatty acids, indicating that there are significant relationships among variables. The factor analysis results of alkanes and PAHs in coarse and fine particulates in Beijing are presented in Table 3. Two factors were sufficient to explain most of the data variance. For alkanes, factor 1, which explained 43.8% and 42.0% of the

total variance with Eigen Value of 7.0 and 6.7 for fine and coarse particulate respectively, represented emissions from biogenic sources such as vegetative detritus and cooking, with strong loadings for carbon numbers C<sub>17-27</sub>. Factor 2 explained 48.0% and 43.0% of the total variance, with Eigen Value of 7.7 and 6.4 for fine and coarse particulates, and respectively represented emissions from combustion sources such as fossil fuel emissions and biomass burning, with strong loadings for carbon numbers C<sub>28-32</sub>.

For PAHs, factor 1 was responsible for 59.3% and 73.2% of the total variance, with Eigen Value of 8.8 and 8.7 for fine and coarse particulates respectively. This factor was associated with vehicular emission, with strong loading of PHEN, FLUR, PY, BaA, ChRY, BbF, BkF, dBAn, IcdP, and Bpe. The secondary factor, which explained 21.1% and 20.0% of the total variance for fine and coarse particulates with high loading of FL, AN, BaP and IcdP, represented coal combustion.

## 3 Conclusions

Samples of PM<sub>2.5</sub> and PM<sub>10</sub> aerosols were analyzed to study the compositions of solvent-extractable organic compounds, including *n*-alkanes, PAHs, and organic acids and water-soluble ions from September 2006 to August 2007. The total concentrations of *n*-alkanes and PAHs showed strong seasonal variation trends, with higher concentrations in the winter than in other seasons, and higher concentrations were found in winter or autumn. Fatty acid concentrations had weak seasonal variation trends. These organic compounds were mainly enriched in PM<sub>2.5</sub> samples. Source identification analyses indicated that the most significant source of *n*-alkanes was fossil fuel combustion, and the most significant source of PAHs in aerosols was coal combustion, followed by gasoline combustion. The main source of fatty acids was vehicle emissions. The major alkane sources were biogenic sources and fossil fuel combustion, as identified through PCA analysis. The major sources of PAHs were vehicular emission and coal combustion.

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