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Progress and prospects of atmospheric environmental sciences in China

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Compositions and sources of organic acids in fine particles (PM_{2.5}) over the Pearl River Delta region, south China

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ABSTRACT

Organic acids as important constituents of organic aerosols not only influence the aerosols' hygroscopic property, but also enhance the formation of new particles and secondary organic aerosols. This study reported organic acids including C14-C32 fatty acids, C4-C9 dicarboxylic acids and aromatic acids in PM_{2.5} collected during winter 2009 at six typical urban, suburban and rural sites in the Pearl River Delta region. Averaged concentrations of C14-C32 fatty acids, aromatic acids and C4-C₉ dicarboxylic acids were 157, 72.5 and 50.7 ng/m³, respectively. They totally accounted for 1.7% of measured organic carbon. C₂₀-C₃₂ fatty acids mainly deriving from higher plant wax showed the highest concentration at the upwind rural site with more vegetation around, while C_{14} - C_{18} fatty acids were more abundant at urban and suburban sites, and dicarboxylic acids and aromatic acids except 1,4-phthalic acid peaked at the downwind rural site. Succinic and azelaic acid were the most abundant among C4-C9 dicarboxylic acids, and 1,2-phthalic and 1,4-phthalic acid were dominant aromatic acids. Dicarboxylic acids and aromatic acids exhibited significant mutual correlations except for 1,4-phthalic acid, which was probably primarily emitted from combustion of solid wastes containing polyethylene terephthalate plastics. Spatial patterns and correlations with typical source tracers suggested that C14-C32 fatty acids were mainly primary while dicarboxylic and aromatic acids were largely secondary. Principal component analysis resolved six sources including biomass burning, natural higher plant wax, two mixed anthropogenic and two secondary sources; further multiple linear regression revealed their contributions to individual organic acids. It turned out that more than 70% of $C_{14}-C_{18}$ fatty acids were attributed to anthropogenic sources, about 50%–85% of the $C_{20}-C_{32}$ fatty acids were attributed to natural sources, 80%-95% of dicarboxylic acids and 1,2-phthalic acid were secondary in contrast with that 81% of 1,4-phthalic acid was primary.

Introduction

Atmospheric aerosols negatively impact human health (Pope et al., 2002), dramatically reduce visual range (Hobbs, 2002) and strongly influence radioactive forcing and climate (Penner et al., 1998). They are highly var-

ied in their spatiotemporal distribution and their health and climate effects are largely attributed to their sizes and/or chemical compositions (Dusek et al., 2006; Pope et al., 2002; Xu et al., 2008), therefore it is necessary to chemically characterize aerosols in different regions due to increasing concern about aerosols' role in environmental health and climate change. As the fraction of atmospheric particulates closely related to health endpoints than the larger ones (Pope et al., 2002), fine particles or particles

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with dynamic diameter less than 2.5 μ m (PM_{2.5}) have been regulated in many nations for the protection of human health. Organic matters are major aerosol constituents in both urban and rural areas (Alves et al., 2012; Wang et al., 2012b), and organic acids, including low molecular weight acids (e.g. oxalic acid) and high molecular weight acids (e.g. fatty acids), are important atmospheric oxygenated organic aerosols in PM2.5 (Rogge et al., 1993; Wang et al., 2006). These acids are either directly emitted from various natural and anthropogenic sources (Ho et al., 2010; Oliveira et al., 2007) or secondarily formed from oxidation of gas-phase precursors followed by gas/particle partition (Wang et al., 2012a). Organic acids are involved in a series of atmospheric chemical reactions occurring in the gas, water and particle phases (Chebbi and Carlier, 1996); they can reduce the surface tension of particles to form cloud condensation nuclei (Facchini et al., 1999), and laboratory experiments also indicate that nucleation of sulfuric acid is considerably enhanced in the presence of organic acids like aromatic acids (Zhang et al., 2004). As recently reported by Chan and Chan (2011), the presence of oleic acid in particles would enhance the reactive uptake of nonanal. Therefore, investigation of organic acids in particles would be helpful in understanding atmospheric chemistry of organics and formation mechanism of secondary organic aerosols and cloud condensation nuclei. Since organic acids in the atmosphere are a pool of compounds with varying structures and sources/origins, their chemical speciation and source apportionment remain to be a big challenge.

Organic acids in urban areas are even more complicated in terms of their primary and secondary contributions, and in their biogenic and anthropogenic sources. In the Pearl River Delta (PRD) region, one of the most industrialized and densely populated regions in Guangdong Province, China, organic acids contributed substantially to particlebound solvent-extractable organics (Feng et al., 2006; Ma et al., 2010; Zheng et al., 2000). Ho et al. (2007) and Wang et al. (2006) investigated seasonal and spatial variation of organic acids in Chinese cities, and found organic acids exhibited higher concentrations in southern China, especially in Guangzhou, a central city in the PRD region. Yet at the moment, information about sources and origins of organic acids in the PRD region is quite limited (Ho et al., 2011). As cooking can directly emit organic acids like fatty acids (He et al., 2004; Schauer et al., 1999, 2002; Zhao et al., 2007a, 2007b), cooking might be an important source of organic acids in the densely populated PRD region with about 40 thousand restaurants in Guangzhou (Guangzhou Statistical Yearbook, 2011); vehicle exhausts, another contributor to organic acids (Fraser et al., 1998; He et al., 2006, 2008), are supposed to be another important source with over 30% annual growth rate of private cars in the PRD region in recent years (Guangdong Statistical Yearbook, 2011). Biogenic emissions, on the other hand,

can give rise to organic acids directly through emission of terrestrial higher plant wax (Simoneit, 1986) or indirectly through oxidation of biogenic volatile organic compounds (VOCs) (Ding et al., 2011; Hu et al., 2008). This biogenic source is expected to be much more significant in the tropical/subtropical PRD region with relatively high annual mean temperature (about 25°C) and more evergreen vegetation (Ho et al., 2011). Secondary organic aerosols including secondary acids should also be important components of fine particles in the PRD region (Ding et al., 2012) as a result of the higher anthropogenic and biogenic precursor emissions (Zhang et al., 2012; Zheng et al., 2009), elevated atmospheric oxidative capacity (Hofzumahaus et al., 2009) and strong aerosols acidity (Ding et al., 2011) in the region. Considering the complex situation in the PRD region, it is an interesting topic to further explore the contributions of organic acids from different sources in addition to investigate their chemical compositions.

In the PRD region, aerosol pollution is typically much more serious in winter largely due to lower mixing height, less wet removal and prevailing winds from polluted area. In Guangzhou, the study by Ma et al. (2010) revealed much higher free organic acids in total suspended particulates (TSP) in winter than in summer. In the present study, we collected $PM_{2.5}$ samples simultaneously at six representative sites in the PRD region during winter 2009, and analyzed these samples for organic acids including fatty acids, dicarboxylic acids and aromatic acids. The purpose of this study is to characterize chemical compositions and to identify and apportion sources for organic acids in this highly industrialized and densely populated subtropical region in south China.

1 Experimental

1.1 Field sampling

The PRD region, with a population of about 56 million and a land area of about 43,000 km², is an economically developed region in south China and it contributes about one tenth of China's national GDP. In the present study, six sampling sites, including two urban sites (GZ and GD), two suburban sites (HD and PY) and two rural sites (KD and WQS), are selected for filter-based sampling of PM_{2.5}. These sites are also among the air quality monitoring stations established by local governmental environmental monitoring center. The locations of these sites are shown in **Fig. 1**.

The two urban sites GZ and GD, about 1.5 km away from each other, are located in densely populated commercial/residential districts of urban Guangzhou City with much heavier traffic around. The site PY is a suburban site in the south of urban Guangzhou with an industrial area about 1 km away in the east. HD is another suburban site



Fig. 1 Sampling sites in the Pearl River Delta (PRD) region, China.

in the northwest of urban Guangzhou. It is close to a forest park but there are also handicraft and machinery industries nearby. KD is a rural site in the north of PRD region without any significant emission sources nearby but with more forests and vegetation in the neighborhood, while WQS is a regional background site in the south of urban Guangzhou and about 50 km away from the city clusters of Guangzhou, Shenzhen, Foshan and Dongguan in the PRD region.

Twenty four hours filter-based PM2.5 samples were collected simultaneously at the six sites from November 28 to December 23, 2009, using medium-volume samplers at a flow rate of 300 L/min. The samplers were placed on the rooftops, 10-20 m above grounds. The quartz filters (5 \times 8 inch; Whatman) were baked at 450°C for at least 4 hours before sampling to reduce potential organic contamination. After weighing, these filters were wrapped with prebaked aluminum foils and stored in zipped Teflon bags when transported to the field. After sampling, the filters were again wrapped with prebaked aluminum foils and put in zipped Teflon bags, and then transported back to laboratory and stored at -4°C till analysis. Samples were only collected on sunny days and totally 140 valid samples were collected in this study. There was a field blank at each site during the campaign.

1.2 Laboratory analysis

Organic carbon (OC) and elemental carbon (EC) were measured on a punch $(1.5 \times 1.0 \text{ cm}^2)$ from each filter by the thermo-optical transmittance method (NIOSH 1999) with an OC/EC analyzer (Sunset Laboratory Inc., USA).

Detailed analysis of organic compounds has been described elsewhere (Ding et al., 2011). Briefly, 1/8 of each filter was extracted twice by sonication with 30 mL hexane/dichloride methane (DCM) (1:1, V/V) each time and then twice with 30 mL DCM/methanol (1:1, V/V) each time. Prior to solvent extraction, 400 µL mixing internal standards including *n*-tetracosane-D₅₀, palmitic acid-D₃₁, lauric acid-D₂₃, phthalic acid-D₄, and levoglucosan-¹³C₆ were spiked into the samples. The extracts of each sample were combined, filtered and concentrated to 2 mL. Then each sample was divided into two aliquots. One part was blown to dryness under a gentle stream of nitrogen, and kept at room temperature for one hour to derivatize carboxylic acids to methyl esters after adding 200 μ L of DCM, 10 μ L of methanol and 300 μ L of fresh prepared diazomethane. The methylated extract was analyzed for organic acids and 1,3,5-triphenylebenzene. Another part of sample was blown to dryness for silylation with 100 μ L pyridine and 200 μ L N,O-bis-(trimethylsilyl)-trifluoroacetamide plus 1% trimethylchlorosilane in an oven at 70°C for 1 hr. The silylated extract was analyzed for levoglucosan.

Samples were analyzed by an Agilent 5975N gas chromatography/mass selective detector (GC-MSD) in the scan mode with a HP-5 MS capillary column (30 m length \times $0.25 \text{ mm i.d.} \times 0.25 \text{ µm filmthickness}$). Splitless injection of 1 µL sample was performed. The GC temperature was initiated at 65°C (held for 2 min) and increased to 290°C at a rate of 5°C/min then held for 20 min. Compounds were identified according to their mass spectra and retention times. Quantification were accomplished by GC-MSD workstation after calibration curves were obtained with authentic standards included C14-C32 fatty acids, C4-C9 dicarboxylic acids and aromatic acids except 1,4-phthalic acids. Due to lacking of authentic standards, 1,4-phthalic acid was quantified using 1,2-phthalic acid as an alternative standard. The method detection limits ranged from 0.02 ng/m³ (1,4-phthalic acid) to 0.33 ng/m³ (octacosanoic acid), when calculated with the average sampling volume of 432 m³. Recoveries of the target compounds were over 70%. The field and laboratory blanks were extracted and analyzed in the same way as the field samples. Target compounds were not detected except for palmitic acid and stearic acid. Concentrations of organic acids were reported with their blanks corrected.

2 Results and discussion

2.1 Levels and compositions

Observed concentrations of fatty acids, dicarboxylic acids and aromatic acids in the PM_{2.5} samples together with OC and EC contents are presented in **Table 1**. The concentrations of OC ranged from 4.35 to 59.9 μ g/m³ with an average value of 17.9 μ g/m³ while EC ranged from 0.92 to 16.5 μ g/m³ with an average value of 4.95 μ g/m³, quite approximate to those previously reported in the PRD region, such as 14.7 μ g/m³ for OC and 6.1 μ g/m³ for EC in winter 2002 (Cao et al., 2003). Detected 40 organic acids totaled 82.6–756 ng/m³ with an average value of 280 ng/m³, and accounted for 0.66%–3.69% of OC with an avTable 1Concentrations of fatty acids, dicarboxylic acids and aromatic acids measured at the six sites in the PRD region over the period Nov28-Dec 23, 2009 (unit: ng/m^3)

| Compounds | GZ | GD | РҮ | HD | KD | WQS |
|---|--------------------------------|------------------------------------|-----------------------------------|--------------------------------|-----------------------------------|------------------------------------|
| | I. Fatty acids | | | | | |
| | Saturated | | | | | |
| Myristic acid (C14:0) | 5.99 ± 2.62 | 5.07 ± 2.15 | 5.18 ± 2.58 | 5.53 ± 2.06 | 3.57 ± 1.34 | 4.22 ± 1.17 |
| Pentadecanoic acid (C15:0) | 2.77 ± 1.23 | 2.58 ± 1.30 | 2.51 ± 1.08 | 2.44 ± 1.08 | 2.25 ± 0.91 | 2.23 ± 0.65 |
| Palmitic acid (C16:0) | 107 ± 48.2 | 87.8 ± 38.7 | 78.4 ± 52.7 | 122 ± 50.9 | 54.6 ± 27.3 | 58.3 ± 22.3 |
| Margaric acid (C17:0) | 1.97 ± 0.89 | 1.84 ± 0.89 | 1.63 ± 1.10 | 2.08 ± 1.10 | 1.55 ± 0.78 | 1.14 ± 0.69 |
| Stearic acid (C18:0) | 35.7 ± 15.2 | 28.7 ± 13.7 | 26.7 ± 19.2 | 38.7 ± 22.8 | 17.7 ± 9.72 | 15.1 ± 9.53 |
| Nonadecanoic acid (C19:0) | 0.61 ± 0.27 | 0.61 ± 0.30 | 0.50 ± 0.34 | 0.63 ± 0.33 | 0.66 ± 0.38 | 0.52 ± 0.26 |
| Arachidic acid (C20:0) | 3.58 ± 1.75 | 3.03 ± 1.45 | 2.44 ± 1.55 | 3.57 ± 1.99 | 3.64 ± 2.12 | 2.96 ± 1.35 |
| Heneicosanoic acid (C21:0) | 1.06 ± 0.50 | 1.06 ± 0.55 | 0.83 ± 0.63 | 1.13 ± 0.64 | 1.28 ± 0.81 | 0.99 ± 0.50 |
| Behenic acid (C22:0) | 5.08 ± 2.44 | 4.61 ± 2.23 | 3.71 ± 2.33 | 5.46 ± 3.06 | 5.91 ± 3.31 | 4.21 ± 2.43 |
| Tricosanoic acid (C23:0) | 2.58 ± 1.42 | 2.49 ± 1.42 | 1.87 ± 1.26 | 2.95 ± 1.92 | 3.42 ± 2.34 | 2.50 ± 1.51 |
| Carnaubic acid (C24:0) | 6.21 ± 3.19 | 5.88 ± 3.14 | 4.39 ± 2.67 | 7.19 ± 4.23 | 8.21 ± 5.00 | 5.96 ± 3.35 |
| Pentacosanoic acid (C25:0) | 1.35 ± 0.73 | 1.37 ± 0.81 | 0.96 ± 0.67 | 1.58 ± 1.06 | 1.74 ± 1.25 | 1.01 ± 0.67 |
| Cerotic acid (C26:0) | 3.11 ± 1.57 | 3.23 ± 1.76 | 2.24 ± 1.57 | 3.76 ± 2.28 | 4.39 ± 2.79 | 2.73 ± 1.71 |
| Heptacosanoic acid (C27:0) | 0.63 ± 0.35 | 0.67 ± 0.41 | 0.43 ± 0.35 | 0.74 ± 0.48 | 0.73 ± 0.51 | 0.53 ± 0.44 |
| Octacosanoic acid (C28:0) | 2.83 ± 1.55 | 3.10 ± 1.84 | 1.99 ± 1.35 | 3.56 ± 2.30 | 3.88 ± 2.67 | 2.31 ± 1.84 |
| Nonacosanoic acid (C29:0) | 0.58 ± 0.38 | 0.64 ± 0.45 | 0.37 ± 0.28 | 0.69 ± 0.52 | 0.66 ± 0.55 | 0.42 ± 0.35 |
| Triacontanoic acid (C30:0) | 2.67 ± 1.76 | 3.02 ± 2.11 | 1.70 ± 1.21 | 3.52 ± 2.51 | 3.64 ± 2.92 | 2.01 ± 1.93 |
| Henriacontanoic acid (C31:0) | 0.36 ± 0.24 | 0.42 ± 0.29 | 0.20 ± 0.16 | 0.45 ± 0.34 | 0.44 ± 0.38 | 0.28 ± 0.22 |
| Lacceroic acid (C32:0) | 2.00 ± 1.42 | 2.25 ± 1.70 | 1.18 ± 0.80 | 2.82 ± 2.18 | 2.79 ± 2.48 | 1.40 ± 1.22 |
| Subtotal | 185 ± 80.5 | 158 ± 70.3 | 137 ± 89.0 | 209 ± 88.3 | 121 ± 62.6 | 106 ± 45.9 |
| Palmitoleic acid (C16:1) | 0.11 ± 0.14 | 0.05 ± 0.03 | 0.03 ± 0.03 | 0.08 ± 0.08 | 0.28 ± 0.23 | 0.18 ± 0.20 |
| Oleic acid (C18:1) | 0.11 ± 0.14 | 1.81 ± 1.23 | 0.03 ± 0.03 | 1.05 ± 1.52 | 0.28 ± 0.23 | 0.13 ± 0.20 0.61 ± 0.28 |
| Lipoleic acid (C18:2) | 4.03 ± 0.89 1 74 ± 2.60 | 1.81 ± 1.23 0.48 ± 0.28 | 1.22 ± 0.93 0.27 ± 0.19 | 1.05 ± 1.52 0.40 ± 0.38 | 1.02 ± 2.42 0.43 ± 0.45 | 0.01 ± 0.28 0.10 ± 0.14 |
| Subtotal | 1.74 ± 2.09 6.46 ± 0.51 | 0.48 ± 0.28 2.33 ± 1.50 | 0.27 ± 0.19 | 0.40 ± 0.50 1.53 ± 1.02 | 0.43 ± 0.43 | 0.10 ± 0.14 0.84 ± 0.45 |
| Total fatty acids | 192 + 865 | 2.35 ± 1.50 161 + 71 3 | 1.31 ± 1.14 130 ± 80.4 | 1.55 ± 1.92 210 + 89 1 | 1.31 ± 0.90 122 ± 63.0 | 107 ± 461 |
| Total fatty actus | I Dicarboxylic acids | 101 ± /1.5 | 157 ± 07.4 | 210 ± 09.1 | 122 ± 00.0 | 107 ± 40.1 |
| Succinic acid (di-C4) | 17.8 ± 7.23 | 338 + 115 | 25.7 ± 9.31 | 23.7 ± 14.0 | 237+725 | 50.1 + 30.5 |
| Glutaric acid (di-C5) | 2.94 ± 1.17 | 5.0 ± 11.0 5.26 ± 1.84 | 3.88 ± 1.19 | 3.26 ± 2.03 | 3.38 ± 1.13 | 6.03 ± 3.99 |
| Adipic acid (di-C6) | 1.41 ± 0.56 | 2.37 ± 1.09 | 1.84 ± 0.48 | 1.87 ± 0.79 | 1.73 ± 0.47 | 3.52 + 2.94 |
| Pimelic acid (di-C7) | 0.71 ± 0.38 | 1.26 ± 0.74 | 0.66 ± 0.24 | 0.69 ± 0.32 | 0.74 ± 0.30 | 1.09 ± 1.13 |
| Suberic acid (di-C8) | 1.68 ± 0.64 | 2.19 ± 1.12 | 1.82 ± 0.73 | 1.92 ± 0.68 | 2.06 ± 0.67 | 4.26 + 3.31 |
| Azelaic acid (di-C9) | 10.6 ± 4.81 | 13.9 ± 8.21 | 10.5 ± 5.66 | 11.8 ± 5.42 | 10.1 ± 4.11 | 21.6 ± 19.6 |
| Subtotal | 35.2 ± 13.7 | 58.8 ± 21.1 | 44.4 ± 16.0 | 43.3 ± 20.7 | 41.7 ± 12.7 | 85.7 ± 58.1 |
| | III. Aromatic acids | | | | | |
| Benzoic acid | 1.97 ± 1.03 | 2.28 ± 1.19 | 2.04 ± 1.04 | 1.72 ± 0.87 | 1.90 ± 0.96 | 5.28 ± 2.93 |
| m-Toluic acid | 0.25 ± 0.11 | 0.27 ± 0.09 | 0.22 ± 0.09 | 0.22 ± 0.09 | 0.25 ± 0.10 | 0.38 ± 0.19 |
| p-Toluic acid | 0.24 ± 0.09 | 0.21 ± 0.09 | 0.20 ± 0.09 | 0.18 ± 0.07 | 0.20 ± 0.06 | 0.38 ± 0.19 |
| 1,2-Phthalic acid (1,2-Ph) | 24.0 ± 5.63 | 18.2 ± 6.24 | 20.0 ± 8.25 | 23.7 ± 6.17 | 25.6 ± 5.36 | 33.9 ± 17.5 |
| 1,4-Phthalic acid (1,4-Ph) | 28.6 ± 19.6 | 21.0 ± 14.0 | 28.1 ± 35.0 | 32.9 ± 27.4 | 29.8 ± 25.2 | 29.0 ± 14.6 |
| 1,3-Phthalic acid (1,3-Ph) | 1.12 ± 0.59 | 0.97 ± 0.61 | 0.86 ± 0.44 | 0.93 ± 0.52 | 0.80 ± 0.41 | 1.27 ± 0.81 |
| 4-Methyl-1,2-phthalic acid(4-Methyl-1,2-ph) | 4.86 ± 1.17 | 3.56 ± 1.05 | 3.73 ± 1.06 | 4.17 ± 1.62 | 4.24 ± 1.30 | 6.58 ± 2.66 |
| Trimellitic acid | 7.37 ± 4.19 | 5.61 ± 2.47 | 5.58 ± 3.05 | 6.19 ± 3.24 | 6.85 ± 3.08 | 9.89 ± 4.86 |
| Trimesic acid | 0.29 ± 0.13 | 0.20 ± 0.08 | 0.21 ± 0.10 | 0.26 ± 0.13 | 0.25 ± 0.11 | 0.38 ± 0.17 |
| Pyromellitic acid | 3.19 ± 2.11 | 1.87 ± 0.76 | 2.05 ± 0.99 | 1.62 ± 0.71 | 2.35 ± 1.16 | 4.31 ± 2.11 |
| 4-Methoxybenzoic acid | 0.95 ± 1.24 | 1.14 ± 0.80 | 1.41 ± 1.30 | 0.79 ± 0.53 | 1.80 ± 1.50 | 1.03 ± 1.61 |
| 3,4-Dimethoxy benzoic acid | 0.94 ± 0.76 | 0.98 ± 0.60 | 1.15 ± 0.83 | 1.24 ± 0.77 | 2.55 ± 2.02 | 0.98 ± 0.98 |
| Subtotal | 73.7 ± 27.9 | 56.2 ± 22.7 | 65.5 ± 38.2 | 73.8 ± 33.6 | 76.5 ± 34.8 | 93.3 ± 38.0 |
| | Carbon fraction | | | | | |
| Fatty acids (ng/µg OC) | 8.97 ± 2.43 | 8.01 ± 2.76 | 11.1 ± 5.52 | 13.1 ± 4.60 | 8.19 ± 1.07 | 6.50 ± 1.93 |
| Dicarboxylic acids (ng/µg OC) | 1.80 ± 0.83 | 3.02 ± 0.97 | 3.72 ± 1.70 | 2.58 ± 0.83 | 3.20 ± 1.21 | 5.44 ± 3.40 |
| Aromatic acids (ng/µg OC) | 3.62 ± 1.18 | 2.86 ± 0.95 | 5.37 ± 3.65 | 4.26 ± 0.78 | 5.30 ± 1.04 | 5.95 ± 3.12 |
| Organic carbon (µg/m ³) | 23.0 ± 12.4 | 20.8 ± 8.66 | 13.6 ± 6.18 | 18.3 ± 10.1 | 15.1 ± 8.16 | 15.5 ± 5.73 |
| Elemental carbon (µg/m ³) | 7.13 ± 3.69 | 5.69 ± 2.42 | 3.78 ± 2.03 | 4.18 ± 2.47 | 4.03 ± 2.56 | 4.84 ± 1.59 |
| Levoglucosan | 118 ± 51.6 | 131 ± 96.8 | 89.6 ± 44.9 | 179 ± 100 | 251 ± 137 | 116 ± 59.5 |
| 1,3,5-Triphenylebenzene | 0.42 ± 0.32 | 0.63 ± 0.50 | 0.49 ± 0.82 | 0.68 ± 0.84 | 0.44 ± 0.36 | 0.48 ± 0.25 |
| NDVI | -0.01 | -0.06 | 0.06 | -0.02 | 0.22 | 0.1 |

NDVI: normalized difference vegetation index. Data are expressed as mean ± standard deviations (SD).

erage value of 1.72%. Fatty acids predominated in organic acids and their average concentration was 157 ng/m^3 . The

mean concentration was 72.5 ng/m³ for aromatic acids and 50.7 ng/m³ for discarboxylic acids.

Saturated fatty acids (C14:0-C32:0) from this study in the PRD region showed a strong even carbon number predominance with a maximum at palmitic acid (C16:0) and stearic acid (C18:0) (Table 1). This was quite similar to the distribution patterns reported in the urban areas (Fraser et al., 2002; Fu et al., 2010; Ho et al., 2011), but different from the bimodal distribution observed at the mountain site (Fu et al., 2008) or in marine aerosols (Fu et al., 2011; Kawamura et al., 2003) with maximums at C16:0 and C24:0/C28:0. The concentrations of C16:0 and C18:0 in the PRD region averaged 85.8 and 27.8 ng/m³, respectively, comparable with those reported in some urban sites, such as Nanjing in China (Wang and Kawamura, 2005) and Chennai in India (Fu et al., 2010). On the Mt. Tai in north China, average C16:0 and C18:0 concentrations in TSP reached 60 and 19 ng/m³, respectively (Fu et al., 2008); other remote sites, however, had much lower C16:0 and C18:0 levels, such as 27.0 and 11.3 ng/m³, respectively, in a Germany forest during summertime (Kourtchev et al., 2008), the concentration in aerosols collected at the North Pacific Island and over the ocean during the ACE-Asia campaign were even lower (Simoneit et al., 2004).

Due to difference in extraction and derivatization procedures in this study, low molecular weight dicarboxylic acids were not available as those by Kawamura and Ikushima (1993), and therefore only C_4 – C_9 dicarboxylic acids are presented (Table 1). Succinic acid and azelaic acid were dominating species in C_4 – C_9 dicarboxylic acids in aerosols from various regions including marines and the Arctic (Narukawa et al., 2002; Simoneit et al., 2004). In the present study, succinic acid and azelaic acid were also the two most abundant dicarboxylic acids with average concentrations of 28.6 ng/m³ and 12.9 ng/m³, respectively. Their concentrations were in the same magnitude as those in other urban sites in Asia, such as 52.5 and 12.9 ng/m³ in roadside Hong Kong (Ho et al., 2006) and 42.8 and 24.5 ng/m³ in megacity India (Pavuluri et al., 2010), but were substantially higher than those reported in Houston, USA (Fraser et al., 2002), in Europe (Hyder et al., 2012; Kubátová et al., 2002), at high mountains (Fu et al., 2008) and in the Arctic region (Narukawa et al., 2002).

Detected aromatic acids, including benzoic acid, mtoluic acid and p-toluic acid, had much lower levels than fatty acids and dicarboxylic acids partly because that these semi-volatiles would partition much more in gas phase under higher temperature in the PRD region (Fraser et al., 2003). Benzoic acid, m-toluic acid and p-toluic acid averaged 2.46, 0.26 and 0.23 ng/m³, respectively, which were lower than those previously reported at four sites in PRD region in both summer and winter (Ho et al., 2011), but in the same magnitude as the benzoic acid of 1.25 ng/m³ and total toluic acids of 0.62 ng/m³ observed in the Indian City of Chennai during winter (Fu et al., 2010).

The aromatic polycarboxylic acids measured in this study included diacids, triacids and a tetraacid (**Table 1**).

Much higher levels of 1,2-phthalic acid than that of 1,4phthalic acid were typically observed (Fu et al., 2008; Ho et al., 2010, 2011). However, our study instead revealed higher 1,4-phthalic acid in average (28.2 ng/m³) than 1,2phthalic acid (23.9 ng/m³), the same as those reported in the polluted city sites, such as Chennai in India and Xi² an in China (Fu et al., 2010; Wang et al., 2012a). The levels of 1,4-phthalic acid and 1,2-phthalic acid quite approximated those reported in China and India, but were higher than those reported in USA.

2.2 Spatial distribution

Among the six sampling sites, the suburban site (HD) had the highest average level of fatty acids and the rural site (WQS) had the lowest; dicarboxylic acids and aromatic acids, however, peaked at WQS (**Table 1**). If the carbon fraction of organic acids are normalized to OC, the two suburban sites (HD and PY) showed the highest shares of fatty acids in OC, which were 11.1 and 13.1 ng/mg OC on average, respectively, and the rural site (WQS) had the lowest (6.50 ng/mg OC). In contrast, the average shares of dicarboxylic acids and aromatic acids in OC reached their peaks at WQS and showed relative lower values at the two urban sites SZ and FC.

2.3 Source attribution

2.3.1 Source identification

Higher molecular weight fatty acids ($\geq C_{20}$) (HFAs) are derived from terrestrial higher plant wax (Oliveira et al., 2007; Simoneit, 1986), while lower molecular weight fatty acids ($\leq C_{20}$) (LFAs) are derived from microbes and marine phytoplankton (Kawamura et al., 2003) and from anthropogenic sources, such as the fossil fuels combustion (He et al., 2006; 2008), biomass burning (Zhang et al., 2007) and cooking (He et al., 2004; Zhao et al., 2007a, 2007b). Higher concentrations of HFAs at the rural site KD suggested more contribution of terrestrial higher plant wax than other sites. This is supported by the highest normalized difference vegetation index (NDVI) from remote sensing data around KD (Table 1). On the other hand, emission of higher plant waxes was reported during biomass burning events (Alves et al., 2012). Higher levels of biomass burning marker levoglucosan and the significant relationship (r = 0.63, p < 0.01) between levoglucosan and HFAs at KD implied biomass burning activity might also promote the HFAs at KD. The major sources of LFAs in this study should be anthropogenic in the highly industrialized and densely populated PRD region, as can also be indicated by the higher LFAs in the urban sites.

Strong positive correlations (r > 0.63, p < 0.01) were observed among the C₄–C₉ dicarboxylic acids at all six sites (**Table 2**). Secondary formation was generally more important for dicarboxylic acids in PM_{2.5} (Chebbi and Carlier, 1996; Fraser et al., 2003), even though primary exhausts were possible sources (Kawamura and Kaplan, 1987).

| 1 | 1 | 5 |
|---|---|---|
| 1 | I | J |

| Table 2 Pearson correlation | tion coefficie | ents between t | he ambient c | oncentrations | of dicarboxylic acids | s and aromatic a | cids | |
|-------------------------------|------------------|----------------|--------------|---------------|-----------------------|---------------------|------------------|-------------------|
| | di-C4 | di-C5 | di-C6 | di-C7 | di-C8 | di-C9 | Benzoic acid | m-Toluic acid |
| di-C4 | 1 | 0.925** | 0.797** | 0.632** | 0.793** | 0.731** | 0.540** | 0.401** |
| di-C5 | | 1 | 0.846** | 0.746** | 0.782** | 0.772** | 0.496** | 0.388** |
| di-C6 | | | 1 | 0.802** | 0.837** | 0.826** | 0.586** | 0.374** |
| di-C7 | | | | 1 | 0.775** | 0.770** | 0.389** | 0.317** |
| di-C8 | | | | | 1 | 0.941** | 0.529** | 0.401** |
| di-C9 | | | | | | 1 | 0.515** | 0.414** |
| Benzoic acid | | | | | | 1 | 1 | 0.815** |
| m-Toluic acid | | | | | | | 1 | 1 |
| n-Toluic acid | | | | | | | | 1 |
| 1.2 Dh | | | | | | | | |
| 1,2-1 II 1 / Dh | | | | | | | | |
| 1,4-1 II 1 2 Dh | | | | | | | | |
| 1,3-FII 4 Mathul 1.2 mb | | | | | | | | |
| 4-Metnyi-1,2-pn | | | | | | | | |
| | | | | | | | | |
| | | | | | | | | |
| ryromeintic acid | 0 (10++ | 0 100** | 0 (5(** | 0 10744 | 0.727** | 0 (21** | 0 500** | 0 411** |
| cis-Pinonic acid | 0.618** | 0.489** | 0.656** | 0.427** | 0.737** | 0.631** | 0.588** | 0.411** |
| Pinic acid | 0.704** | 0.578** | 0.744** | 0.50/** | 0.812** | 0.717** | 0.554** | 0.355** |
| C16:0 | 0.112 | 0.219* | 0.134 | 0.242** | 0.118 | 0.301** | 0.109 | 0.240** |
| C18:0 | 0.162 | 0.267** | 0.143 | 0.221* | 0.060 | 0.253** | 0.143 | 0.224* |
| C18:1 | -0.056 | 0.008 | -0.023 | 0.086 | -0.054 | 0.000 | 0.054 | 0.129 |
| OC | 0.297** | 0.367** | 0.163 | 0.270** | 0.158 | 0.274** | 0.220* | 0.336** |
| EC | 0.292** | 0.368** | 0.192* | 0.256** | 0.177* | 0.298** | 0.278** | 0.358** |
| Levoglucosan | 0.219* | 0.243** | 0.222* | 0.314** | 0.304** | 0.337** | 0.195* | 0.367** |
| 1,3,5-Triphenylebenzene | 0.293** | 0.375** | 0.242** | 0.281** | 0.208* | 0.330** | 0.249** | 0.298** |
| | p-Toluic acid | 1,2-Ph | 1,4-Ph | 1,3-Ph | 4-Methyl-1,2-ph | Trimellitic acid | Trimesic acid | Pyromellitic acid |
| | | | | | | | | |
| di-C4 | 0.547** | 0.464** | 0.258** | 0.354** | 0.482** | 0.479** | 0.490** | 0.514** |
| di-C5 | 0.51/** | 0.3/6** | 0.321** | 0.424** | 0.409** | 0.441** | 0.453** | 0.4/1** |
| di-C6 | 0.525** | 0.45/** | 0.205* | 0.370** | 0.445** | 0.440** | 0.429** | 0.499** |
| di-C/ | 0.397** | 0.267** | 0.208* | 0.437** | 0.214* | 0.423** | 0.356** | 0.408** |
| di-C8 | 0.596** | 0.486** | 0.235** | 0.365** | 0.478** | 0.520** | 0.521** | 0.583** |
| di-C9 | 0.608** | 0.416** | 0.374** | 0.428** | 0.480** | 0.465** | 0.490** | 0.521** |
| Benzoic acid | 0.808** | 0.656** | 0.337** | 0.659** | 0.648** | 0.612** | 0.566** | 0.496** |
| m-Toluic acid | 0.805** | 0.644** | 0.372** | 0.739** | 0.576** | 0.666** | 0.615** | 0.429** |
| p-Toluic acid | 1 | 0.704** | 0.370** | 0.658** | 0.663** | 0.714** | 0.703** | 0.641** |
| 1,2-Ph | | 1 | 0.163 | 0.509** | 0.799** | 0.791** | 0.751** | 0.611** |
| 1,4-Ph | | | 1 | 0.603** | 0.329** | 0.295** | 0.406** | 0.187* |
| 1,3-Ph | | | | 1 | 0.422** | 0.695** | 0.656** | 0.418** |
| 4-Methyl-1,2-ph | | | | | 1 | 0.596** | 0.702** | 0.596** |
| Trimellitic acid | | | | | | 1 | 0.916** | 0.825** |
| Trimesic acid | | | | | | | 1 | 0.785** |
| Pyromellitic acid | | | | | | | | 1 |
| cis-Pinonic acid | 0.618** | 0.489** | 0.656** | 0.427** | 0.737** | 0.631** | 0.588** | 0.411** |
| Pinic acid | 0.704** | 0.578** | 0.744** | 0.507** | 0.812** | 0.717** | 0.554** | 0.355** |
| C16:0 | 0.112 | 0.219* | 0.134 | 0.242** | 0.118 | 0.301** | 0.109 | 0.240** |
| C18:0 | 0.162 | 0.267** | 0.143 | 0.221* | 0.060 | 0.253** | 0.143 | 0.224* |
| C18:1 | -0.056 | 0.008 | -0.023 | 0.086 | -0.054 | 0.000 | 0.054 | 0.129 |
| OC | 0.297** | 0.367** | 0.163 | 0.270** | 0.158 | 0.274** | 0.220* | 0.336** |
| EC | 0.292** | 0.368** | 0.192* | 0.256** | 0.177* | 0.298** | 0.278** | 0.358** |
| Levoglucosan | 0.219* | 0.243** | 0.222* | 0.314** | 0.304** | 0.337** | 0.195* | 0.367** |
| 1,3,5-Triphenylebenzene | 0.293** | 0.375** | 0.242** | 0.281** | 0.208* | 0.330** | 0.249** | 0.298** |

** Value with p < 0.01; * value with p < 0.05.

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These low molecular-weight diacids are produced photochemically in the polluted atmosphere from oxidation of cyclic olefins, diolefins, monocarboxylic acids, mid-chain ketocarboxylic acids, ω-ketocarboxylic acids (Mochida et al., 2007; Wang et al., 2011) and also unsaturated fatty acid (Kawamura and Sakaguchi, 1999; Kawamura et al., 1996). As for the higher succinic acid, it is interesting that significant correlation between succinic acid and levoglucosan (r = 0.62, p < 0.01) was found in the rural site KD, suggesting the contribution of biomass burning to succinic acid in this site. In fact, high levels of succinic acid were observed in aerosols under the influence of biomass burning (Falkovich et al., 2005; Narukawa et al., 1999; Wang et al., 2011; Yan et al., 2008). However, biomass burning should not be the major source for succinic acids at other sites and instead secondary formation would be more important, as much higher levels were observed at the downwind WQS. Azelaic acid is regarded as an oxidation product of oleic acid by O₃, H₂O₂ or OH radical (Kawamura and Gagosian, 1987; Stephanou and Stratigakis, 1993). Oleic acid is reported to be rich in marine phytoplankton and terrestrial higher plant leaves (Kawamura and Gagosian, 1987). In the PRD region, anthropogenic sources such as cooking (He et al., 2004; Zhao et al., 2007a, 2007b), would be much more important. As can be seen, concentrations of oleic acid from urban to rural sites (Fig. 2), and the highest concentration of azelaic acid were observed at the downwind rural site WQS. The ratios of azelaic to oleic acid were much higher at the two rural sites with averages of 40.4 at WQS and 33.9 at KD, further confirming the oxidizing of oleic acids to azelaic acids during transport from the urban source areas to the rural (Kawamura and Gagosian, 1987; Kawamura and Sakaguchi, 1999). Similar formation process of dicarboxylic acid was also observed in a background site in Sweden (Hyder et al., 2012).

1,2-Phthalic acid has been proposed as secondarily formed by the oxidation of naphthalene (Fine et al., 2004; Kleindienst et al., 2012) or other polycyclic aromatic hydrocarbons such as benz(a)antharacene (Jang and McDow, 1997). Concentration of naphthalene which is a ubiquitous



Fig. 2 Spatial variation of azelaic acid, oleic acid and the ratio of azelaic acid to oleic acid at the six sites.

pollutant in the atmosphere was reported to be as high as 3.5 μ g/m³ in Hong Kong in the PRD region (Lee et al., 2001). In the present study, the higher abundance of 1,2-phthalic acid in the rural site WOS was supposed to be secondarily formed during transport of naphthalene from the polluted urban areas. 1,2-Phthalicacid was highly correlated with dicarboxylic acids, benzoic acid and other aromatic polycarboxylic acids except for 1,4-phthalic acid (Table 2), also suggesting that secondary formation was much more important for 1,2-phthalic acid, and 1,4phthalic acid had sources other than secondary formation. As an important industrial raw material in manufacturing polyethylene terephthalate (PET) fiber and plastic bottles (Kawamura and Pavuluri, 2010), 1,4-phthalic acid was reported to have elevated levels derived from open burning of plastics, roadside litter and landfill trash (Simoneit et al., 2005). In this study, 1,4-phthalic acid was found to be significantly correlated with 1,3,5-triphenylebenzene (r = 0.70, p < 0.01), which is a specific marker for the open-burning of plastics (Simoneit et al., 2005). This source of 1,4-phthalic acid can largely explain its higher levels in many Asian cities (Fu et al., 2010; Ho et al., 2010; 2011; Wang et al., 2012a) and its rare occurrence in aerosols from oceans and the Arctic (Narukawa et al., 2002; Simoneit et al., 2004). The quite different spatial patterns of 1,4-phthalic acid comparing to the secondarily formed 1,2-phthalic acid, as well as significant correlation between 1,4-phthalic acid and EC (r = 0.59, p < 0.01; Table 2), also supports that 1,4-phthalic acid was a primary pollutant. Additionally, cis-pinonic acid and pinic acid, as typical SOA tracers from biogenic monoterpenes (Ding et al., 2011) in the region, also had good correlations (p < 0.01, Table 2) with the detected diacids and aromatic acids except 1,4-phthalic acid, further supporting that 1,4phthalic acid was primary while other aromatic acids and diacids were mainly secondary.

2.3.2 Principal component analysis

Principal component analysis (PCA) followed by multiple linear regression was applied to the dataset obtained in the present study to explore the major sources of organic acids (Larsen and Baker, 2003). The purpose of PCA is to represent the total variability of the original organic acids data in a minimum number of factors. Each factor is orthogonal to all others, which results in the smallest possible covariance. The first factor represents the weighted (factor loadings) linear combination of the original variables that account for the greatest variability. Each subsequent factor accounts for less variability than the previous (Table 3). By critically evaluating the factor loadings, an estimate of the chemical source responsible for each factor can be made. Six factors were resolved which altogether explained 87% of the total variance. Factor 1 was heavily loaded by C₁₉- C_{32} *n*-alkanoic acids, and therefore was identified to be · Jose . He . Ch associated with natural sources such as plant wax (Oliveira

Table 3 Varimax rotated component matrix of organic acids from the PRD region

| | | Principal components | | | | |
|----------------------------|--------|----------------------|--------|--------|--------|--------|
| | 1 | 2 | 3 | 4 | 5 | 6 |
| Variance (%) | 46.6 | 17.6 | 7.73 | 6.69 | 4.52 | 3.66 |
| C14:0 | 0.378 | 0.120 | 0.115 | 0.828 | -0.110 | 0.062 |
| C15:0 | 0.589 | 0.156 | 0.139 | 0.635 | -0.040 | 0.114 |
| C16:0 | 0.330 | -0.005 | 0.094 | 0.865 | -0.006 | 0.116 |
| C17:0 | 0.573 | 0.054 | 0.096 | 0.753 | 0.113 | 0.100 |
| C18:0 | 0.405 | -0.021 | 0.081 | 0.837 | -0.042 | 0.111 |
| C19:0 | 0.792 | 0.158 | 0.213 | 0.482 | 0.146 | 0.043 |
| C20:0 | 0.808 | 0.183 | 0.158 | 0.460 | 0.104 | 0.157 |
| C21:0 | 0.871 | 0.180 | 0.187 | 0.335 | 0.116 | 0.019 |
| C22:0 | 0.882 | 0.160 | 0.128 | 0.343 | 0.132 | 0.089 |
| C23:0 | 0.944 | 0.204 | 0.119 | 0.150 | 0.068 | -0.001 |
| C24:0 | 0.936 | 0.198 | 0.131 | 0.161 | 0.095 | 0.038 |
| C25:0 | 0.932 | 0.121 | 0.087 | 0.198 | 0.050 | 0.017 |
| C26:0 | 0.934 | 0.124 | 0.104 | 0.229 | 0.129 | 0.048 |
| C27:0 | 0.941 | 0.128 | 0.070 | 0.227 | -0.030 | 0.026 |
| C28:0 | 0.968 | 0.103 | 0.045 | 0.177 | 0.044 | 0.024 |
| C29:0 | 0.949 | 0.089 | 0.031 | 0.164 | -0.051 | 0.060 |
| C30:0 | 0.980 | 0.097 | -0.007 | 0.067 | -0.019 | 0.007 |
| C31:0 | 0.911 | 0.078 | 0.009 | 0.108 | -0.045 | 0.085 |
| C32:0 | 0.961 | 0.071 | -0.009 | 0.063 | -0.022 | 0.035 |
| C16:1 | 0.101 | 0.091 | 0.304 | -0.263 | 0.271 | 0.626 |
| C18:1 | 0.093 | 0.027 | -0.080 | 0.345 | -0.124 | 0.858 |
| C18:2 | 0.103 | 0.089 | -0.147 | 0.279 | -0.087 | 0.874 |
| di-C4 | 0.138 | 0.342 | 0.814 | 0.017 | -0.035 | -0.049 |
| di-C5 | 0.133 | 0.258 | 0.860 | 0.153 | -0.001 | -0.033 |
| di-C6 | -0.027 | 0.317 | 0.884 | 0.064 | -0.042 | 0.011 |
| di-C7 | 0.081 | 0.127 | 0.840 | 0.163 | 0.088 | 0.058 |
| di-C8 | 0.117 | 0.351 | 0.870 | -0.029 | 0.005 | -0.006 |
| di-C9 | 0.165 | 0.309 | 0.839 | 0.170 | -0.030 | -0.024 |
| Benzoic acid | -0.091 | 0.749 | 0.342 | 0.200 | 0.215 | 0.055 |
| m-Toluic acid | 0.102 | 0.748 | 0.155 | 0.274 | 0.296 | 0.065 |
| p-Toluic acid | 0.086 | 0.799 | 0.329 | 0.194 | 0.152 | -0.027 |
| 1,2-Ph | 0.121 | 0.856 | 0.199 | -0.155 | 0.156 | -0.018 |
| 1,4-Ph | 0.503 | 0.167 | 0.156 | 0.557 | 0.180 | 0.160 |
| 1,3-Ph | 0.237 | 0.586 | 0.183 | 0.527 | 0.281 | 0.112 |
| 4-Methyl-1,2-ph | 0.207 | 0.790 | 0.216 | -0.033 | -0.153 | 0.103 |
| Trimellitic acid | 0.251 | 0.824 | 0.229 | 0.037 | 0.185 | 0.003 |
| Trimesic acid | 0.406 | 0.805 | 0.209 | 0.059 | 0.016 | 0.048 |
| Pyromellitic acid | 0.185 | 0.716 | 0.338 | -0.056 | -0.096 | 0.045 |
| 4-Methoxybenzoic acid | -0.005 | 0.240 | -0.036 | 0.031 | 0.893 | -0.038 |
| 3,4-Dimethoxy benzoic acid | 0.229 | 0.182 | -0.011 | -0.020 | 0.911 | -0.012 |

et al., 2007; Simoneit, 1986). Factor 4 was dominated by $C_{14}-C_{18}$ *n*-alkanoic acids and 1,4-phthalic acid, and Factor 6 by the three unsaturated acids. These two factors were mixed anthropogenic sources including vehicle exhausts (He et al., 2006, 2008) and cooking (He et al., 2004; Zhao et al., 2007a, 2007b). Factor 5 was strongly correlated with the two oxygenated aromatic acids, 4-methoxybenzoic acid and 3,4-dimethoxy benzoic acid, which were lignin pyrolysis products primarily emitted from biomass burning (Simoneit et al., 1993). Besides, Factor 5 had significant

correlations with the biomass burning marker levoglucosan (r = 0.43, p < 0.01). Therefore this factor was associated with biomass burning. C₄–C₉ dicarboxylic acids associated with the photochemical oxidation showed good correlation with Factor 3, while benzoic acid, toluic acid and most of aromatic polycarboxylic acids except 1,4-phthalic acid were heavily loaded in Factor 2. These two factors all represented secondary sources but with different precursors.

The ultimate goal of performing PCA/MLR is to de-



Fig. 3 Source attribution of individual organic acids (a) and the contribution of the six factors to organic acids at the six sites (b).

termine the percent contribution of different organic acids sources for a given ambient sample. The basic equation of a multiple linear regression (MLR) of $Y = \sum B_i X_i$ was performed using factor scores (X_i) as independent variables and total organic acids mass concentration (Y) as dependent variable. With the regression coefficients (B_i) by MLR, the influence of each factor on the organic acids mass could then be directly compared and the mean contribution percentage by source *i* was calculated as $100 (B_i/\sum B_i)$.

The contribution of different sources of individual compounds by multiple linear regressions followed PCA was presented in Fig. 3a. More than 70% of C₁₄-C₁₈ fatty acids were attributed to anthropogenic sources, 80%-95% of dicarboxylic acids and 1,2-phthalic acid were secondary in contrast with that 81% of 1,4-phthalic acid was primary. Biomass burning contributed about 70% to 4-methoxybenzoic acid and 3,4-dimethoxy benzoic acid, and natural sources accounted for 50%-85% of the HFAs. These findings were well consistent with discussions about source attribution in the above parts. Histograms of the four major factor groups at the six sampling sites are shown in Fig. 3b. The spatial distribution showed that the urban and suburban sites had higher average level of anthropogenic sources than rural sites. The rural site KD had the highest nature sources loadings (84.3 ng/m^3). Contributions of biomass burning were also significant higher at rural site KD (11.0 ng/m³), where the biomass burning was more frequent. The secondary organic acids significantly increased at the downwind rural site WQS with the average value of 99.3 ng/m^3 .

3 Conclusions

 C_{14} - C_{32} fatty acids, C_4 - C_9 dicarboxylic acids and aromatic acids in PM_{2.5} were investigated during winter 2009 at typical urban, suburban and rural sites in the Pearl River Delta (PRD) region. C14-C32 fatty acids, aromatic acids and C₄-C₉ dicarboxylic acids averaged 157, 72.5 and 50.7 ng/m³, respectively. Palmitic acid and stearic acid were dominant fatty acids, succinic acid and azelaic acid were the most abundant among C_4 – C_9 diacids, and 1,4phthalic acid and 1,2-phthalic acid were major aromatic acids. The sources of the organic acids were explored through three aspects: spatial patterns, mutual correlation as well as their correlation with typical source tracers, and principal component analysis followed by multiple linear regressions. Fatty acids were mainly primary with C₂₀-C₃₂ fatty acids mainly from natural higher plant wax and C14-C18 fatty acids mainly from anthropogenic emissions including cooking. Dicarboxylic acids and aromatic acids except 1,4-phthalic acid were largely secondarily formed, and 1,4-phthalic acid was probably primarily emitted from combustion of solid wastes containing PET plastics. Principal component analysis resolved six sources including biomass burning, natural higher plant wax, two mixed anthropogenic and two secondary sources, and it turned out that more than 70% of C_{14} - C_{18} fatty acids were attributed to anthropogenic sources, 50%-85% of the HFAs were attributed to natural sources, 80%-95% of dicarboxylic acids and 1,2-phthalic acid were secondary in contrast with that 81% of 1,4-phthalic acid was primary.

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