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TELLUS

Selected water-soluble organic compounds found in size-resolved aerosols collected from urban, mountain and marine atmospheres over East Asia

By GEHUI WANG[1,2,*], KIMITAKA KAWAMURA[2], MINGJIE XIE[3], SHUYUAN HU[3], JIANJUN LI[1], BIANHONG ZHOU[1], JUNJI CAO[1] and ZHISHENG AN[1], [1]*State Key Laboratory of Loess and Quaternary Geology, Institute of Earth Environment, Chinese Academy of Sciences, Xi'an 710075, China;* [2]*Institute of Low Temperature Science, Hokkaido University, Sapporo 060-0819, Japan;* [3]*School of the Environment, Nanjing University, Nanjing 210093, China*

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ABSTRACT

Primary (i.e. sugars and sugar-alcohols) and secondary (i.e. carboxylic acids) water-soluble organic compounds (WSOCs) in size-segregated aerosols from the urban and mountain atmosphere of China and from the marine atmosphere in the outflow region of East Asia were characterized on a molecular level. Levoglucosan is the most abundant compound among the quantified WSOCs in the urban and mountain atmosphere, whose concentration at the urban site was 1–2 orders of magnitude higher than that at the mountain and marine sites. In contrast, malic, succinic and phthalic acids were dominant among the measured WSOCs at the marine site. In the urban air, sugars except levoglucosan gave a bimodal size distribution with a large peak in fine range (<2.1 μm) and a small peak in coarse range (≥ 2.1 μm) during winter, being opposite to those in spring. In contrast, these WSOCs at the mountain and marine sites dominated in the coarse range but diminished and even disappeared in the fine range. Geometric mean diameters (GMDs) of the measured WSOCs in the fine mode at the urban site were larger in winter than in spring. Levoglucosan and carboxylic acids except for azelaic and benzoic acids showed a larger GMD in the coarse mode at the marine site probably due to an increased hygroscopic growth.

1. Introduction

Water-soluble organic compounds (WSOCs) constitute a substantial fraction of atmospheric organic matter, accounting for 10–90% of organic carbon content in fine ambient aerosols depending on locations (Mader et al., 2004; Ming and Russell, 2004; Yu et al., 2004; Pöschl, 2005). WSOCs play an important role in global climate change by altering the hygroscopicity of atmospheric aerosols (Cruz and Pandis, 2000; Decesari et al., 2005; Engling and Gelencser, 2010; Kuwata et al., 2008). WSOCs can also cause a negative effect on human health by increasing the solubility of toxic pollutants (Sorjamaa et al., 2004). Moreover, some of WSOCs are allergens, leading to asthma and other diseases (Franze et al., 2004, 2005).

^{1}H NMR (Decesari et al., 2007; Graham et al., 2002) and ^{13}C NMR (Sannigrahi et al., 2006) analyses indicate that WSOCs are predominantly composed of aliphatic compounds containing carboxyl, carbonyl and hydroxyl groups (carboxylic acids, aldehydes, sugars, alcohols, etc.) with a minor content of aromatic rings bearing carboxylic and phenolic groups. Dehydrated sugars (e.g. levoglucosan, mannosan and galactosan) are the pyrolysis products of cellulose and hemicellulose (Fraser and Lakshmanan, 2000; Simoneit et al., 2004a; Simoneit et al., 2003; Wang et al., 2009a; Wang et al., 2009c), whereas other sugars (e.g. glucose, fructose and sucrose) and sugar-alcohols (e.g. arabitol, mannitol and inositol) are the metabolic products of biota including pollen, spores, fungi, algae and bacteria in soil (Wang et al., 2009b). These sugars and sugar-alcohols are directly emitted from the sources and generally taken as primary WSOCs. Sizes of those primary aerosols range from a few decade nanometres to hundreds of micrometres (Carvalho et al., 2003; Wang et al., 2009c). Dicarboxylic acids and related compounds are another important class of WSOCs in the atmosphere, major portion of which results from photo-oxidation of organic precursors and are thus considered in general as secondary products (Blando et al., 1998; Claeys et al., 2007; Glasius et al., 2000; Kawamura and Yasui, 2005; Wang et al., 2009b).

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[*]Corresponding author.
e-mail: wanggh@ieecas.cn or gehuiwang@yahoo.com.cn
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Numerous studies have demonstrated that chemical and physical properties of atmospheric aerosols are area and location specific (Pierce and Katz, 1975; Yoshizumi and Hoshi, 1985; Venkataraman et al., 2002; Greenwald et al., 2007). For better understanding the relationships between East Asian aerosols and their climate impacts, it is necessary to investigate differences in the chemical and physical characteristics of organic aerosols in the urban, mountain and marine atmosphere over East Asia. In this study, we first report the difference in the composition and concentration of the primary and secondary WSOCs in the three types of atmospheric environments, then discuss the difference in their size distributions and sources.

2. Experiment

2.1 Sample collection

Baoji is a mid-scale city located in central China with a population of 0.75 million (Fig. 1). The sampler was set on the rooftop of a three-floor building at the Environment Monitoring Station of Baoji, which is located in the urban centre. Winter sampling was performed on 11–14 January 2008 and 12–20 February 2008, whereas spring sampling was conducted on 12–24 April 2008. Size-segregated mountain aerosols were collected on the mountaintop (elevation of 1534 m a.s.l.) of Mt. Tai located in the central North China Plain, which faces to the East China Sea, Korean Peninsula and Japanese Islands (Fig. 1). The mountain sampling was performed for 12 days (12–24 January 2008) in the winter on the square in front of the Meteorological Station located on the mountaintop. Size-segregated marine aerosols were collected from 18 March to 12 April 2008 at Cape Hedo, Okinawa Island, Japan (Fig. 1). There is no major industry in the island and local anthropogenic activities are insignificant.

Size-segregated aerosols at the urban and mountain sites were collected using Andersen 8-stage air sampler (Thermo Electron Corporation, Franklin, MA, USA) with the cutoff points at 9.0, 5.8, 4.7, 3.3, 2.1, 1.1, 0.7 and 0.4 μm at an airflow rate of 28.3 L min^{-1} (filter size Ø90 mm). On the other hand, the marine size-resolved aerosols were collected on pre-combusted quartz filters (Ø80 mm) by using another type of Andersen 8-stage sampler (Tokyo Dylec Company, Japan) at a flow rate of 100 L min^{-1} with cutoff points at 11.3, 7.0, 4.7, 3.3, 2.1, 1.1, 0.7 and 0.4 μm. All the collected samples were stored at -20 °C before analysis. Information including dates, sample numbers, sampling durations and particle concentrations is shown in Table 1.

2.2 Sample extraction, derivatization and GC/MS analysis

Detailed methods for extraction, derivatization and gas chromatography/mass spectrometer (GC/MS) analysis are described

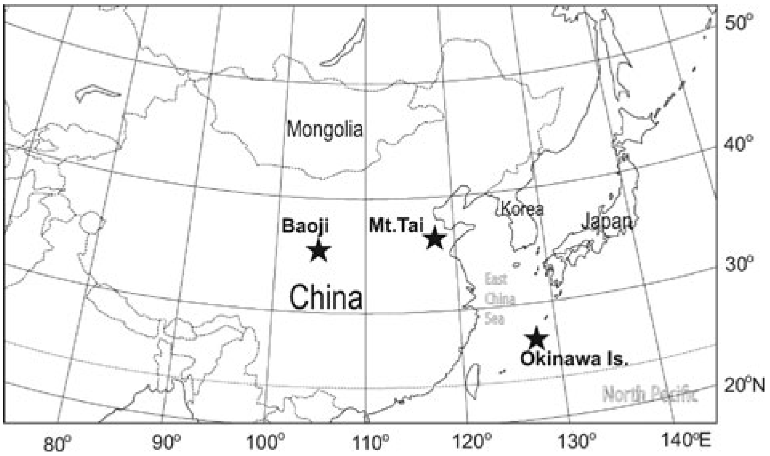

Fig. 1. A map description for the sampling sites and surroundings.

Table 1. Sampling information during the 2008 Campaign

Site	Season	Date	Duration of each sampling	No. of sample sets	TSP[a] (μg m^{-3})
Urban (Baoji, China)	Winter	11–14 Jan 12–20 Feb	4 days	3	316 ± 76
	Spring	12–24 Apr	4 days	3	286 ± 150
Mountain (Mt. Tai, China)	Winter	12–24 Jan	4 days	3	102 ± 60
Marine (Okinawa Is., Japan)	Spring	18 Mar–12 Apr	3–5 days	5	ND[b]

[a] TSP, total suspended particles, the sum of particle concentration on all stages of the impactor.

[b] ND, not determined.

elsewhere (Wang et al., 2009d). Briefly, aliquots of the sample and blank filter were extracted with a mixture of dichloromethane and methanol (2:1, v/v) under ultrasonication. The extracts were concentrated to dryness and reacted with BSTFA at 70 °C for 3 h. Finally, the derivatives were diluted with *n*-hexane prior to GC/MS determination.

GC/MS analysis of the derivatized fraction was performed using a Hewlett–Packard 6890 GC coupled to a Hewlett–Packard 5973 MSD. The GC separation was carried out on a DB-5MS fused silica capillary column with the GC oven temperature programmed from 50 °C (2 min) to 120 °C at 15 °C min^{-1} and then to 300 °C at 5 °C min^{-1} with final isothermal hold at 300 °C for 16 min. The sample was injected in a splitless mode at an injector temperature of 280 °C, and scanned from 50 to 650 Da using electron impact (EI) mode at 70 eV. GC/MS response factors were determined using authentic standards.

Average recoveries of the target compounds were better than 80% except for oxalic acid. Oxalic acid was somewhat depleted because of the volatility of the trimethylsilyl ester derivatized in this study compared to the previous data (Wang and Kawamura, 2005; Wang et al., 2009b). Malonic acid co-eluted with other unknown compounds in the GC/MS analysis (Simoneit et al., 2004c; Wang and Kawamura, 2005). Therefore, both compounds are omitted here. No serious contamination was found in the field blanks. Detection limits of all the target compounds are in the range of 0.003 ng m^{-3} for benzoic acid to 0.032 ng m^{-3} for malic acid. Here, we focus on chemical compositions and size distributions of the primary (i.e. sugars and sugar-alcohols) and

Table 2. Concentrations of water-soluble organic compounds in the fine (<2.1 μm) and coarse (≥2.1 μm) particles from the urban, mountain and marine atmosphere over East Asia, μg m^{-3}

	Urban				Mountain		Marine	
	Winter		Spring		Winter		Spring	
	Fine	Coarse	Fine	Coarse	Fine	Coarse	Fine	Coarse
I. Primary water-soluble organic compounds								
PM[a] (μg m^{-3})	203 ± 45	113 ± 35	124 ± 57	162 ± 93	54 ± 38	48 ± 23	NA[b]	NA[b]
Levoglucosan	729 ± 42	210 ± 37	336 ± 97	51 ± 14	90 ± 42	13 ± 7.7	2.7 ± 1.7	0.8 ± 0.2
Fructose	5.0 ± 4.1	5.6 ± 3.6	10 ± 4.0	11 ± 2.3	0.6 ± 0.3	1.6 ± 1.5	0.0 ± 0.0	11 ± 6.6
Glucose	16 ± 2.6	11 ± 6.3	7.9 ± 1.9	25 ± 3.3	1.4 ± 0.7	3.1 ± 2.8	0.2 ± 0.3	17 ± 10
Sucrose	4.1 ± 1.6	12 ± 9.1	1.5 ± 0.5	43 ± 23	0.2 ± 0.2	1.4 ± 1.0	0.0 ± 0.0	31 ± 7.5
Trehalose	0.3 ± 0.3	5.7 ± 4.6	0.2 ± 0.2	11 ± 4.1	0.0 ± 0.1	2.5 ± 1.7	0.0 ± 0.0	5.8 ± 2.1
Arabitol	11 ± 2.1	5.2 ± 2.9	2.2 ± 0.4	11 ± 5.0	0.4 ± 0.2	1.8 ± 1.5	0.0 ± 0.0	3.4 ± 3.1
Mannitol	3.8 ± 0.9	4.3 ± 2.9	1.1 ± 0.4	11 ± 4.1	0.0 ± 0.0	2.1 ± 2.0	0.0 ± 0.0	4.5 ± 3.1
Inositol	4.1 ± 1.1	0.8 ± 0.3	3.1 ± 0.8	1.2 ± 0.4	0.2 ± 0.1	0.2 ± 0.2	0.0 ± 0.0	0.4 ± 0.2
Glycerol	80 ± 21	20 ± 6.7	51 ± 25	22 ± 11	6.1 ± 2.4	4.4 ± 2.0	0.3 ± 0.2	2.2 ± 0.8
Subtotal	854 ± 69	275 ± 70	413 ± 125	186 ± 50	99 ± 46	30 ± 20	3.3 ± 1.6	75 ± 27
Subtotal/PM (%)	0.4 ± 0.1	0.3 ± 0.0	0.4 ± 0.1	0.1 ± 0.0	0.2 ± 0.1	0.1 ± 0.0	NA[b]	NA[b]
II. Secondary water-soluble organic compounds								
Glyceric acid	31 ± 16	5.0 ± 2.7	16 ± 4.8	3.1 ± 1.9	6.3 ± 4.8	1.0 ± 1.1	2.7 ± 1.2	1.2 ± 0.3
Malic acid	32 ± 20	3.7 ± 3.4	40 ± 16	3.7 ± 2.3	10 ± 8.8	0.6 ± 0.8	14 ± 5.1	2.1 ± 0.7
Succinic acid	87 ± 35	24 ± 11	49 ± 5.3	22 ± 10	23 ± 15	10 ± 8.0	8.6 ± 2.6	6.8 ± 1.0
Glutaric acid	39 ± 17	10 ± 5.6	17 ± 2.2	9.4 ± 3.7	12 ± 8.6	6.9 ± 6.3	1.4 ± 0.5	2.3 ± 0.5
Azelaic acid	46 ± 17	13 ± 7.0	26 ± 5.1	15 ± 3.1	7.6 ± 5.0	1.8 ± 1.6	0.9 ± 1.2	0.5 ± 0.8
Maleic acid	2.3 ± 0.7	1.2 ± 0.3	1.5 ± 0.2	0.9 ± 0.2	1.2 ± 0.8	0.6 ± 0.3	0.1 ± 0.0	0.1 ± 0.0
Fumaric acid	23 ± 7.2	4.4 ± 1.6	8.5 ± 1.5	3.4 ± 1.6	6.2 ± 4.6	2.1 ± 2.0	1.9 ± 0.5	1.0 ± 0.1
Benzoic acid	10 ± 2.6	5.7 ± 1.2	4.1 ± 1.4	3.6 ± 1.5	5.6 ± 2.9	3.5 ± 1.6	0.3 ± 0.1	0.4 ± 0.2
o-Phthalic acid	102 ± 26	47 ± 19	46 ± 8.0	32 ± 14	62 ± 52	26 ± 16	6.0 ± 4.1	1.7 ± 1.0
m-Phthalic acid	7.5 ± 3.5	1.4 ± 0.8	2.8 ± 0.8	0.7 ± 0.5	2.9 ± 2.6	0.7 ± 1.0	0.4 ± 0.3	0.1 ± 0.1
p-Phthalic acid	101 ± 48	52 ± 26	50 ± 13	21 ± 7.1	30 ± 23	8.2 ± 7.4	2.4 ± 1.7	0.6 ± 0.3
m-Salicyclic acid	14 ± 5.8	3.2 ± 1.6	2.7 ± 0.9	0.9 ± 0.3	1.4 ± 0.7	0.3 ± 0.2	0.1 ± 0.0	0.0 ± 0.0
p-Salicyclic acid	28 ± 13	19 ± 23	4.7 ± 1.6	2.0 ± 0.5	4.4 ± 2.4	0.8 ± 0.7	0.1 ± 0.1	0.1 ± 0.0
Subtotal	523 ± 197	190 ± 89	267 ± 52	118 ± 46	173 ± 129	62 ± 47	39 ± 149	17 ± 2.3
Subtotal/PM (%)	0.3 ± 0.1	0.2 ± 0.0	0.2 ± 0.1	0.1 ± 0.0	0.3 ± 0.1	0.1 ± 0.0	NA[b]	NA[b]

[a] PM, particle mass.

[b] NA, not available.

secondary (i.e. dicarboxylic and aromatic acids) WSOCs, whereas those of water-insoluble organic compounds in the samples have been reported in a companion paper (Wang et al., 2009d).

3. Results and discussion

3.1. Compositions and size distributions of primary WSOCs

Concentrations of WSOCs in the impactor stages with cutoff sizes less than 2.1 μm and larger than 2.1 μm were summed in Table 2 as fine (<2.1 μm) and coarse (≥ 2.1 μm) modes, respectively. Among the sugar and sugar-alcohol class, levoglucosan shows concentrations 1–3 orders of magnitude higher than other compounds in the fine and coarse particles at the urban and mountain sites, accounting for 70–90% of the primary WSOCs. Levoglucosan in the fine mode at the marine site is much more abundant than other sugars and sugar-alcohols, but its concentrations on the coarse mode are much less than other primary WSOCs (Table 2). Ambient concentration of levoglucosan in the urban air is around one order of magnitude higher than that in the mountain air and two orders of magnitude higher than that in the marine atmosphere. Levoglucosan, a tracer for biomass burning smokes, is chemically stable in the atmosphere (Fraser and Lakshmanan, 2000). Thus, such a difference in concentrations among the three sites can be ascribed to the difference in sources, that is, the urban aerosols are mostly contributed from local sources whereas the mountain and marine aerosols are largely derived from regional transport (Wang et al., 2009d).

Figure 2 illustrates the size distributions of levoglucosan and glucose in the three types of atmospheric environments. Levoglucosan showed a unimodal size distribution with a peak at the range of 0.7–1.1 μm for all the samples (Figs 2a–c). Levoglucosan is a dehydrated sugar produced by pyrolysis of cellulose-containing materials. It is initially emitted from the flame as gaseous molecule cluster and subsequently condenses onto pre-existing particles as temperature decrease, thus, exists mostly in fine particles (Simoneit and Elias, 2001). In contrast, glucose showed a bimodal size distribution in the urban and mountain air, peaking at 0.7–2.1 and >3.1 μm ranges, and a unimodal distribution in the marine atmosphere, peaking at the size >2.1 μm (Figs 2d–f). Other sugars and sugar-alcohols showed a size distribution pattern similar to glucose in the three atmospheric environments. Sugars, except for anhydrosaccharides, and sugar-alcohols mostly originate from ablation of the phyllosphere off fresh leaf surfaces, pollen, spores, fungi and other biota in soil (Medeiros et al., 2006). Therefore, they are enriched in coarse particles and are more abundant in spring than in winter due to enhanced biological activity and soil resuspension in the warm season. We found that sugars (e.g. glucose, Figs 2d–f) and sugar-alcohols (e.g. arabitol and mannitol) in the fine mode abundantly existed in the urban aerosols, but diminished in the mountain air and almost disappeared in the marine atmosphere. Such a phenomenon on the primary WSOCs that exist abundantly in fine particles has broadly been observed at many locations in the world (Carvalho et al., 2003; Ion et al., 2005; Yttri et al., 2007; Kourtchev et al., 2008). However, the origins of glucose and related sugars and sugar-alcohols in fine particles are unclear, and they may be formed via secondary pathways (Pun et al., 1999;

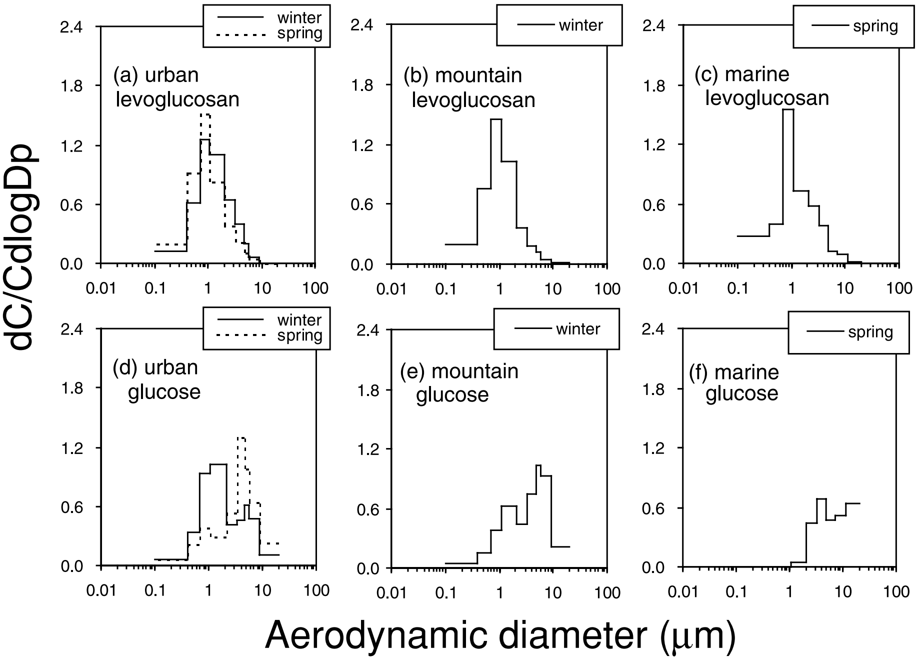

Fig. 2. Size distributions of levoglucosan and glucose in the urban, mountain and marine atmosphere.

Carvalho et al., 2003) or from uncombusted materials (Medeiros et al., 2006).

3.2. Compositions and size distributions of secondary WSOCs

In general, oxalic acid is the dominant aliphatic diacid species in the atmosphere, followed by malonic (C_3), succinic (C_4), glutaric (C_5) and azelaic (C_9) acids (Kawamura and Yasui, 2005; Wang et al., 2006). Oxalic and malonic acids are excluded in the current study because of the limitation of the TMS method mentioned earlier. Thirteen organic acids in total were quantified in the size-resolved samples (Table 2). These acids in the atmosphere are mostly derived from photochemical oxidation of organic precursors although a minor fraction of them can be produced directly by biomass and fossil fuel combustion, and grouped as secondary organic compounds (Table 2). Among the measured secondary organic aerosols (SOA) in the urban and mountaintop air, *o*-phthalic acid showed the highest concentration during winter and spring, followed by succinic, malic, azelaic and glutaric acids. *o*-Phthalic acid is a photo-oxidation product of aromatic compounds such as naphthalene and *o*-xylene (Kawamura and Ikushima, 1993). Such a molecular composition of SOA at the urban and mountain sites indicates a significant pollution caused by anthropogenic activities.

However, the composition of the measured secondary WSOCs is different in the marine atmosphere, where malic and succinic acids are dominant, being two times more abundant than *o*-phthalic acid (Table 2). Malic acid is believed to originate from the oxidation of unsaturated fatty acids (Kawamura and Ikushima, 1993; Rogge et al., 1993; Claeys et al., 2004), whereas succinic acid is efficiently produced by cyclopentene oxidation (Chebbi and Carlier, 1996). Therefore, such a difference in the SOA compositions between the continental and marine samples may suggest a significant dilution of anthropogenic pollutants and a continuous photochemical oxidation of organic precursors to produce malic and succinic acids during the long-range transport of organic precursors from inland China to the North Pacific. Glyceric and malic acids in all the size-segregated samples showed a strong correlation each other (Fig. 3). Such a site- and size-independent linear correlation indicates that both compounds may be produced via similar photochemical pathways.

In a previous study we found high concentrations of dicarboxylic acids such as succinic and malic acids in the smoke particles from wheat straw burning (Wang et al., 2011). Kawamura and Kaplan (1987) also found low molecular weight (LMW, C_2–C_6) saturated dicarboxylic acids in vehicular exhausts. However, these primary diacids are relatively minor in the atmosphere, and the major fraction of the atmospheric LMW dicarboxylic acids is secondarily formed via photochemical oxidation of gaseous precursors (Kawamura and Ikushima, 1993). Therefore, it is reasonable that glyceric and malic acids showed a unimodal size distribution, peaking at the range of 0.7–1.1 μm, for all the samples (Fig. 4). Size distribution of succinic acid at the three sites is similar to that of glyceric and malic acids except in the urban air during spring, where succinic acid showed an additional minor peak in the coarse mode (Figs 5a–c). This was probably caused by an increased adsorption of gaseous succinic acid onto alkaline particles due to an enhanced soil suspension in the warm season (Yao et al., 2003; Wang et al., 2010). Azelaic acid displayed different patterns among the three sample types; the urban samples showed a unimodal distribution with a peak at 0.7–1.1 μm (Fig. 5d), the mountain samples had a bimodal pattern with a major peak at the range 0.7–1.1 μm and a minor peak at the range 4.7–5.8 μm (Fig. 5e), and the marine samples presented a bimodal distribution with two equivalent peaks at the sizes of 0.7–1.1 and 3.3–4.7 μm (Fig. 5f). Other marine

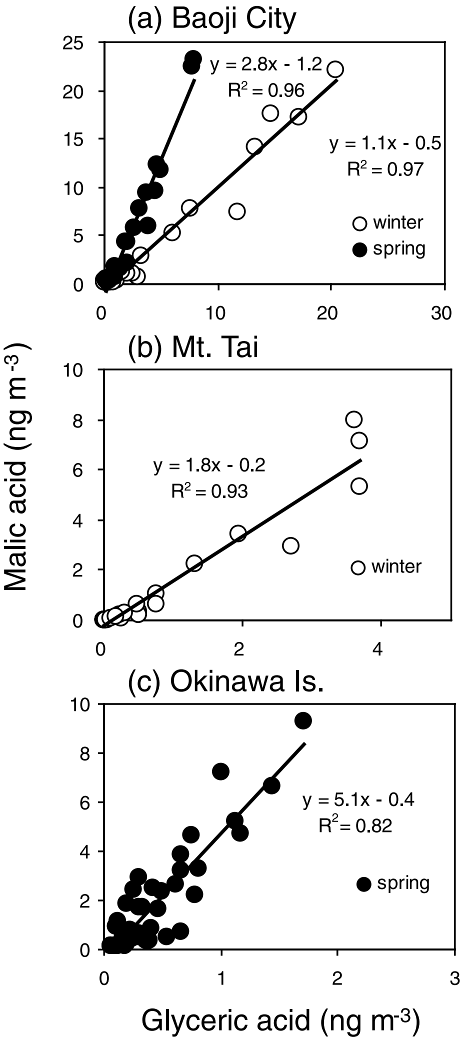

Fig. 3. Linear correlation of glyceric and malic acids in all the size-segregated particles from the urban, mountain and marine atmosphere.

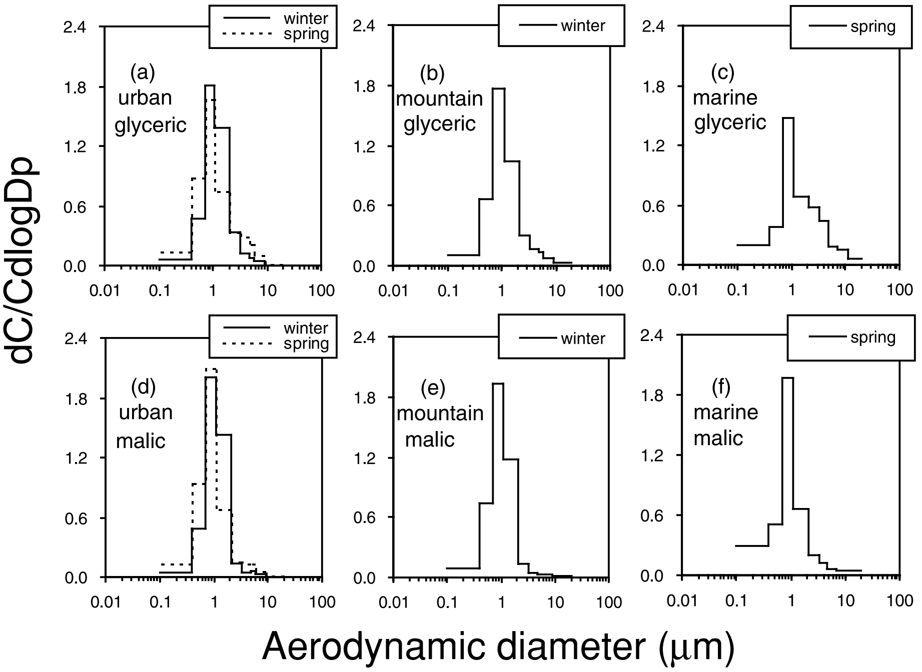

Fig. 4. Size distributions of glyceric and malic acids in the urban, mountain and marine atmosphere.

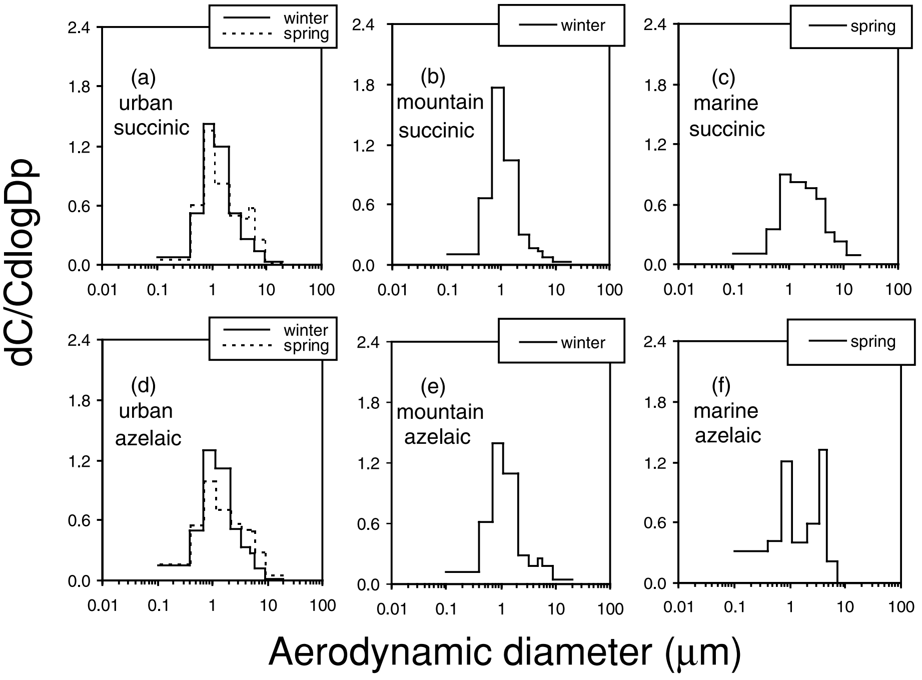

Fig. 5. Size distributions of succinic and azelaic acids in the urban, mountain and marine atmosphere.

aerosol studies also found a similar bimodal pattern for azelaic acid in the North Pacific atmosphere (Mochida et al., 2007; Miyazaki et al., 2010). Azelaic acid is believed to be the photo-oxidation product of unsaturated fatty acid $C_{18:1}$ (Kawamura and Gagosian, 1987), which enriches in fine particles in urban area mostly due to cooking emissions and in coarse particles (i.e. phytoplankton) in marine region due to sea wave splashing (Schauer et al., 1996; Simoneit et al., 2004b). Therefore, azelaic acid had a pronounced peak at the coarse fraction for the marine samples compared with the urban and mountain samples (Fig. 5f).

Benzoic, phthalic and salicyclic acids are photo-oxidation products of anthropogenic precursors such as toluene, xylene

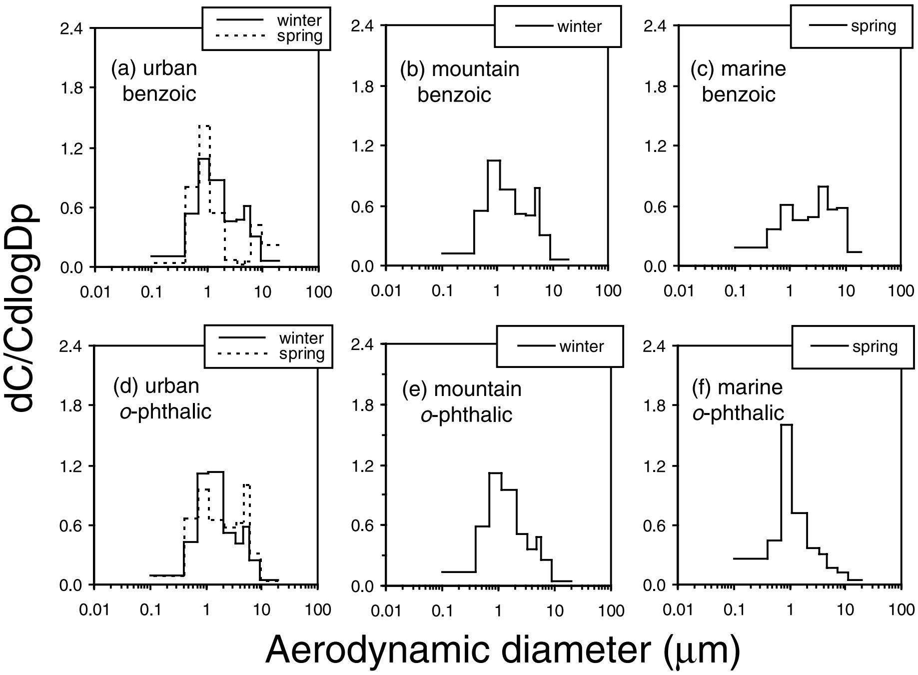

Fig. 6. Size distributions of benzoic and *o*-phthalic acids in the urban, mountain and marine atmosphere.

and naphthalene (Kawamura and Ikushima, 1993; Schauer et al., 1996). These secondarily produced aromatic acids are initially formed as gaseous products and subsequently condensed onto pre-existing particles. On the other hand, some of these gaseous acids may also be absorbed onto coarse particles that contain alkaline metals such as K, Na and Ca (Yao et al., 2003). Therefore, a bimodal size distribution was observed in the urban and mountain atmospheres with a large peak at the range 0.7–1.1 μm and a relatively small peak at the size >3.3 μm (Figs 6a, b, d and e). Compared to the case in winter, a more distinct bimodal pattern in spring was found for benzoic and *o*-phthalic acids at the urban site with a pronounced coarse peak (Figs 6a and d), further indicating an increased adsorption of the aromatic acids onto the alkaline particles due to more soil dust occurring in the warm season. Unlike *o*-phthalic acid, benzoic acid exists largely in gas phase due to its high volatility, thus benzoic acid in fine particles can easily be evaporated into the air and then adsorbed onto coarse particles and/or onto water droplets emitted by sea spray, thus this acid at the marine site moved toward larger size with a maximum in the coarse mode (Fig. 6c), resulting in its fine fraction (0.3 ± 0.1 ng m^{-3}, Table 2) less than its coarse fraction (0.4 ± 0.2 ng m^{-3}). Such an evaporation/condensation effect was also observed in our previous study on PAHs in the three types of the atmosphere (Wang et al., 2009d).

3.3. *Geometric mean diameter of WSOCs*

To further recognize the difference in the size distributions of the WSOCs in the three environments, we calculated the geometric mean diameter (GMD) of the fine (<2.1 μm) and coarse (≥ 2.1 μm) modes and the whole size range for all the measured species (Table 3). In the urban atmosphere, GMDs of levoglucosan and the secondary WSOCs in fine mode are larger in winter than in spring (Table 3). Characteristics of airborne particle size distribution commonly depend on the processes including emissions, condensation, coagulation (Hinds, 1999) and hygroscopic growth (Furutani et al., 2008). Biomass burning for house heating is active during winter in the rural area nearby Baoji city, from which particulate matter emitted are of larger sizes than those from vehicle exhausts (Yang et al., 2004). Moreover, an enhanced condensation of secondarily produced organics due to more abundant gas-phase precursors and an increased coagulation of fine particles due to increased concentration of airborne particulate matter may also be responsible for the wintertime larger GMDs. Both condensation and coagulation rates increase as the increases in the concentrations of gaseous precursors and particles (Herner et al., 2006), and are thus more significant in the urban region than in the mountain and marine areas, resulting in the fine mode GMDs of most WSOCs to be largest in the urban air and smallest in the marine air during the same season (Table 3).

As discussed in Section 3.2, benzoic acid in fine mode may migrate onto coarse mode via the evaporation/adsorption effect. As a result, benzoic acid presented the largest GMD in the whole particle size range compared to other secondary WSOCs. Such a shift toward larger particles is more significant as temperature increases, leading to its GMDs in the total size range larger in spring than in winter at the urban site (Table 3). Levoglucosan

Table 3. Geometric mean diameter (GMD[a], μm) of water-soluble organic compounds in the fine (<2.1 μm) and coarse ($\geq 2.1 \mu$m) particles and the whole range of impactor particles size (total) in the urban, mountain and marine atmosphere over East Asia

	Urban (Baoji, China)						Mountain (Mt. Tai, China)			Marine (Okinawa Is., Japan)		
	Winter ($n = 3$)			Spring ($n = 3$)			Winter ($n = 3$)			Spring ($n = 5$)		
	Fine	Coarse	Total	Fine	Coarse	Total	Fine	Coarse	Total	Fine	Coarse	Total
I. Primary water-soluble organic compounds												
Levoglucosan	0.89 ± 0.03	3.46 ± 0.16	1.21 ± 0.02	0.76 ± 0.04	3.52 ± 0.05	0.94 ± 0.09	0.80 ± 0.07	3.72 ± 0.16	0.97 ± 0.14	0.74 ± 0.08	4.05 ± 0.63	1.17 ± 0.23
Fructose	0.63 ± 0.37	6.20 ± 1.01	2.80 ± 1.78	0.68 ± 0.03	8.54 ± 1.05	2.66 ± 0.78	1.04 ± 0.55	7.61 ± 0.99	3.91 ± 1.95	Nd[b]	14.2 ± 0.82	14.2 ± 0.82
Glucose	1.00 ± 0.03	5.74± 1.23	2.06 ± 0.55	0.81 ± 0.09	6.47 ± 0.42	3.93 ± 0.68	0.98 ± 0.26	6.67 ± 0.37	3.46 ± 1.12	Nd[b]	9.89 ± 0.60	9.89 ± 0.60
Sucrose	0.99 ± 0.04	7.08 ± 0.49	3.85 ± 1.45	0.82 ± 0.10	11.0 ± 0.14	9.92 ± 0.58	0.69 ± 0.30	7.38 ± 0.21	5.01 ± 1.42	Nd[b]	14.7 ± 0.49	14.7 ± 0.49
Trehalose	Nd[b]	6.01 ± 0.88	5.26 ± 1.77	Nd[b]	5.76 ± 0.51	5.63 ± 0.49	Nd[b]	6.86 ± 0.17	6.69 ± 0.46	Nd[b]	10.7 ± 1.93	10.7 ± 1.93
Arabitol	1.01 ± 0.06	4.72 ± 1.03	1.67 ± 0.31	0.87 ± 0.04	6.20 ± 0.45	4.45 ± 0.59	0.88 ± 0.15	6.78 ± 0.73	4.37 ± 0.95	Nd[b]	7.76 ± 1.20	7.76 ± 1.20
Mannitol	0.97 ± 0.07	5.36 ± 0.96	2.35 ± 0.68	0.81 ± 0.05	5.71 ± 0.14	4.72 ± 0.30	Nd[b]	7.18 ± 1.30	7.18 ± 1.30	Nd[b]	7.65 ± 1.32	7.65 ± 1.32
Inositol	0.82 ± 0.06	5.15 ± 0.96	1.11 ± 0.05	0.67 ± 0.05	7.56 ± 0.44	1.32 ± 0.02	0.89 ± 0.61	6.44 ± 1.06	2.73 ± 1.66	Nd[b]	12.2 ± 0.87	12.2 ± 0.87
Glycerol	0.88 ± 0.05	5.20 ± 1.64	1.25 ± 0.12	0.72 ± 0.05	5.58 ± 0.28	1.39 ± 0.32	0.66 ± 0.06	7.14 ± 0.93	1.77 ± 0.26	1.02 ± 0.52	7.57 ± 1.17	5.76 ± 1.19
II. Secondary water-soluble organic compounds												
Glyceric acid	1.04 ± 0.07	3.41 ± 0.58	1.22 ± 0.10	0.80 ± 0.05	4.28 ± 0.19	1.05 ± 0.13	0.80 ± 0.20	4.60 ± 1.72	1.04 ± 0.26	0.80 ± 0.10	5.18 ± 0.38	1.52 ± 0.51
Malic acid	1.04 ± 0.07	3.96 ± 0.94	1.18 ± 0.13	0.79 ± 0.02	4.76 ± 0.19	0.93 ± 0.10	0.80 ± 0.23	4.77 ± 1.95	0.90 ± 0.22	0.71 ± 0.06	6.51 ± 0.92	1.03 ± 0.34
Succinic acid	0.99 ± 0.07	4.26 ± 0.20	1.35 ± 0.07	0.90 ± 0.05	4.67 ± 0.31	1.50 ± 0.23	0.88 ± 0.12	5.42 ± 0.37	1.52 ± 0.30	0.88 ± 0.05	5.27 ± 0.27	2.01 ± 0.38
Glutaric acid	1.01 ± 0.07	4.18 ± 0.23	1.34 ± 0.07	0.87 ± 0.06	4.92 ± 0.16	1.61 ± 0.21	0.85 ± 0.13	5.70 ± 0.15	1.64 ± 0.34	0.80 ± 0.22	5.19 ± 0.17	2.57 ± 0.64
Azelaic acid	0.90 ± 0.08	4.01 ± 0.32	1.24 ± 0.05	0.76 ± 0.03	5.13 ± 0.23	1.52 ± 0.06	0.83 ± 0.15	5.48 ± 0.86	1.17 ± 0.31	0.61 ± 0.10	3.54 ± 0.22	1.12 ± 0.15
Maleic acid	0.88 ± 0.07	4.79 ± 0.53	1.59 ± 0.06	0.78 ± 0.05	4.32 ± 0.47	1.53 ± 0.26	0.79 ± 0.17	4.87 ± 0.35	1.46 ± 0.14	0.80 ± 0.10	4.78 ± 0.60	1.80 ± 0.21
Fumaric acid	1.00 ± 0.03	3.91 ± 0.47	1.24 ± 0.07	0.81 ± 0.04	4.52 ± 0.05	1.31 ± 0.18	0.83 ± 0.16	5.61 ± 0.54	1.30 ± 0.31	0.75 ± 0.04	4.99 ± 0.34	1.48 ± 0.40
Benzoic acid	0.85 ± 0.02	5.46 ± 0.55	1.67 ± 0.18	0.72 ± 0.02	6.69 ± 0.70	2.03 ± 0.25	0.78 ± 0.06	5.32 ± 0.25	1.68 ± 0.07	0.67 ± 0.06	6.31 ± 0.11	2.28 ± 0.25
o-Phthalic acid	0.97 ± 0.04	4.75 ± 0.27	1.59 ± 0.06	0.81 ± 0.07	4.72 ± 0.69	1.67 ± 0.40	0.80 ± 0.10	5.13 ± 0.65	1.57 ± 0.36	0.74 ± 0.03	5.11 ± 0.54	1.24 ± 0.28
m-Phthalic acid	1.01 ± 0.03	3.76 ± 0.43	1.23 ± 0.08	0.81 ± 0.09	4.19 ± 0.14	1.14 ± 0.23	0.88 ± 0.13	5.34 ± 0.64	1.23 ± 0.31	0.77 ± 0.03	5.72 ± 1.17	1.59 ± 0.68
p-Phthalic acid	1.09 ± 0.05	3.62 ± 0.22	1.64 ± 0.04	0.99 ± 0.05	3.64 ± 0.11	1.46 ± 0.12	0.97 ± 0.11	3.65 ± 0.10	1.29 ±0 .14	0.89 ± 0.07	4.41 ± 0.82	1.32 ± 0.21
m-Salicyclic acid	0.97 ± 0.03	3.61 ± 0.18	1.23 ± 0.03	0.74 ± 0.12	4.41 ± 0.15	1.17 ± 0.11	0.86 ± 0.09	4.55 ± 0.43	1.13 ± 0.20	0.74 ± 0.03	5.18 ± 0.29	1.52 ± 0.44
p-Salicyclic acid	0.98 ± 0.02	3.51 ± 0.52	1.45 ± 0.25	0.73 ± 0.10	4.78 ± 0.08	1.28 ± 0.06	0.84 ± 0.11	4.56 ± 0.46	1.07 ± 0.18	0.73 ± 0.04	6.52 ± 0.48	1.90 ± 0.28

[a] $\log \mathrm{GMD} = (\sum C_i \log Dp_i)/ \sum C_i$, where C_i is the concentration of compound in size i and Dp_i is the geometric mean particle diameter (Hinds, 1999).

[b] Nd, not determined.

and the secondary WSOCs except for azelaic and benzoic acids showed the largest GMDs values in the coarse mode at the marine site and the smallest values at the urban site, which is likely caused by an enhanced hygroscopic growth of the particles due to higher humidity in the marine environment in addition to the difference in sources. Furthermore, levoglucosan and the secondary WSOCs showed an increase in concentration ratios of coarse fraction ($\geq$2.1 μm) to fine fraction ($<$2.1 μm) from 0.4 ± 0.2 in the urban air to 0.6 ± 0.4 in the marine air in spring, again suggesting the hygroscopic growth of particles due to high humidity of the marine atmosphere.

4. Summary and conclusion

Molecular compositions and size distributions of primary (i.e. sugars and sugar-alcohol) and secondary (i.e. dicarboxlic acids and related compounds) WSOCs were characterized in the urban, mountain and marine aerosols over East Asia. The measured WSOCs in the urban and mountain areas are dominated by levoglucosan. In contrast, these WSOCs in the marine air are dominated by malic and succinic acids, suggesting a continuous photo-oxidation of organic precursors to produce SOA. Sugars and sugar-alcohols excluding levoglucosan in the urban and mountain atmospheres presented a bimodal size distribution (i.e. accumulation mode and coarse mode) with two peaks at the sizes of 0.7–2.1 and $>$3.1 μm, being in contrast to a unimodal size distribution (i.e. coarse mode) of these primary WSOCs found in the marine air with a peak at the range larger than 3.1 μm. Levoglucosan and all the secondary WSOCs, except for azelaic and benzoic acids, were dominated in fine particles for all the aerosol samples, with a maximum at 0.7–2.1 μm.

Fine mode GMDs of levoglucosan and all the secondary WSOCs are larger in the urban air than in the mountain and marine air, and also larger in winter than in spring, which is, in addition to the difference in sources, probably caused by the enhancements in condensation of secondary products and coagulation of fine particles in the urban air especially in winter due to more abundant gaseous precursors and particulate matters. Levoglucosan and the secondary WSOCs except for azelaic and benzoic acids showed the largest GMDs values in the coarse mode at the marine site and the smallest values at the urban site, highlighting the importance of hygroscopic growth due to high humidity in the marine air, in addition to difference in sources.

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