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# Observation of biogenic secondary organic aerosols in the atmosphere of a mountain site in central China: temperature and relative humidity effects

J. J. Li[1], G. H. Wang[1], J. J. Cao[1], X. M. Wang[2], and R. J. Zhang[3]

[1]State Key Laboratory of Loess and Quaternary Geology, Institute of Earth Environment, Chinese Academy of Sciences, Xi'an 710075, China
[2]State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, China
[3]Key Laboratory of Regional Climate-Environment for Temperate East Asia, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China

Correspondence to: G. Wang (wanggh@ieecas.cn)

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## Abstract

Secondary organic aerosols (SOA) derived from isoprene, pinene and caryophyllene were determined for $PM_{10}$ and size-segregated (9-stage) aerosols collected at the summit (2060 m, a.s.l.) of Mt. Hua, central China during the summer of 2009. Concentrations of estimated isoprene, $\alpha$-/$\beta$-pinene and $\beta$-caryophyllene derived SOC are $81\pm53$, $29\pm14$ and $98\pm53\,\mathrm{ng\,m^{-3}}$, accounting for $2.7\pm1.0\,\%$, $0.8\pm0.2\,\%$ and $2.1\pm1.0\,\%$ of OC, respectively. Concentrations of biogenic (BSOA, the isoprene/pinene/caryophyllene oxidation products) and anthropogenic (ASOA, mainly aromatic acids) SOA positively correlated with temperature ($R = 0.57 - 0.90$). However, a decreasing trend of BSOA concentration with an increase in relative humidity (RH) was observed during the sampling period, although a clear trend between ASOA and RH was not found. Based on the AIM Model calculation, we found that during the sampling period an increase in RH resulted in a decrease in the aerosol acidity and thus reduced the effect of acid-catalysis on BSOA formation. Size distribution measurement showed that most of the determined isoprene derived SOA may form in aerosol phase and enriched in the fine mode ($< 2.1\,\mu m$). 3-Hydroxyglutaric acid, 3-methyl-1,2,3-butanetricarboxylic acid and $\beta$-caryophyllinic acid are only presented in fine particles. However, *cis*-pinonic acid presents a large peak in the coarse mode ($> 2.1\,\mu m$) due to its highly volatile nature.

## 1 Introduction

Volatile organic compounds (VOCs) produced from the biosphere have a substantial impact on the atmospheric chemistry. On a global scale, biogenic volatile organic compounds (BVOCs, $1150\,\mathrm{Tg\,yr^{-1}}$), mostly consisting of isoprene, monoterpenes (such as $\alpha$-/$\beta$-pinene) and sesquiterpenes (such as $\beta$-caryophyllene), are one order of magnitude more abundant than anthropogenic VOCs (Guenther et al., 2006). The high emission of BVOCs can significantly increase the occurrence of secondary organic aerosols (SOA) in the atmosphere, which influences the atmospheric radiation budget directly

by absorbing and scattering sun light, and indirectly by acting as cloud condensation nuclei (CCN) (Kanakidou et al., 2005; Kawamura et al., 2005; Turpin and Huntzicker, 1995). Therefore, numerous studies in the last decade have been devoted to characterizing biogenic secondary organic aerosol (BSOA) in the atmosphere (Kleindienst, 2009; Froyd et al., 2010; Jaoui et al., 2008; Offenberg et al., 2009; Claeys et al., 2004).

No other country in the world is as large and diverse a source of aerosols and trace gases as China. Biogenic emission is high in some forested areas of the country. For example, a model study showed that monthly average emission rates of isoprene are more than $30\,\mathrm{mg\,m^{-2}\,day^{-1}}$ in southeastern, northeastern and central China in summer (Guenther et al., 2006). On the other hand, air pollution in China is serious (Fang et al., 2009; Wang et al., 2006). North China Plain, Guanzhong Basin, and Sichuan Basin in the country are the three most heavily polluted regions in the world, where the annual level of $PM_{2.5}$ on the ground surface was more than $80\,\mathrm{\mu g\,m^{-3}}$ during 2001–2006 (van Donkelaar et al., 2010). In the past decade, SOA derived from anthropogenic sources in China has been given much attention. However, BSOA in the country has only been documented by a limited number of studies (Fu et al., 2010; Wang et al., 2008; Ding et al., 2011; Hu et al., 2008) with no reports for BSOA from the central part. Atmospheric environment over mountain area is unique because of lower temperature, higher humidity and stronger solar radiation, and thus chemical and physical properties of mountain aerosols differ from those on lowlands (e.g., urban area). Furthermore, mountain area is a receptor for anthropogenic gas and aerosols originating from lowlands by long-range transport. Thus characterizations of alpine BSOA can improve our understanding on physicochemical properties of aerosols in the atmosphere.

In the current paper, we first investigate molecular composition of SOA, including those derived from isoprene, pinene and caryophyllene, in airborne particles at the summit (2060 m altitude) of Mt. Hua in central China and their relationship with temperature and relative humidity (RH), and then measure their size distributions to investigate sources and formation mechanisms of BSOA in the alpine atmosphere.

## 2 Experiment section

### 2.1 Aerosols sampling

Mt. Hua (34°29′ N, 110°05′ E, 2060 m a.s.l., Fig. 1) is located in the east border of Guanzhong Plain, central China. It is part of the Qin Ling Mountain Range, where around 3000 species of vegetation are known to grow in the region. The low-elevation forests of the foothills are dominated by temperate deciduous trees. At the middle elevations, conifers are mixed with broadleaf birch, oak and hornbeam. However, subalpine forests of fir, cunninghamia, and birch are dominated at high-elevation forests (http://en.wikipedia.org/wiki/Qin_Ling). Collections of $PM_{10}$ and size-segregated particles were simultaneously performed at the summit of Mt. Hua during the summer of 2009 (15 July ∼ 25 August). A medium-volume air sampler (KC-120H, made in China) was operated for $PM_{10}$ collection at an airflow rate of $100 \, L \, min^{-1}$. Size-segregated samples were collected using an Andersen 9-stage air sampler (Thermo electronic Company, USA) with the cutoff points at 9.0, 5.8, 4.7, 3.3, 2.1, 1.1, 0.7, and 0.4 μm under an airflow rate of $28.3 \, L \, min^{-1}$. A total of 34 $PM_{10}$ samples were collected each lasting for 24 h. The size-resolved aerosols (9 samples per set, 3 sets) were collected for 5–6 days in each set. All samples were collected on pre-baked (450 °C for 8 h) quartz microfiber filters (Whatman 42). After sampling, the filter was sealed in an aluminum bag and stored at −18 °C prior to analysis. Three field blank samples were collected before and after sampling by mounting the filters onto the sampler for about 10 min without sucking any air.

### 2.2 Sample extraction, derivatization and GC/MS quantification

Detailed methods for extraction, derivatization and gas chromatography/mass spectrometry (GC/MS) analysis were described elsewhere (Wang et al., 2006, 2009). Briefly, one fourth of the sample/blank filter was cut in pieces and extracted with a mixture of dichloromethane and methanol (2 : 1, $v/v$) under ultrasonication (3 times and

15 min per time). The extracts were concentrated using a rotary evaporator under a vacuum condition and then blown down to dryness using pure nitrogen. After reaction with a mixture of N,O-bis-(trimethylsilyl) trifluoroacetamide (BSTFA) and pyridine (5 : 1, $v/v$) at 70 °C for 3 h, the derivatives were determined using a GC/MS technique below.

Gas chromatography–mass spectrometry (GC/MS) analysis of the derivatized fraction was performed using an Agilent 7890A GC coupled with an Agilent 5975C 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 a final isothermal holds 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 Daltons using electron impact (EI) mode at 70 eV. Detailed chemical characterization and possible sources of the 15 detected organic compounds in this study are listed in Table 1. In addition, GC/MS response factors of *cis*-pinonic acid, succinic acid, glutaric acid, malic acid, phthalic acids, levoglucosan and arabitol were determined using authentic standards, while GC/MS response factors of 2-methylglyceric acid (MGA), 2-methyltetrols (MT/ME), 3-hydroxyglutaric acid (HGA), 3-methyl-1,2,3-butanetricarboxylic acid (MBTCA) and $\beta$-caryophyllinic aicd (PA) were replaced by those of glyceric acid, erythritol, tartaric acid, suberic acid and *cis*-pinic acid, because the authentic standards are not commercially available. Average recoveries of the target compounds were better than 70 %. No significant contamination ($< 10\%$ of those in the samples) was found in the blanks. Data presented were corrected for the field blanks but not corrected for the recoveries.

OC and EC in the $PM_{10}$ samples were analyzed using DRI Model 2001 Carbon analyzer following the Interagency Monitoring of Protected Visual Environments (IMPROVE) thermal/optical reflectance (TOR) protocol (Chow et al., 2004). Inorganic ions including $SO_4^{2-}$, $NO_3^-$ and $NH_4^+$ in $PM_{10}$ and size-resolved samples were analyzed using ion chromatography (Li et al., 2011; Shen et al., 2008). Particle in-situ pH ($pH_{IS}$) and aerosol liquid water content (LMC) were calculated by Aerosol Inorganic Model (AIM) using a $SO_4^{2-}$-$NO_3^-$-$NH_4^+$-$H^+$ system (AIM-II) (Clegg et al., 1998a,b)

(http://www.aim.env.uea.ac.uk/aim/aim.php). These data are used in the current study to explore the sources and formation mechanisms of BSOA.

## 3   Results and discussion

### 3.1   Abundance of OC, EC and SOA in $PM_{10}$

#### 3.1.1   Overall results

Concentrations of detected organic compounds, OC, EC and inorganic ions are summarized in Table 2. $PM_{10}$, OC and EC concentrations at Mt. Hua in summer were $43 \pm 17$, $3.5 \pm 1.3$ and $0.65 \pm 0.27\,\mu g\,m^{-3}$, respectively, and lower than those in winter ($54 \pm 24$, $5.9 \pm 2.5$ and $0.9 \pm 0.6\,\mu g\,m^{-3}$) (Li et al., 2012) mainly due to decreased emissions of biomass and coal burning for house heating. However, the favorable meteorological conditions (i.e., higher temperature and RH, and increased boundary layer height) resulted in higher concentrations of $SO_4^{2-}$ and $NH_4^+$ ($21\pm12$ and $4.3\pm2.5\,\mu g\,m^{-3}$, respectively) in summer compared to those ($5.8\pm3.6$ and $1.6\pm0.9\,\mu g\,m^{-3}$, respectively) in winter (Li et al., 2011).

Three compounds were determined as oxidation products of isoprene in the $PM_{10}$ samples, which are 2-methylglyceric acid (MGA, $4.1\pm2.2\,ng\,m^{-3}$), 2-methylthreitol (MT, $3.6\pm2.8\,ng\,m^{-3}$) and 2-methylerythritol (ME, $4.9\pm3.6\,ng\,m^{-3}$) (Table 2). *cis*-Pinonic acid (PA), 3-hydroxyglutaric acid (HGA) and 3-methyl-1,2,3-butanetricarboxylic acid (MT-BCA) are the major photooxidized products of $\alpha$-/$\beta$-pinene, and their concentrations are $3.0 \pm 1.4$, $2.1 \pm 1.1$ and $1.6 \pm 1.1\,ng\,m^{-3}$, respectively. $\beta$-Caryophyllinic acid, one of $\beta$-caryophyllene (a sesquiterpene) oxidation products, was also determined in this study, and its concentration is $2.2\pm1.2\,ng\,m^{-3}$. Contributions of BVOCs to secondary organic carbon (SOC) in the atmosphere of Mt. Hua were estimated using a tracer-based method reported by Kleindienst et al. (2007). Concentrations of estimated isoprene, $\alpha$-

/$\beta$-pinene and $\beta$-caryophyllene derived SOC are $81 \pm 53$, $29 \pm 14$ and $98 \pm 53 \, \mathrm{ng \, C \, m^{-3}}$, accounting for $2.7 \pm 1.0 \%$, $0.8 \pm 0.2 \%$ and $2.1 \pm 1.0 \%$ of OC, respectively.

Succinic, glutaric and malic acids in the atmosphere are produced mainly by photochemical reactions of unsaturated hydrocarbons and fatty acids from biomass burning and coal combustion (Kawamura and Yasui, 2005; Wang et al., 2010). Concentrations ($2.2$–$5.8 \, \mathrm{ng \, m^{-3}}$) of these compounds in the Mt. Hua $PM_{10}$ samples are comparable to the BSOA above. Phthalic acids are derived from photooxidation of polycyclic aromatic hydrocarbons (PAHs) (Wang et al., 2007; Simoneit et al., 2004b), and pyrolysis of plastic materials (Kawamura and Pavuluri, 2010). Their concentrations in the Mt. Hua samples are $0.16$–$4.9 \, \mathrm{ng \, m^{-3}}$. Levoglucosan and arabitol are tracers of biomass burning and biological emissions, respectively (Simoneit et al., 2004a; Engling et al., 2009). Levoglucosan is lower in summer ($14 \pm 12 \, \mathrm{ng \, m^{-3}}$) than in winter ($65 \pm 30 \, \mathrm{ng \, m^{-3}}$), in contrast to arabitol ($5.5 \pm 3.5 \, \mathrm{ng \, m^{-3}}$ in summer and $3.0 \pm 2.0 \, \mathrm{ng \, m^{-3}}$ in winter), which is higher in summer (Li et al., 2012), indicating a decreased biomass burning and an increased biogenic activity in the hot season.

### 3.1.2 Comparison between different air masses classified by back-trajectories analysis

As shown in Fig. 1, air masses reaching the sampling site during the campaign were from three directions: southerly, easterly and northerly. Thus, concentrations of total detected compounds, temperatures and RH during the summer campaign are categorized into the three groups (Table 2). Although $PM_{10}$ concentration ($46 \pm 6.6 \, \mathrm{\mu g \, m^{-3}}$, Table 2) from northerly is slightly higher than those ($42 \pm 17 \, \mathrm{\mu g \, m^{-3}}$, Table 2) from the southerly and easterly, $SO_4^{2-}$, $NH_4^+$ and $o$-phthalic acid in the samples showed higher concentrations when air mass was transported from the south and east directions, indicating more anthropogenic pollutants in these regions. As shown in Table 2, nearly all the BSOA tracers presented higher concentrations for the southerly and easterly samples than those for the northerly samples, which is reasonable as plant vegeta-

tion is much more abundant in southern and eastern China (Guenther et al., 2006). Furthermore, the higher concentration of $SO_4^{2-}$ and $NO_3^-$ can also enhance the BSOA formation by an acid-catalyzed reaction (Surratt et al., 2010).

### 3.1.3  Comparison of BSOA tracers with other studies

A comparison of concentration and diagnostic ratio of isoprene, $\alpha$-/$\beta$-pinene and $\beta$-caryophyllene products was shown in Table 3. In this study, aerosol samples are collected at a summit of Mt. Hua (2060 m a.s.l), where vegetation is very limited and the ground surfaces are mostly rocks. Thus, BSOA tracers present lowest concentration in the elevated region than those in other areas except Alert (82.5° N, 62.3° W) in the Canadian High Arctic. All the tracers at the Alert site are 1–3 orders of magnitude lower than those in other regions. 2-Methyltetrols (sum of 2-methylthreitol and 2-methylerythritol) are the major products of isoprene (Kleindienst et al., 2006; Edney et al., 2005; Claeys et al., 2010), thus their contribution to organic carbon of airborne particles can be used to roughly compare the variation of isoprene emission. The ratios present a decreasing trend along with an increase in latitude, highest at the equatorial region (Balbina, Brazil, 1.8 %) (Claeys et al., 2004) and lowest at the Arctic site (Alert, Canada 0.019 %) (Fu et al., 2009). The carbon contribution of 2-methyltrols in Mt. Hua (0.11 ± 0.06 %) is lower than those (0.17–0.75 %, Table 3) in other Chinese regions excluding Wangqingsha and Chongming, which can be explained by lower levels of biogenic precursors due to less abundant vegetation in central and western China compared to the eastern part of the country (Guenther et al., 2006). However, the lowest ratios in Wangqingsha (0.070 %) and Chongming (0.020 %) most likely resulted from the high anthropogenic OC input since both sites are close to the urban areas.

MBTCA is aged biogenic product from gas-phase oxidation of *cis*-pinonic acid (PA) by OH (Szmigielski et al., 2007; Henry and Donahue, 2012). Thus the ratio of MBTCA/PA ($R_{MBTCA/PA}$) is indicative of aerosol ageing (Kulmala et al., 2011). The ratios (0.54 and 1., Table 3) at Mt. Hua and Mt. Tai are higher than that (0.32, Table 3) at Wangqingsha, a lowland rural site in southern China, indicating the mountain aerosols

are more oxidized. Such a phenomenon may be related to the stronger mountaintop radiation that causes the BSOA being highly oxidized. Averaged ratios of MBTCA/PA ratios are 0.71±0.27, 0.43±0.31 and 0.29±0.12 for the southerly, easterly and northerly samples, respectively, indicating that aerosols derived from the southerly air masses were more aged. The Mexico Campaign reported that MBTCA/PA (4.8, Table 3) in the rural area is nearly 5 times higher than that (0.91) in urban region due to more aged BSOA (Stone et al., 2010), which is consistent with the larger contributions ($25 \pm 5$ %) of aged low-MW acids (mainly oxalic acid) to WSOC at the peripheral rural site than that at urban site ($8.2 \pm 3.4$ %) (Stone et al., 2010). Concentration of MBTCA at the Arctic site is almost one order of magnitude higher than PA ($R_{\text{MBTCA/PA}}$ = 9.4 at Alert, Table 3). Such a spatial variation in the ratios demonstrates a continuous oxidation of BSOA during transport.

## 3.2   Calculations of particle in-situ pH and liquid water content

Chemical reactions in aerosol phase or gas-particle distributions in aerosol surface are much linked to the actual pH (i.e., in-situ pH, $\text{pH}_{\text{IS}}$) in the aqueous phase and liquid water content (LWC) of particles (Xue et al., 2011). Thus, aerosol in-situ pH and liquid water content are calculated in this study using the following equation:

$$\text{pH}_{\text{IS}} = -\log \alpha_{\text{H}^+} = -\log(\gamma_{\text{H}^+} \times n_{\text{H}^+} \times 1000/V_{\text{a}}) \tag{1}$$

Where $\alpha_{\text{H}^+}$ is activity of $\text{H}^+$ in $\text{mol L}^{-1}$ in the aqueous phase on the particle, $\gamma_{\text{H}^+}$ is the activity coefficient of $\text{H}^+$, $n_{\text{H}^+}$ is free $\text{H}^+$ in the unit of $\text{mol m}^{-3}$ of air, and $V_{\text{a}}$ is volume concentration of the aqueous phase of aerosol in ambient atmosphere in the unit of $\text{cm}^3 \text{m}^{-3}$. $\gamma_{\text{H}^+}, n_{\text{H}^+}$ and $V_{\text{a}}$ as well as LWC ($\mu\text{mol m}^{-3}$) are derived using Aerosol Inorganic Model (AIM) (Clegg et al., 1998b,a). As $SO_4^{2-}$, $NO_3^-$ and $NH_4^+$ are detected in this study, we chose AIM-II model for calculation, which considers a $SO_4^{2-}$-$NO_3$-$NH_4^+$-$H^+$ system and allows variable temperature and relative humidity. As shown in Table 2, $\text{pH}_{\text{IS}}$ are $-0.08 \pm 0.64$ in $PM_{10}$ at Mt. Hua, which is comparable with those in $PM_{2.5}$ samples in Hong Kong ($-0.08 \pm 0.81$ during the summer of

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2009) (Xue et al., 2011) but higher than those at Qinghai Lake ($-1.20 \pm 0.32$ during the summer of 2010) (Li et al., 2013), a highland site with a distance of around 1000 km west to Mt. Hua. It is worth noting that in-situ acidity of the Mt. Hua $PM_{10}$ samples may be somewhat overestimated due to the relatively higher level of mineral ions (e.g., $Ca^{2+}$ and $Mg^{2+}$) in the samples, which were not included by the model. LWC of $PM_{10}$ at Mt. Hua are $4.87 \pm 4.90\,\mu mol\,m^{-3}$, and much larger than those of $PM_{2.5}$ in Hong Kong ($0.39 \pm 0.12\,\mu mol\,m^{-3}$) (Xue et al., 2011), probably suggesting that the mountain aerosols are more hygroscopic.

## 3.3 Effects of temperature on BSOA formation

Temporal variations of BSOA tracers, OC and EC in $PM_{10}$ and meteorological conditions are shown in Fig. 3. The sharp declines during the rainy days suggest a significant scavenging effect of wet deposition. The variation of the seven BSOA tracers exhibits the same pattern, because they are formed via similar pathways.

The relationships between BSOA and meteorological parameters are plotted in Fig. 4a–l according to the three different classes of air masses. Nearly all of the detected BSOA tracers showed a robust linear relation with ambient temperature, largely due to enhancements in BVOCs emission and BSOA production under higher temperature. However, the temperature dependence of gas-particle partitioning of the compounds may be relatively weaker. For example, the temperature dependence of gas-particle partitioning of pinonic acid, a highly volatile compound, can be approximated by an Arrhenius activation energy of $\sim 0.21\,kJ\,mol^{-1}$ (Zhang et al., 2010). Such a temperature dependence is also observed in other areas, such as in the central Pearl River Delta region, Southern China (except MGA) (Ding et al., 2011) and Research Triangle Park, North Carolina, USA (Offenberg et al., 2011). As shown in Fig. 4b, the slopes of 2-methyltetrols in this study are 2.7, 2.0 and 0.79 for the $PM_{10}$ samples from southerly, easterly and northerly air masses, respectively, much lower than that (5.27) (Ding et al., 2011) in southern China. This can be explained by a higher isoprene emission due to

more abundant vegetation in the south lowland region (Guenther et al., 2006). The slopes of the regression equation in the southerly and easterly air masses are higher than those from northerly, which is consistent with plant abundance distributions in the three regions.

## 3.4  Effects of relative humidity on BSOA formation

As far as we know, field observations of the influence of relative humidity (RH) on SOA formation from biogenic VOCs were documented in very limited number. Some chamber studies explored the effects of humidity on BSOA production. For example, Zhang et al. (2011) conducted a chamber study and found that 2-methylglyceric acid and its corresponding oligomers were enhanced in the particle-phase under lower RH condition, in contrast, the 2-methyltetrols did not substantially vary under different RH conditions. However, in this study we found that all of the detected isoprene, $\alpha$-/$\beta$-pinene and $\beta$-caryophyllene showed a significant negative linear correlation with relative humidity (Fig. 4g–l).

In fact, an enhancement effect of aerosol acidity on BSOA formation was found by several chamber and field studies (Ding et al., 2011; Iinuma et al., 2004; Offenberg et al., 2009). Surratt et al. (2010) found that isoprene epoxydiols (IEPOX) are key intermediates of SOA formation from isoprene under low $NO_x$ condition, which are initially formed in the gas-phase and subsequently partitioned into aerosol aqueous-phase. The aerosol phase IEPOX further convert to 2-methyltetrols (MT/ME) and $C_5$-alkene triols, the three most common SOA of isoprene, by an acid-catalyzed ring opening of this epoxydiol and a subsequent nucleophilic addition. Lin et al. (2012) again confirmed that IEPOX reactively uptake onto acidified sulfate aerosols to produce MT and ME as well as other IEPOX-derived SOA tracers, including the $C_5$-alkene triols, 3-methyltetrahydrofuran-3,4-diols, dimers, and organosulfates. However, aerosol in-situ acidity is strongly dependent on relative humidity. Based on the AIM model calculations, we found that relative humidity (RH) of the mountaintop atmosphere exhibits a robust linear correlation with the in-situ pH ($pH_{IS}$) of the $PM_{10}$ particles ($R^2 = 0.95$)

and a moderate correlation with liquid water content of the particles ($R^2 = 0.64$). As shown in Fig. 4m–o, all the calculated SOC concentrations showed significant negative linear correlations with $pH_{IS}$, suggesting that the suppressed effect of RH on BSOA formation is most likely related to reducing aerosol acidity. Similar researches have been performed in the southeastern United States although the AIM model for most samples did not work due to the aerosol either being fully neutralized or the RH being too low in the region (Lin et al., 2013b; Budisulistiorini et al., 2013). Interestingly, Budisulistiorini et al. (2013) found that the IEPOX-OA factor obtained by PMF (positive matrix factorization) analysis of ACSM (Aerosol Chemical Speciation Monitor) data also shows a moderate correlation ($R^2 = 0.25$) with in situ acidity of particles.

Moreover, Zhang et al. (2011) reported that low RH circumstance can improve BSOA yields by promoting particle phase organic esterification process. Nguyen et al. (2011) also pointed out that the suppression of condensation and addition reactions are the most important chemical effects of the increased RH on BSOA formation. However, we found there were no clear correlation between biogenic SOC (BSOC) concentrations and liquid water content (LWC) of the particles (Fig. 4p–r). High LWC is favorable for a conversion of BSOA precursors into aqueous phase and thus enhances BSOA production. On the other hand, however, high LWC may reduce particle acidity, which is unfavorable for BSOA production. Moreover, the effect of RH on precursor emission strength may be another potential factor influencing the BSOA production. In any case, more chamber and field studies are warranted to conduct for understanding the exact mechanism of effect of RH on BSOA formation.

Phthalic acids may be produced by the oxidation of naphthalene and other polycyclic aromatic hydrocarbons (Kawamura et al., 2005; Kawamura and Ikushima, 1993). Similar to BSOA formation, these anthropogenically derived SOA also showed a positive relation with temperature ($R > 0.6$). A clear correlation between RH and concentration of phthalic acids was not observed in this study ($R < 0.1$, $p > 0.7$). However, recent investigations indicated that the yields of SOA derived from some aromatic compounds (such as xylenes and toluene) positively correlated with the particle water content (Ka-

mens et al., 2011; Zhou et al., 2011), emphasizing the important impact of liquid water content in the particles (RH depended) on anthropogenic SOA formation.

## 3.5 Size distribution

To further discuss the source and formation mechanism of the BSOA in the mountain atmosphere, detailed information of their size distribution was investigated. Table 5 shows the concentration and geometric mean diameters (GMD) of the detected organic compositions and inorganic ions in fine ($< 2.1 \, \mu m$) and coarse ($> 2.1 \, \mu m$) modes, and their concentrations as a function of size are plotted in Fig. 5.

2-Methyltetrols present a unimodal size distribution pattern with a peak at the size of 0.7–1.1 μm (Fig. 5b). As mentioned above, IEPOX are key intermediates of isoprene oxidation products of 2-methyltetrols under low-$NO_x$ condition through heterogeneous reactions in aerosol-phase (Surratt et al., 2010; Lin et al., 2012). In contrast, MGA is formed via nucleophilic attack of water on the oxirane ring of 2-methyloxirane-2-carboxylic acid (methacrylic acid epoxide, MAE), which is produced by isoprene oxidation under high-$NO_x$ condition (Lin et al., 2013a). MGA in the Mt. Hua aerosols present a bimodal size distribution pattern with a major peak in the fine mode ($< 2.1 \, \mu m$) and a minor peak in the coarse mode ($> 2.1 \, \mu m$) (Fig. 5a).

*cis*-Pinonic acid (PA) exhibits a bimodal pattern with a large peak in the coarse mode ($> 2.1 \, \mu m$) (Fig. 5c), in contrast to HGA and MBTCA, which only present in the fine mode (Fig. 5d and e). Such distributions are consistent with those in Mainz, Germany (Zhang et al., 2010). PA is formed by a gas-phase oxidation of pinene with ozone and a subsequent adsorption onto pre-existing particles (Hallquist et al., 2009). On the contrary, MBTCA is likely formed in the gas phase by further reactions of *cis*-pinonic acid or *cis*-pinic acid involving participation of OH radical (Jaoui et al., 2005; Szmigielski et al., 2007). The differences in size distributions of PA and MBTCA can be ascribed to the highly volatile nature of *cis*-pinonic acid. Zhang et al. (2010) pointed out that *cis*-pinonic acid predominantly present in the gas phase. Coarse particle are generally basic, because they are in most cases enriched with mineral species. Therefore, *cis*-

pinonic acid in gas-phase may prefer to reactively uptake onto coarse particles. Only a fine mode of $\beta$-caryophyllinic acid (CA) was observed in the mountain atmosphere (Fig. 5f).

Succinic ($C_4^{di}$) and glutaric ($C_5^{di}$) acids are formed mostly from anthropogenic volatile organic compounds, which are firstly oxidized as gaseous keto-carboxylic acids and then partitioned into particle phase followed by a further oxidation as the dicarboxylic acids (Wang et al., 2011), thus both present a bimodal size distribution (Fig. 5g and h). On the contrary, malic acid is found to be mostly produced in biomass combustion process and emitted into the air as fine particles (Wang et al., 2011, 2012), thus malic acid showed the fine mode pattern (Fig. 5i). $o$-Phthalic acid is formed via a gaseous oxidation of naphthalene and followed by a subsequent adsorption/condensation onto pre-existing particles, thus, showed a bimodal pattern (Fig. 5j), in contrast to a unimodal pattern of $p$-phthalic acid (Fig. 5l), which is largely formed by a pyrolysis of plastic materials and emitted into the air as fine particles (Wang et al., 2012). Levoglucosan, a key tracer for biomass burning, presents a unimodal distributing, peaking at a size of 0.7–1.1 µm in summer (Fig. 5m). In contrast, arabitol is dominated in coarse particle as it mostly originates from pollen, spore and other biota (Fig. 5n). $SO_4^{2-}$ and $NH_4^+$ are dominated in the fine mode (Fig. 5o and q), similar to those in winter (Li et al., 2011). However, coarse particles in the mountain atmosphere are more significantly enriched in $NO_3^-$ compared to that in Xi'an (unreported data), a mega-city near Mt. Hua (Fig. 5p). Such a shift to large particles is caused by an evaporation of nitrate from fine particles and a subsequent adsorption onto coarse particles during the air mass transport onto the mountaintop.

## 4  Summary and conclusion

BSOA derived from isoprene, pinene and sesquiterpene were determined for $PM_{10}$ and size-resolved aerosols collected at the mountaintop of Mt. Hua. Back-trajectories analysis showed that all the BSOA tracers present higher concentrations in the sam-

ples from the southerly and easterly than in those from the northerly, indicating higher BVOC emissions and/or more BSOA production. Concentrations of the BSOA in the mountain samples increased with an increase in temperature during the campaign, because higher temperature is favorable for BVOCs emission and subsequent oxida-
tion. In contrast, RH showed significant linear negative correlations with concentrations of all the isoprene, pinene and caryophyllene products. Based on the AIM model calculation, we found that an increase in RH can result in a decrease in particle in-situ acidity, which reduces the effect of acid-catalysis on BSOA formation. However, such a significant relation was not observed for LWC and BSOA. Size distribution details
were also explored for the target compounds. Most of the BSOA is enriched in the fine mode ($< 2.1\,\mu m$) except for PA, which is dominated in the coarse mode ($> 2.1\,\mu m$) due to its highly volatile nature. Size distributions of levoglucosan, arabitol and other secondary aerosols were also investigated. These size distribution details are helpful for recognizing their atmospheric fate including formation, adsorption and evaporation.

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**Table 1.** Chemical character and possible source of detected organic markers.

| Order | Organic marker | Chemical structure | Molecular formula | Molecular weight | $m/z$[a] | Possible source |
|-------|----------------|--------------------|-------------------|------------------|----------|-----------------|
| 1 | Succinic acid ($C_4^{di}$)[b] | | $C_4H_6O_4$ | 118 | 247 | Photochemical product of unsaturated hydrocarbons and fatty acids; direct emission from coal and biomass burning. |
| 2 | 2-Methylglyceric acid (MGA)[b] | | $C_4H_8O_4$ | 120 | 103 | Photochemical product of isoprene. |
| 3 | Glutaric acid ($C_5^{di}$) | | $C_5H_8O_4$ | 132 | 147 | Photochemical product of unsaturated hydrocarbons and fatty acids; direct emission from coal and biomass burning. |
| 4 | Malic acid (MA) | | $C_4H_6O_5$ | 134 | 147 | Photochemical product of unsaturated hydrocarbons and fatty acids; direct emission from coal and biomass burning. |
| 5 | *cis*-Pinonic acid (PA) | | $C_{10}H_{16}O_3$ | 184 | 171 | Photochemical product of monoterpene. |
| 6 | 2-Methylthreitol (MT) | | $C_5H_{12}O_4$ | 136 | 147 | Photochemical product of isoprene. |
| 7 | 2-Methylerythritol (ME) | | $C_5H_{12}O_4$ | 136 | 147 | Photochemical product of isoprene. |
| 8 | 3-Hydorxyglutaric acid (HGA) | | $C_5H_8O_5$ | 148 | 147 | Photochemical product of monoterpene. |
| 9 | *o*-Phthalic acid (*o*-Ph) | | $C_8H_6O_4$ | 166 | 295 | Photochemical product of toluene, xylene, naphthalene and other anthropogenic pollutants. |
| 10 | Levoglucosan (Levo) | | $C_6H_{10}O_5$ | 162 | 217 | Pyrolysis product of materials containing cellulose. |
| 11 | Arabitol (Arab) | | $C_5H_{12}O_5$ | 152 | 217 | Direct emission by microorganism, plants and animals. |
| 12 | 3-Methyl-1,2,3-butane tricarboxylic acid (MBTCA) | | $C_8H_{12}O_6$ | 204 | 147 | Photochemical product of monoterpene. |
| 13 | *m*-Phthalic acid (*m*-Ph) | | $C_8H_6O_4$ | 166 | 295 | Photochemical product of toluene, xylene, naphthalene and other anthropogenic pollutants. |
| 14 | *p*-Phthalic acid (*p*-Ph) | | $C_8H_6O_4$ | 166 | 295 | Direct emission by open buring of new plastic bags, roadside litter and landfill trash; secondary formation. |
| 15 | *β*-Caryophyllinic acid | | $C_{14}H_{22}O_4$ | 254 | 383 | Photochemical product of *β*-caryophyllene. |

[a] Mass-to-charge ratio of fragment ions for qualification and quantification.

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**Table 2.** Concentrations ($ng\,m^{-3}$) of biogenic secondary organic aerosols and other compounds in $PM_{10}$ of Mt. Hua.

| | Southerly ($N = 17$)[a] | Easterly ($N = 11$)[a] | Northerly ($N = 6$)[a] | Average ($N = 34$) |
|---|---|---|---|---|
| $T$ (°C) | 18 ± 2.1 | 16 ± 1.1 | 18 ± 2.2 | 17 ± 2.1 |
| RH (%) | 85 ± 10 | 91 ± 5.0 | 64 ± 13 | 84 ± 13 |
| $PM_{10}$ ($\mu g\,m^{-3}$) | 42 ± 17 | 42 ± 17 | 46 ± 6.6 | 43 ± 17 |
| OC ($\mu g\,m^{-3}$) | 4.0 ± 1.2 | 3.0 ± 1.2 | 3.0 ± 0.87 | 3.5 ± 1.3 |
| EC ($\mu g\,m^{-3}$) | 0.65 ± 0.26 | 0.57 ± 0.20 | 0.78 ± 0.37 | 0.65 ± 0.27 |
| $SO_4^{2-}$ ($\mu g\,m^{-3}$) | 19 ± 9.9 | 23 ± 11 | 15 ± 6.8 | 21 ± 12 |
| $NO_3^-$ ($\mu g\,m^{-3}$) | 1.2 ± 1.7 | 1.5 ± 1.3 | 2.0 ± 0.4 | 1.4 ± 1.4 |
| $NH_4^+$ ($\mu g\,m^{-3}$) | 4.0 ± 2.0 | 5.1 ± 2.5 | 3.1 ± 1.4 | 4.3 ± 2.5 |
| $pH_{IS}$ | 0.0 ± 0.55 | 0.30 ± 0.39 | −1.08 ± 0.47 | −0.08 ± 0.64 |
| $LWC^b$ ($\mu mol\,m^{-3}$) | 4.85 ± 5.27 | 7.01 ± 4.51 | 0.93 ± 0.34 | 4.87 ± 4.90 |
| | | | | |
| **I. Isoprene derived SOA** | | | | |
| 2-methylglyceric acid | 4.7 ± 1.7 | 3.9 ± 2.4 | 2.7 ± 1.7 | 4.1 ± 2.2 |
| 2-methylthreitol | 5.1 ± 2.9 | 2.4 ± 1.8 | 1.5 ± 1.2 | 3.6 ± 2.8 |
| 2-methylerythritol | 6.7 ± 3.5 | 3.7 ± 3.1 | 2.6 ± 1.8 | 4.9 ± 3.6 |
| subtotal | 16 ± 7.6 | 10 ± 6.9 | 6.8 ± 4.7 | 13 ± 8.2 |
| $SOC_{isoprene}$ ($ng\,C\,m^{-3}$)$^c$ | 106 ± 49 | 65 ± 45 | 44 ± 30 | 81 ± 53 |
| | | | | |
| **II. $\alpha$-/$\beta$-pinene derived SOA** | | | | |
| *cis*-pinonic acid | 3.3 ± 1.4 | 2.5 ± 1.1 | 3.1 ± 1.4 | 3.0 ± 1.4 |
| 3-hydorxyglutaric acid | 2.6 ± 1.1 | 1.6 ± 0.78 | 1.4 ± 0.67 | 2.1 ± 1.1 |
| MBTCA | 2.2 ± 1.1 | 1.0 ± 0.64 | 0.88 ± 0.49 | 1.6 ± 1.1 |
| subtotal | 8.1 ± 3.1 | 5.1 ± 2.3 | 5.3 ± 2.4 | 6.6 ± 3.3 |
| $SOC_{pinene}$ ($ng\,C\,m^{-3}$)$^c$ | 35 ± 13 | 22 ± 9.8 | 23 ± 10 | 29 ± 14 |
| | | | | |
| **III. $\beta$-caryophyllene derived SOA** | | | | |
| $\beta$-caryophyllinic acid | 2.7 ± 1.1 | 1.9 ± 1.0 | 2.3 ± 1.6 | 2.2 ± 1.2 |
| $SOC_{caryophyllene}$ ($ng\,C\,m^{-3}$)$^c$ | 116 ± 47 | 81 ± 42 | 98 ± 72 | 98 ± 53 |
| | | | | |
| **IV. Other organic compounds** | | | | |
| Succinic acid | 6.7 ± 3.1 | 6.8 ± 3.5 | 1.6 ± 0.41 | 5.8 ± 3.6 |
| Glutaric acid | 2.4 ± 1.0 | 2.2 ± 1.3 | 0.88 ± 0.31 | 2.2 ± 1.5 |
| Malic acid | 7.3 ± 2.4 | 5.1 ± 2.6 | 2.7 ± 1.5 | 5.8 ± 3.2 |
| *o*-Phthalic acid | 6.2 ± 2.3 | 4.4 ± 2.1 | 2.9 ± 1.5 | 4.9 ± 2.5 |
| *m*-Phthalic acid | 0.18 ± 0.08 | 0.13 ± 0.06 | 0.15 ± 0.08 | 0.16 ± 0.08 |
| *p*-Phthalic acid | 2.7 ± 2.2 | 1.7 ± 1.7 | 3.0 ± 3.0 | 2.3 ± 2.2 |
| Levoglucosan | 17 ± 13 | 12 ± 12 | 14 ± 6.8 | 14 ± 12 |
| Arabitol | 6.1 ± 4.0 | 4.3 ± 2.8 | 5.9 ± 0.63 | 5.5 ± 3.5 |

[a] Mean concentrations of detected compounds in $PM_{10}$ of Mt. Hua from southerly, easterly and northerly air masses based on back-trajectories analysis.
[b] LWC: liquid water content in the aqueous phase of $PM_{10}$.
[c] The total mass concentrations of SOC (secondary organic carbon) produced by isoprene, $\alpha$-/$\beta$-pinene, and $\beta$-caryophyllene were calculated using a tracer-based method reported by Kleindienst et al. (2007).

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**Temperature and relative humidity effects**

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**Table 3.** Comparison of biogenic secondary organic aerosol tracers in the Mt. Hua aerosols with those reported by other studies during summer.

| Location | Latitude (°) | Aerosols type | Isoprene products | $\alpha$-/$\beta$-pinene products | $\beta$-caryophyllene products | 2-methyltetrols -C/OC (%) | $R_{MBTCA/PA}$ | Reference |
|---|---|---|---|---|---|---|---|---|
| Changbai, China | 42°24′ N | PM$_{2.5}$ | 53 | 31 | NA[a] | 0.45 | NA | Wang et al. (2008) |
| Mt. Tai, China | 36°41′ N | TSP | 171 | 30 | 12 | 0.32 | 1.0 | Fu et al. (2010) |
| Mt. Hua, China | 34°29′ N | PM$_{10}$ | 13 | 6.6 | 2.2 | 0.11 | 0.54 | This study |
| Chongming, China | 30°50′ N | PM$_{2.5}$ | 48 | 1.8 | NA | 0.02 | NA | Wang et al. (2008) |
| Dinghu, China | 23°10′ N | PM$_{2.5}$ | 26 | NA | NA | 0.17 | NA | Wang et al. (2008) |
| Wangqingsha, China | 22°42′ N | PM$_{2.5}$ | 31 | 6.6 | 0.54 | 0.07 | 0.32 | Ding et al. (2011) |
| Hong Kong, China | 22°13′ N | PM$_{2.5}$ | 30 | 198 | 13 | 0.24 | NA | Hu et al. (2008) |
| Hainan, China | 18°40′ N | PM$_{2.5}$ | 51 | 9.4 | NA | 0.75 | NA | Wang et al. (2008) |
| Alert, Canada | 82°30′ N | TSP | 0.30 | 1.6 | 0.12 | 0.02 | 9.4 | Fu et al. (2009) |
| Hyytiälä, Finland | 61°51′ N | PM$_1$ | 17 | 65 | NA | 0.25 | NA | Kourtchev et al. (2008a) |
| Jülich, Germany | 50°54′ N | PM$_{2.5}$ | 21 | 26 | NA | 0.14 | NA | Kourtchev et al. (2008b) |
| RTP, NC, USA | 35°54′ N | PM$_{2.5}$ | 137 | 153 | 11 | NA | NA | Lewandowski et al. (2007) |
| An urban site in Mexico | 19°29′ N | PM$_{2.5}$ | 34 | 48 | 3.4 | 0.17 | 0.91 | Stone et al. (2010) |
| A rural site in Mexico | 19°42′ N | PM$_{2.5}$ | 32 | 55 | 5.7 | 0.28 | 4.8 | Stone et al. (2010) |
| Balbina, Brazil | 1°55′ S | PM$_{2.5}$ | 57 | NA | NA | 1.8 | NA | Claeys et al. (2004) |

[a] NA: not available.

**Table 4.** Concentration (ng m$^{-3}$) and geometric mean diameters (GMD, µm) of BSOA and other components in fine (< 2.1 µm) and coarse modes (> 2.1 µm).

| | Fine mode (< 2.1 µm) | | Coarse mode (> 2.1 µm) | |
| | Concentration | GMD[a] | Concentration | GMD[a] |
|---|---|---|---|---|
| Particle mass (µg m$^{-3}$) | 26 ± 3.7 | 0.82 ± 0.01 | 20 ± 3.9 | 9.82 ± 1.27 |
| SO$_4^{2-}$ (µg m$^{-3}$) | 11 ± 1.4 | 0.89 ± 0.03 | 3.0 ± 0.18 | 10.0 ± 1.51 |
| NO$_3^-$ (µg m$^{-3}$) | 0.81 ± 0.45 | 0.58 ± 0.17 | 1.2 ± 0.32 | 9.41 ± 2.06 |
| NH$_4^+$ (µg m$^{-3}$) | 2.4 ± 0.51 | 0.90 ± 0.04 | 0.04 ± 0.01 | 5.97 ± 2.01 |
| | | | | |
| I. Isoprene derived SOA | | | | |
| MGA | 2.3 ± 0.54 | 0.87 ± 0.01 | 1.3 ± 0.27 | 8.77 ± 2.68 |
| MT | 3.4 ± 0.24 | 0.79 ± 0.06 | 1.3 ± 0.37 | 7.02 ± 1.88 |
| ME | 6.8 ± 1.09 | 0.82 ± 0.03 | 2.0 ± 0.56 | 6.38 ± 1.52 |
| | | | | |
| II. $\alpha$-/$\beta$-pinene derived SOA | | | | |
| PA | 1.6 ± 0.57 | 0.74 ± 0.03 | 2.4 ± 0.81 | 8.40 ± 2.81 |
| HGA | 1.6 ± 0.32 | 0.77 ± 0.02 | ND[b] | ND[b] |
| MBTCA | 1.5 ± 0.30 | 0.80 ± 0.04 | 0.05 ± 0.01 | 2.70 ± 0.00 |
| | | | | |
| III. $\beta$-caryophyllene derived SOA | | | | |
| CA | 1.2 ± 0.10 | 0.79 ± 0.01 | ND[b] | ND[b] |
| | | | | |
| III. Other organic compounds | | | | |
| C$_4^{di}$ | 4.9 ± 0.97 | 0.98 ± 0.02 | 12.0 ± 0.54 | 9.96 ± 2.76 |
| C$_5^{di}$ | 1.5 ± 0.11 | 0.88 ± 0.04 | 0.62 ± 0.17 | 9.22 ± 2.93 |
| MA | 5.5 ± 0.95 | 0.82 ± 0.04 | 0.47 ± 0.15 | 9.12 ± 3.70 |
| $o$-P | 4.6 ± 0.87 | 0.76 ± 0.03 | 4.3 ± 1.8 | 9.76 ± 3.11 |
| $m$-P | 0.11 ± 0.01 | 0.73 ± 0.04 | 0.04 ± 0.01 | 8.97 ± 3.19 |
| $p$-P | 1.8 ± 0.44 | 0.72 ± 0.06 | 0.38 ± 0.22 | 9.52 ± 5.94 |
| Levo | 11 ± 1.1 | 0.63 ± 0.05 | 1.3 ± 0.11 | 6.83 ± 2.28 |
| Arab | 0.32 ± 0.06 | 0.68 ± 0.08 | 7.1 ± 1.34 | 8.21 ± 2.78 |

[a] log GMD = ($\Sigma C_i \log Dp_i$)/$\Sigma C_i$, where $C_i$ is the concentration of compound in size $i$ and $Dp_i$ is the geometric mean particle diameter collected on stage $i$ (Hinds, 1999).
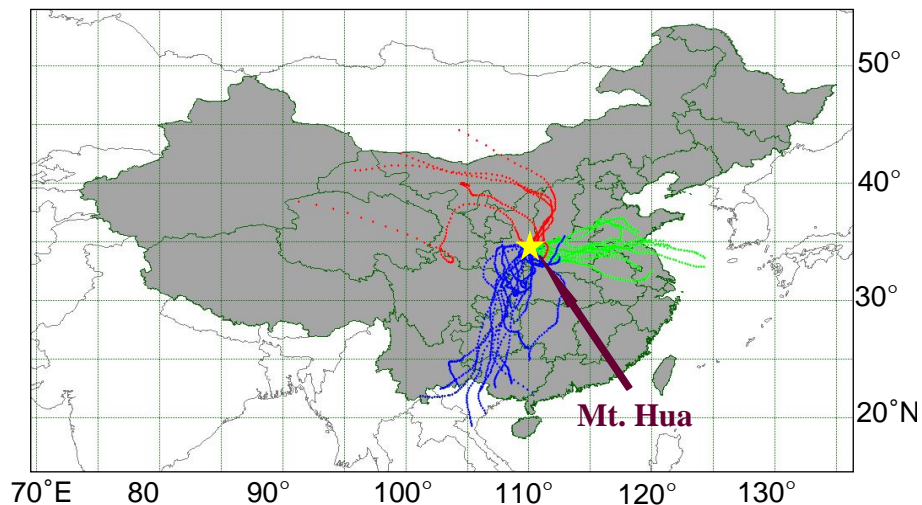[b] ND: not detected.

**Fig. 1.** Location of the sampling site (Mt. Hua; 34°29′ N, 110°05′ E; 2060 m a.s.l.) and 72 h backward air mass trajectories reaching the summit during the sampling in the summer of 2009.
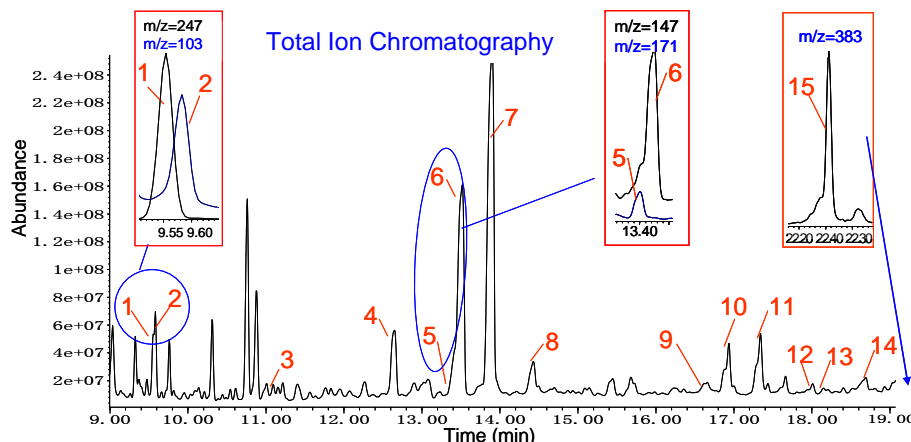
**Fig. 2.** GC/MS ion chromatography obtained for a trimethylsilylated PM$_{10}$ sample collected on 15 August 2009 (1. succinic acid; 2. 2-methylglyceric acid; 3. glutaric acid; 4. malic acid; 5. *cis*-pinonic acid; 6. 2-methylthreitol; 7. 2-methylerythritol; 8. 3-hydorxyglutaric acid; 9. *o*-phthalic acid; 10. levoglucosan; 11. arabitol; 12. 3-methyl-1,2,3-butanetricarboxylic acid; 13. *m*-phthalic acid; 14. *p*-phthalic acid; 15. *β*-caryophyllinic acid).
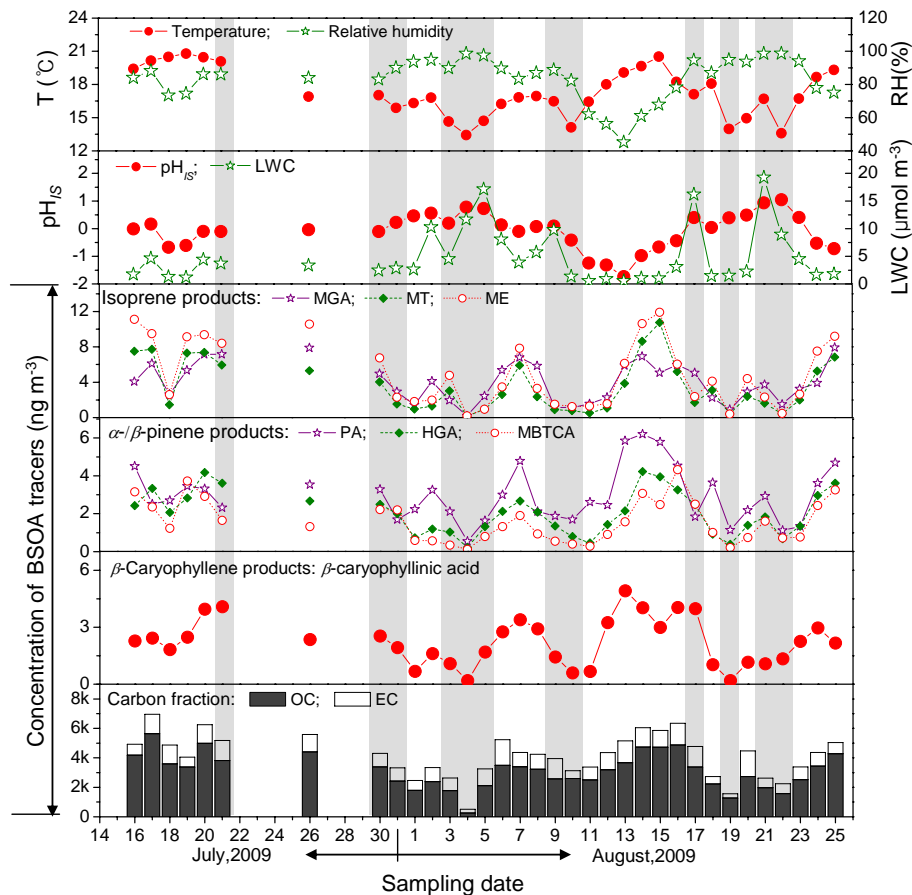
**Fig. 3.** Temporal variation of biogenic products as well as temperature, relative humidity, $pH_{IS}$ (in-situ acidity of particles), LWC (liquid water content of particles), OC and EC (organic and elemental carbon). The shadows denote rainy periods.
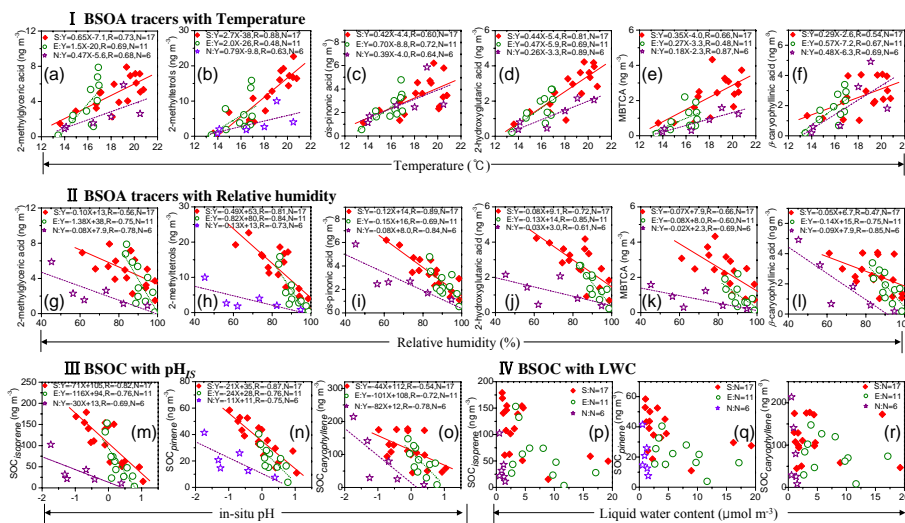
**Fig. 4.** Linear regression of **(a–f)** BSOA tracers with temperature, **(g–l)** BSOA tracers with relative humidity, **(m–o)** BSOC with in-situ pH (pH$_{IS}$), and **(p–r)** BSOC with liquid water content (LWC). S, E, N: the PM$_{10}$ samples in Mt. Hua from southerly (S), easterly (E) and northerly (N) air masses. 2-mehyltetrols: the sum of 2-methylthreitol and 2-methylerythritol; MBTCA: 3-methyl-1,2,3-butanetricarboxylic acid. SOC$_{isoprene}$, SOC$_{pinene}$ and SOC$_{caryophellene}$: the total mass concentrations of SOC (secondary organic carbon) produced by isoprene, $\alpha$-/$\beta$-pinene, and $\beta$-caryophyllene were calculated using a tracer-based method reported by Kleindienst et al. (2007).

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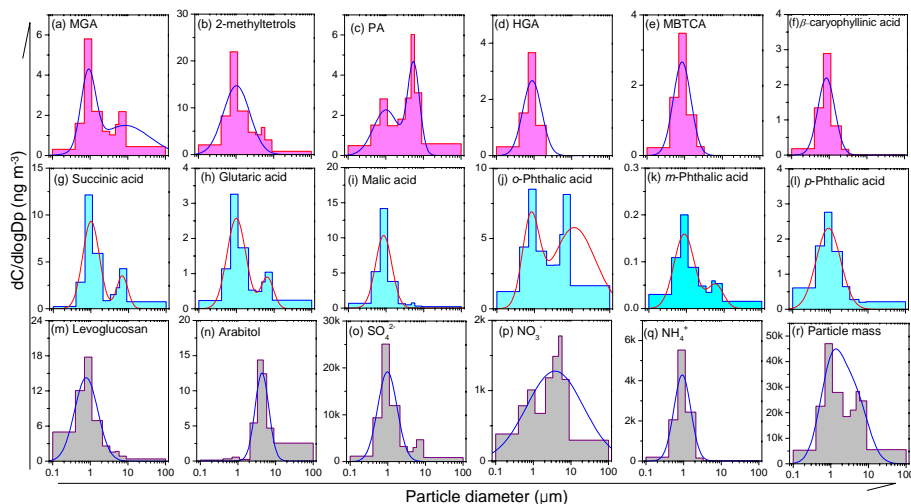
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**Fig. 5.** Size distribution of **(a–n)** organic aerosols, **(o–p)** inorganic ions, and **(r)** particle mass (MGA: 2-methylglyceric acid, 2-methyltetrols: the sum of 2-methylthreitol and 2-methylerythritol; PA: *cis*-pinonic acid, HGA: 3-hydroxyglutaric acid, MBTCA: 2-methyl-1,2,3-butanetricarboxylic acid).