

**Polar organic tracers
in PM_{2.5} aerosols
from Chinese forests**

W. Wang et al.

Polar organic tracers in PM_{2.5} aerosols from forests in eastern China

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Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Abstract

Photooxidation products of biogenic volatile organic compounds, mainly isoprene and monoterpenes, are significant sources of atmospheric particulate matter in forested regions. The objectives of this study were to examine time trends and diurnal variations of polar organic tracers for the photooxidation of isoprene and α -pinene to investigate whether they are linked with meteorological parameters or trace gases and to estimate their regional carbon contributions. PM_{2.5} (particulate matter with an aerodynamic diameter <2.5 μ m) aerosol samples were collected from forests in eastern China and compared with data from forested sites in Europe and America. Aerosol sampling was conducted at four sites located along a gradient of ecological succession in four different regions of China, i.e. Changbai Mountain Nature Reserve (boreal-temperate), Chongming National Forest Park (temperate), Dinghu Mountain Nature Reserve (sub-tropical) and Jianfengling Nature Reserve (tropical) during summer periods when the meteorological conditions are believed to be favorable for photochemical processes. Fifty PM_{2.5} samples were collected; seventeen organic compounds, organic carbon (OC), elemental carbon and trace gases were measured. Results indicate that the concentration trends of the secondary organic compounds reflected those of the trace gases and meteorological parameters. The 24-h average concentrations of isoprene oxidation products, α -pinene oxidation products, sugars and sugar alcohols vary systematically along gradients of ecological succession, except malic acid which may have both biogenic and anthropogenic sources. The maximum carbon contribution of isoprene and α -pinene oxidation products to the OC was 2.4% (293 ng/m³, Changbai day-time) and 0.3% (41.3 ng/m³, Changbai night-time), respectively.

1 Introduction

Vegetation releases numerous volatile organic compounds (VOCs) into the atmosphere, particularly isoprene, monoterpenes, and sesquiterpenes, as well as a series

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Polar organic tracers in PM_{2.5} aerosols from Chinese forests

W. Wang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Polar organic tracers
in PM_{2.5} aerosols
from Chinese forests**

W. Wang et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

of oxygen-containing compounds (Fehsenfeld et al., 1992; Sharkey and Yeh, 2001). Knowledge of the atmospheric fate of these biogenic VOCs (BVOCs) is important because of the large quantities emitted globally compared with those of anthropogenic VOCs as well as their high reactivity (Hoffmann et al., 1997; Griffin et al., 1999).

5 Secondary organic aerosol (SOA) is formed in the troposphere from the oxidation of BVOCs through reactions with ozone and OH or NO₃ radicals, where the resultant low-vapor-pressure oxidation products partition between the gas and aerosol phases or undergo further heterogeneous reactions in the aerosol phase (for a review, see Kanakidou et al., 2005). SOA produced over forests has received considerable attention for different reasons: it impairs visibility and may affect climate by scattering and absorbing solar radiation and by serving as cloud condensation nuclei.

China is comparable in size to Europe; hence, the isoprene and monoterpene emissions from China's ecosystems may be nearly the same as those for Europe (Klinger et al., 2002). So far, detailed organic speciation studies including oxidation products of isoprene and α -pinene have only been conducted for forested sites in Europe and America, e.g. Hungary (Ion et al., 2005), Germany (Plewka et al., 2006; Böge et al., 2006), Finland (Kourtchev et al., 2005, 2008a), the United States of America (USA) (Edney et al., 2005; Xia and Hopke, 2006; Cahill et al., 2006; Clements and Seinfeld, 2006; Lewandowski et al., 2007) and Brazil (Schkolnik et al., 2005). Information is available on the chemical composition of urban PM_{2.5} organic aerosol from Beijing (He et al., 2001), Shanghai (Ye et al., 2003), Pearl River Delta cities (Duan et al., 2007), inland and east coastal China (Wang et al., 2007), and Horqin sand land (Shen et al., 2007) in China. However, detailed organic composition studies on fine aerosol from forested sites in China are lacking.

25 In this study, day- and night-time and also 24-h PM_{2.5} aerosol samples were collected along latitude in boreal, temperate, subtropical, tropical ecosystems, during summer periods when the meteorological conditions are favorable for photochemical reactions of BVOCs. The concentrations of oxidation products of isoprene and α -pinene as well as of other marker compounds that provide information on aerosol

**Polar organic tracers
in PM_{2.5} aerosols
from Chinese forests**W. Wang et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

sources, i.e. fungal spores (Lewis and Smith, 1967; Bielecki, 1982) (arabitol, mannitol), plant pollen (Pacini, 2000) (glucose and fructose), and the oxidation of unsaturated fatty acids (Kawamura and Ikushima, 1993; Yu et al., 2005) (malic acid), were measured. As regards to oxidation products of isoprene, the following marker compounds were included: 2-methyltetrols (2-methylthreitol and 2-methylerythritol), C₅-alkene triols [2-methyl-1,3,4-trihydroxy-1-butene (*cis* and *trans*) and 3-methyl-2,3,4-trihydroxy-1-butene] and 2-methylglyceric acid. Both the 2-methyltetrols (Claeys et al., 2004a) and the C₅-alkene triols (Wang et al., 2005) were first characterized in fine tropical rainforest aerosol collected from the Amazon basin, Brazil, while 2-methylglyceric acid was first reported in rural fine aerosol collected from a deciduous/coniferous forest site in Kpuszta, Hungary (Claeys et al., 2004b). In subsequent laboratory studies (Edney et al., 2005; Böge et al., 2006; Surratt et al., 2006), it was confirmed that these compounds are formed from the photooxidation of isoprene. As regards to marker compounds for the photooxidation of α -pinene, pinic acid, norpinic acid, 3-hydroxyglutaric acid and 3-methyl-1,2,3-butanetricarboxylic acid were considered. These compounds have been reported in both field and smog chamber studies (Claeys et al., 2007); however, it is worth noting that the chemical structure of 3-methyl-1,2,3-butanetricarboxylic acid, which is believed to be a stable end-oxidation product formed through further oxidation of pinonic acid, was only recently elucidated (Szmigielski et al., 2007).

2 Experimental section

2.1 Site descriptions and sample collection

Study sites were located in four forests in eastern China along the north latitude from 42° to 18° (see Fig. 1). The boreal-temperate Changbai Mountain Forest Ecosystem Research Station (42°24′N, 128°28′E, 763 m a.s.l., Jilin Province) is on the edge of Erdaobaihe, a small tourist town with 60 000 inhabitants, surrounded by 200 km² of condensed pine/hardwood forest (the region of Changbai Mountain Nature Reserve) with

dominant species as pine (*Pinus koraiensis*), oak (*Quercus mongolica*), spruce (*Picea koreana*) and maple (*Acer mono*), etc. It is about 680 km to the west of Changchun (with a population of about 7 million), the capital city of Jilin province. This site is called Changbai, hereafter. The temperate site was inside a water fir (*Metasequoia glyptostroboides* Hu and Cheng) plantation in the Dongping National Forest Park (30° 50'N, 121° 40'E, ~ 0 m a.s.l., Chongming island, Shanghai). The area of this water fir plantation is about 3 km², surrounded by orchard and grassland. It is 12 km to the north of Chongming County (with about 700 000 inhabitants) and 28 km away from Shanghai Municipal center (about 20 million inhabitants) with Huangpu River in between. This site is called Chongming, hereafter. The subtropical Dinghu Mountain Forest Ecosystem Research Station (23° 10'N, 112° 32'E, 320 m a.s.l., Guangdong Province) is within the Dinghu Mountain Nature Reserve which covers an area of approximately 11 km² and is a monsoon evergreen broadleaf forest with some needle tree species that is dominated by hairy chestnut (*Castanopsis chinensis* Hance), Chinese cryptocarya (*Cryptocarya chinensis*), white gironniera (*Gironniera subaequalis* Planch) and schima (*Schima superba* Gardn. Et Champ). It is about 85 km to the west of Guangzhou (the capital city of Guangdong Province with a population of 7.5 million) and 18 km to the northeast of Zhaoqing (with about 3.7 million inhabitants). The sampling site was located at the hilltop in the Dinghu Mountain Biosphere Reserve, which is a station of the Man and the Biosphere Programme of the United Nations Educational, Scientific and Cultural Organization. This site is called Dinghu, hereafter. The fourth site was in the Jianfengling Long-term Research Station of Tropical Forest Ecosystem (18° 40'N, 108° 49'E, 820 m a.s.l., Hainan Province). It is a tropical evergreen monsoon forest with an area of 475 km², dominated by the species variegated (*Vatica mangachapoi*), white olive (*Canarium album*) and white tea (*Coelodepas hainanensis*). It is about 120 km from Sanya (the second largest city in Hainan Province, with a population of 500 000) and 315 km from Haikou, the capital city of Hainan Province. This site is called Hainan, hereafter. The central and western parts of Hainan island is mainly mountainous. The meaning of Jianfengling is sharp mountain peak, it is the highest

**Polar organic tracers
in PM_{2.5} aerosols
from Chinese forests**

W. Wang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Polar organic tracers
in PM_{2.5} aerosols
from Chinese forests**

W. Wang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

peak with an altitude of 1410 m among generally 400 m–700 m high mountains. The whole mountain area has an unsymmetrical circular distribution, which makes that the Jianfengling Long-term Research Station of Tropical Forest Ecosystem is situated in a rain forest basin. Therefore, the concentration of SOA from the photooxidation of isoprene and α -pinene is expected to be high at this site, unlike at coastal sites.

All sampling periods were selected taking into account the meteorological conditions and the maximum solar radiation. The sampling periods were 23–29 July 2007 in Changbai, 12–19 June 2006 in Chongming, 2–14 August 2006 in Dinghu, and 20–27 November 2006 during the dry season in Hainan.

For each site, two high-volume PM_{2.5} air samplers (Graseby-Andersen) operated at a flow rate of 1.13 m³ min⁻¹ were used, one collected day-/night-time samples (06:30 to 18:00 for day-time, 18:30 to 06:00 for night-time sampling), while the other collected 24-h samples, except at Dinghu where only 24-h samples were obtained. Whatman quartz fiber filters of 20.3×25.4 cm were used. Blank filters were obtained in the same way as the samples except that the collection time was only 15 s. No aerosol collection was performed on rainy days. In total, 50 field samples and 12 blanks were obtained.

2.2 Meteorological parameters and trace gases

Meteorological parameters (i.e. temperature, wind speed, relative humidity, and altitude) were measured by a Kestrel 4000 Pocket Weather meter (Nielsen-Kelleman, USA). In the case of the Changbai site, measurements of O₃, SO₂, NO, NO₂, NO_x and CO₂ were performed simultaneously with Thermo Environmental Instruments: an ultraviolet light absorption O₃ analyzer (TEI 49C), a fluorescence SO₂ analyzer (TEI 43C), a chemiluminescence NO-NO₂-NO_x analyzer (TEI 42C) and an infrared light absorption analyzer, respectively. The Shanghai Environmental Monitoring Center supplied concentrations of SO₂, NO₂ and O₃ for the Chongming site.

2.3 Aerosol analysis

Filter samples were analyzed for organic carbon (OC) and elemental carbon (EC) by a thermo-optical transmission (TOT) technique (Birch and Cary, 1996). For analysis of polar organic compounds by gas chromatography/mass spectrometry (GC/MS), the samples were spiked with appropriate amounts of internal recovery standards (IS), i.e. methyl- β -D-xylanopyranoside (MXP, Sigma) and deuterated D₃-malic acid (DMA, CDN isotopes, Canada), extracted with methanol, and derivatized with *N*-methyl-*N*-trimethylsilyltrifluoroacetamide (MSTFA)+1%trimethylchlorosilane (TMCS) (Pierce) and anhydrous pyridine (2:1, v/v). Details about the sample workup procedure can be found in Kourtchev et al. (2008a). The GC/MS instrument consisted of a Hewlett-Packard model 6890 gas chromatograph equipped with a DP-5MS (30 m×0.25 mm i.d., 0.25 μ m film thickness), coupled to a Hewlett-Packard model 5975 MSD quadrupole analyzer. Data were acquired and processed with ChemStation software (Hewlett-Packard). The temperature program was as follow: initial temperature at 100°C held for 2 min, a gradient of 3°C min⁻¹ up to 200°C, held for 5 min, then 30°C min⁻¹ up to 310°C, held for 2 min. Individual compounds were identified by comparison of mass spectra with those of authentic standards or literature data (Claeys et al., 2004a; Wang et al., 2004, 2005; Claeys et al., 2007). For quantitative analysis, calibration curves were constructed by using authentic standards that were processed as described above. For the quantification of compounds for which no standards were available the response factors of the following surrogate compounds were used: erythritol for 2-methyltetrols and C₅-alkene triols, pinic acid for norpinic acid, 2-hydroxyglutaric acid for 3-hydroxyglutaric acid, and 1,4-cyclohexanedicarboxylic acid for 3-methyl-1,2,3-butanetricarboxylic acid.

3 Results and discussion

A summary of the average concentrations and concentration ranges of OC, EC, organic compounds, and trace gases for 24-h, day-time and night-time PM_{2.5} aerosol samples,

Polar organic tracers in PM_{2.5} aerosols from Chinese forests

W. Wang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



and meteorological data at the four forest sites is listed in Table 1.

3.1 Isoprene oxidation products

Significant amounts of the 2-methyltetrols, marker compounds for the photooxidation of isoprene, were observed in boreal-temperate Changbai, subtropical Dinghu and tropical Hainan with average concentrations of 49 ng/m^3 , 25 ng/m^3 and 41 ng/m^3 , and concentration ranges between $22\text{--}109 \text{ ng/m}^3$, $3.5\text{--}84 \text{ ng/m}^3$, $16\text{--}67 \text{ ng/m}^3$ for 24-h samples, respectively. These values are comparable to the average concentration of the 24-h Hi-Vol samples from Potsdam, NY, during June and August of 2005 ($\sim 50 \text{ ng/m}^3$) (Xia and Hopke, 2006), and of the 48-h low volume samples from Hyytiälä, Finland during a 2004 summer period (26 ng/m^3) (Kourtchev et al., 2005).

The highest day-time average concentration of the 2-methyltetrols was 131 ng/m^3 and 60 ng/m^3 , for the Changbai and Hainan sites, respectively, while the corresponding night-time average concentration was 113 ng/m^3 and 50 ng/m^3 . A diel variation of the 2-methyltetrols with highest concentration during day-time was found for all four study sites (Table 1). This phenomenon can be regarded as evidence for the fast photochemical formation of 2-methyltetrols from locally emitted isoprene. While the 2-methyltetrols concentration of 60 ng/m^3 for the tropical rain forest site Hainan is similar to the day-time concentration of 65 ng/m^3 in Balbina, Brazil (Claeys et al., 2004a), the 2-methyltetrols concentration of 131 ng/m^3 in boreal-temperate Changbai is much higher than the day-time concentration of 15.7 ng/m^3 in boreal forest at Hyytiälä, Finland, during a 2005 summer period (Kourtchev et al., 2008a). This also holds for the night-time 2-methyltetrols concentration, 113 ng/m^3 , which is much higher than the concentration of 10.3 ng/m^3 in Hyytiälä at night. This may be due to discrepancies of latitude, vegetation types, leaf surface area and concentrations of trace gases in the atmosphere. The lower latitude of Changbai ($42^\circ 24' \text{N}$) compared to Hyytiälä ($61^\circ 51' \text{N}$) implies that there is more solar radiation and a higher temperature in Changbai (average 25.7°C in Changbai, 15.9°C in Hyytiälä, see Table 1). As to isoprene emissions from the vegetation, (Yan et al., 2005) estimated that the annual biogenic isoprene emission

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Polar organic tracers
in PM_{2.5} aerosols
from Chinese forests**

W. Wang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



is about 4.8 Tg C in whole China, and found that its spatial distribution corresponds with that of the vegetation. The highest annual emission flux exceeded 5000 kg C/km² in northeastern China, including Changbai Mountain, and Hainan island. As to trace gases, the concentration of SO₂, which is photooxidized to sulfuric acid and thought to be crucial in the formation of 2-methyltetrols, was 2.29 ppbv in Changbai compared to 0.04 ppbv in Hyytiälä. Therefore, the combined effects of higher isoprene emissions, higher photo-activity and higher concentrations of SO₂ may explain the fairly high maximum 2-methyltetrols concentration (282 ng/m³) in Changbai during day-time.

Furthermore, the C₅-alkene triols, compounds related to the 2-methyltetrols, were also detected at the four study sites. Recently, C₅-alkene triols and 2-methyltetrols were both reported to be generated from isoprene under low-NO_x conditions and are believed to be formed through reaction of isoprene with the OH radical. In addition, the formation of C₅-alkene triols was explained through rearrangement reactions of hydroxyhydroperoxy radicals that are formed in the initial photooxidation reaction of isoprene (Surratt et al., 2006). The sum concentration of the three C₅-alkene triol isomers was 10 ng/m³ and 19.3 ng/m³ in Changbai and Hainan, respectively. The ratio of the concentrations of C₅-alkene triols and 2-methyltetrols was 1:13 in Changbai versus 1:3 in Hainan. The highest ratio was thus found for tropical Hainan, which can be explained by favorable conditions at this site for the degradation of the above mentioned hydroxyhydroperoxy radicals that are formed in the initial photooxidation reaction of isoprene, such as low concentrations of SO₂ (which is oxidized to sulfuric acid) favoring the formation of C₅-alkene triols instead of 2-methyltetrols (Wang et al., 2005). 2-methylglyceric acid, another marker compound for the photooxidation of isoprene, was only found at very low concentrations in this study (Table 1).

The sum concentration of the 2-methyltetrols in 24-h samples from Changbai shows a good correlation with SO₂ ($r^2=0.9455$), see Fig. 2. In addition, 2-methyltetrols are well correlated with ozone ($r^2=0.982$) during day-time, and with NO₂ ($r^2=0.9618$) and NO_x ($r^2=0.8805$) at night. Clearly, these high correlations reveal a direct impact of SO₂ and the atmospheric oxidants, ozone and nitric oxide, on the formation of the

**Polar organic tracers
in PM_{2.5} aerosols
from Chinese forests**

W. Wang et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

2-methyltetrols. They may also imply complex synergies of solar radiation, meteorological conditions and trace gases. Similar results were obtained for the Chongming site. The good correlation between the concentrations of the 2-methyltetrols and SO₂ is consistent with sulfuric acid playing a crucial role in the formation of the 2-methyltetrols (Edney et al., 2005; Surratt et al., 2007). It cannot be ruled out that part of the 2-methyltetrols were originally present in the samples as sulfate esters which are also known to occur in ambient aerosol (Surratt et al., 2007a, 2007b; Gómez-González et al., 2008); these sulfate ester derivatives will be measured as 2-methyltetrols with the derivatization GC/MS method employed in the present study.

As regards to the good correlation found for the concentrations of the 2-methyltetrols and NO₂ at night, a recent smog chamber study by Ng et al. (2008) demonstrated that nitrate derivatives of the 2-methyltetrols can be formed through night-time NO₃ radical chemistry; these nitrate derivatives will not be stable upon derivatization GC/MS and will also be measured as 2-methyltetrols. Interestingly, the day-time 2-methyltetrol concentrations show a reverse correlation with CO₂ (Fig. 2; $r^2=0.9467$). A good correlation was found between the 2-methylthreitol and 2-methylerythritol concentrations for the four sites; the r^2 value for Changbai, Chongming, Dinghu and Hainan was 0.87, 0.94, 0.85 and 0.81, respectively, consistent with their formation through the same aerosol source process, i.e. photooxidation of isoprene.

3.2 α -pinene oxidation products

The recently identified markers for SOA from the photooxidation of α -pinene, i.e. 3-hydroxyglutaric acid and 3-methyl-1,2,3-butanetricarboxylic acid, were detected in samples from the boreal-temperate Changbai site. It is noted that at this site the concentration of the α -pinene oxidation products is much lower than that of the isoprene oxidation products. The average 24-h concentrations of 3-hydroxyglutaric acid and 3-methyl-1,2,3-butanetricarboxylic acid were 17.2 ng/m³ and 13.5 ng/m³, respectively, close to the values of 16.1 ng/m³ and 13.5 ng/m³ during July and August 2005 in Hyytiälä, Finland. In the tropical site of Hainan, 3-methyl-1,2,3-butanetricarboxylic acid

**Polar organic tracers
in PM_{2.5} aerosols
from Chinese forests**

W. Wang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

could only be detected in 24-h samples, norpinic acid was found at a concentration of 5 ng/m³, lower than 22.2 ng/m³ reported for the Finnish samples. The concentration of 3-hydroxyglutaric acid was 2.4 ng/m³, lower than Changbai (17.2 ng/m³) and Finland (16.1 ng/m³). These differences can for a part be explained by differences in the vegetation along latitude with isoprene-emitting species dominating in tropical regions and α -pinene-emitting species being prevalent in boreal regions. Pinic acid, a major photooxidation product of α -pinene in laboratory experiments, could only be detected at trace levels in all samples. In this respect, it is noted that low to rather high concentrations of pinic acid (range: 0.4–135 ng/m³) have been reported in field studies conducted at European and USA sites, including Pertouli in Greece (Kavouras et al., 1999a), Tábua in Portugal (Kavouras et al., 1999b), Hyytiälä in Finland (Kourtchev et al., 2005, 2008a), the Blodgett forest in California, USA (Cahill et al., 2006), the Fichtel mountains in Germany (Plewka et al., 2006), K-puszta in Hungary (Claeys et al., 2007), and Jülich in Germany (Kourtchev et al., 2008b¹). The relatively low sum concentration of isoprene oxidation products (4.7 ng/m³) is probably due to isoprene emissions from surrounding orchards and less from water fir at Chongming site. No α -pinene oxidation products could be detected in subtropical Dinghu.

3.3 Other identified compounds

In contrast to isoprene and α -pinene oxidation products, the concentrations of malic acid for Changbai, Chongming, Dinghu and Hainan were comparable, i.e. 43.5 ng/m³, 58.3 ng/m³, 24.9 ng/m³ and 24.1 ng/m³, respectively, similar to 46 ng/m³ in Hyytiälä, Finland (Kourtchev et al., 2005), 39 ng/m³ in K-puszta, Hungary (Ion et al., 2005), 22 ng/m³ in Balbina, Brazil (Claeys et al., 2004a), and 39.1 ng/m³ in Jülich, Germany (Kourtchev et al., 2008b¹). The sugar alcohols, arabitol and mannitol, showed sum con-

¹Kourtchev, I., Warnke, J., Maenhaut, W., Hoffmann, T., and Claeys, M.: Polar organic marker compounds in PM_{2.5} aerosol from a mixed forest site in western Germany, Chemosphere, in review, 2008b.



centrations of 36.9 ng/m³, 2.8 ng/m³, 7.8 ng/m³ and 30.8 ng/m³ for Changbai, Chongming, Dinghu and Hainan, respectively. The sum concentrations of fructose and glucose were 55.8 ng/m³, 3.6 ng/m³, 15.9 ng/m³ and 33.7 ng/m³, respectively. Both the latter sugar alcohols and sugars thus revealed substantial concentrations, as could be expected for ecosystems that are active during the considered periods.

3.4 Time trends and species attribution to OC

Figure 3 shows time trends for OC and the measured species, isoprene and α -pinene oxidation products, malic acid, mannitol and glucose, in Hainan. Isoprene oxidation products, mannitol (similar to arabitol) and glucose (similar to fructose) gave the same diel pattern with the highest concentration during day-time. Enhanced day-time isoprene oxidation products are consistent with the light- and temperature-dependent isoprene emission. Malic acid did not show day/night variations and followed closely the OC, indicating that its formation involves a longer time scale than that of isoprene oxidation. Similar trends for the 2-methyltetrols, malic acid and mannitol have been reported for K-puszt, Hungary during a 2003 summer period (Ion et al., 2005).

4 Conclusions

It can be concluded that all biogenic tracer compounds contribute significantly to the OC in both Hainan and Changbai (Fig. 4). Isoprene photooxidation is an important process for SOA formation with the average 24-h oxidation products of isoprene contributing 0.91% and 0.49% to the OC in Hainan and Changbai, respectively. The maximum carbon contribution of isoprene and α -pinene oxidation products to the OC was 2.4% (293 ng/m³, Changbai day-time) and 0.3% (41.3 ng/m³, Changbai night-time), respectively. Sugars and sugar alcohols also represent a significant fraction of OC at these two sites, indicating an active ecosystem. For the Chongming site, α -pinene oxidation products contribute 0.86% to the OC, which is the highest for the four sites.

Polar organic tracers in PM_{2.5} aerosols from Chinese forests

W. Wang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Polar organic tracers
in PM_{2.5} aerosols
from Chinese forests**

W. Wang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



The contributions of malic acid to OC for the four sites were comparable. The large part of the organic compounds that were not extractable in methanol or not detectable by GC/MS such as oligomeric humic-like substances and biopolymeric material likely constitute a major fraction of the unidentified organic matter. More research is warranted to better characterize the latter large missing portion of the organic matter using analytical methodology that is complementary to GC/MS such as, for example, liquid chromatography/mass spectrometry with electrospray ionization detection. Considering that SO₂ emissions are quite high in China, it would be very worthwhile to measure organosulfates of isoprene and α -/ β -pinene oxidation products that have only recently been reported (Surratt et al., 2007a; Iinuma et al., 2007; Gómez-González et al., 2008).

In general, the chemical constituents of SOA and their concentrations depend on vegetation area, plant species, meteorological conditions or trace gases. The correlation between SO₂ and isoprene oxidation products support that sulfuric acid plays an important role in the formation of isoprene-related SOA. On the other hand, night-time NO₃ radical chemistry also appears to be involved in SOA formation from isoprene.

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**Polar organic tracers
in PM_{2.5} aerosols
from Chinese forests**

W. Wang et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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**Polar organic tracers
in PM_{2.5} aerosols
from Chinese forests**

W. Wang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**Polar organic tracers
in PM_{2.5} aerosols
from Chinese forests**

W. Wang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Polar organic tracers
in PM_{2.5} aerosols
from Chinese forests**

W. Wang et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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**Polar organic tracers
in PM_{2.5} aerosols
from Chinese forests**

W. Wang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Table 1. Average concentrations and concentration ranges of OC, EC, organic compounds, and trace gases, for day-time, night-time and 24-h PM_{2.5} samples collected in Changbai and meteorological data.

Species	24-h		Day-time		Night-time	
	Average	Conc. Range	Average	Conc. Range	Average	Conc. Range
Changbai (23–29 July 2007)						
OC ($\mu\text{g}/\text{m}^3$)	n=5 4.8	3.6~5.7	n=5 5.6	3.3~6.7	n=4 6.1	5.1~6.9
EC	0.41	0.32~0.52	0.48	0.28~0.58	0.52	0.44~0.59
<i>Organics compounds</i> (ng/m ³)						
2-methylthreitol	16.7	4.7~37	35	14.2~80	26	6.9~45
2-methylerythritol	32	17.6~72	96	55~202	86	24~118
Σ 2-methyltetrols	49	22~109	130	69~282	113	31~144
C ₃ -alkene triols	4.4	0.19~11	10	0.23~28	1.2	n.d~3.3
2-methylglyceric acid	0.50	n.d~2.5	3.0	n.d~12	–	–
Σ isoprene oxidation products	53	23~120	140	69~290	114	31~166
3-hydroxyglutaric acid	17	7.7~27	16	*	25	*
3-methyl-1,2,3-butanetricarboxylic acid	14	8.5~18	22	*	16	*
Σ α -pinene oxidation products	31	16~46	38	*	41	*
malic acid	44	31~63	77	65~88	109	*
Arabitol	6.9	0.99~11	11	n.d~35	5.3	n.d~16
Mannitol	30	12~66	32	24~46	49	21~87
Fructose	5.2	3.9~7.2	15	6.5~25	96	4.6~278
Glucose	51	16~149	77	21~111	323	25~672
Threitol	1.3	n.d~4.1	0.57	n.d~1.5	2.9	n.d~8.8
Erythritol	1.9	0.054~4.4	1.2	0.71~2.01	4.9	n.d~14
2-hydroxyglutaric acid	15	13~18	48	*	18	*
SUM	239	99~343	384	252~532	730	108~1479
<i>Trace gases</i> (ppbv)						
SO ₂	2.04	1.7~7.1	2.3	1.7~4.1	1.8	17~2.6
NO	0.35	0.15~1.8	0.28	0.15~2.8	0.4	0.15~2.8
NO ₂	0.47	n.d~6.4	0.66	n.d~4.2	0.60	0.01~4.2
NO _x	0.67	0.05~6.4	0.92	0.05~7.0	0.93	0.24~7.0
O ₃	31	0.25~60	43	12~75	25	0.25~53
CO ₂	456	447~473	436	421~448	485	476~501
<i>Meteorological parameters</i>						
Temperature (°C)	25	22~27	26	24~27	23	21~24
Wind speed (m/s)	0.27	0.12~0.48	0.57	0.09~1.2	0.17	0.10~0.25
Relative humidity (%)	59	44~74	64	56~76	72	63~77

* only one sample was analyzed, no concentration range is available;

– not detected, the detection limit was ~50 pg/m³ for pinic acid, ~20 pg/m³ for norpinic acid.

Polar organic tracers in PM_{2.5} aerosols from Chinese forests

W. Wang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Table 2. Average concentrations and concentration ranges of OC, EC, organic compounds, and trace gases, for day-time, night-time and 24-h PM_{2.5} samples collected in Chongming and meteorological data.

Species	24-h		Day-time		Night-time	
	Average	Conc. Range	Average	Conc. Range	Average	Conc. Range
Chongming (12–19 June 2006)						
OC ($\mu\text{g}/\text{m}^3$)	n=4 9.9	5.8~15	n=4 10.5	6.4~17	n=1 5.0	*
EC	1.7	0.98~2.6	1.8	1.1~2.8	0.84	*
<i>Organic compounds (ng/m³)</i>						
2-methylthreitol	1.4	0.87~2.3	1.7	0.4~3.4	1.6	*
2-methylerythritol	3.0	2.2~4.3	4.3	1.4~8.1	4.2	*
Σ 2-methyltetrols	4.4	3.1~6.6	6.0	1.8~12	5.8	*
C ₅ alkene triols	0.09	n.d~0.23	0.10	n.d~0.22	0.7	*
2-methylglyceric acid	0.33	n.d~0.95	–	–	–	–
Σ isoprene oxidation products	4.8	3.1~7.8	6.1	1.8~12	6.6	*
3-hydroxyglutaric acid	–	–	–	–	–	–
norpinic acid	1.8	n.d~3.6	–	–	–	–
3-methyl-1,2,3-butanetricarboxylic acid	–	–	–	–	–	–
Σ α -pinene oxidated products	1.8	n.d~3.6	–	–	–	–
malic acid	58	37~88	79	16~173	32	*
Arabitol	0.72	0.28~1.4	2.9	0.18~8.4	0.69	*
Mannitol	1.9	0.43~4.6	5.8	1.5~12	1.3	*
Fructose	1.3	0.78~2.1	4.3	2.1~8.3	1.9	*
Glucose	2.4	0.73~4.2	10.3	2.0~25	2.1	*
Threitol	0.95	0.073~1.7	0.39	n.d~1.0	0.042	*
Erythritol	0.61	0.40~0.90	0.80	0.05~2.2	3.3	*
SUM	74	82~111	109	25~242	48	*
<i>Trace gases (ppbv)</i>						
SO ₂	10	7.0~17	16	8.0~27	23	*
NO ₂	22	13~22	25	18~34	36	*
O ₃	43	33~57	34	15~54	21	*
<i>Meteorological parameters</i>						
Temperature (°C)	29	29~30	27	26~28	27	*
Wind speed (m/s)	0.7	0.4~1.2	0.5	0.32~0.78	0.25	*
Relative humidity (%)	68	60~75	73	64~80	82	*

* only one sample was analyzed, no concentration range is available;

– not detected, the detection limit was $\sim 50 \text{ pg}/\text{m}^3$ for pinic acid, $\sim 20 \text{ pg}/\text{m}^3$ for norpinic acid.

Polar organic tracers in PM_{2.5} aerosols from Chinese forests

W. Wang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Table 3. Average concentrations and concentration ranges of OC, EC, organic compounds, and trace gases, for 24-h PM_{2.5} samples collected in Dingdu and meteorological data.

Species	24-h	
	Average	Conc. Range
Dinghu (2–14 August 2006)		
	n=7	
OC ($\mu\text{g}/\text{m}^3$)	5.1	1.8~8.0
EC	0.69	0.25~1.1
<i>Organics compounds (ng/m³)</i>		
2-methylthreitol	5.7	1.3~13
2-methylerythritol	20	2.2~70
Σ 2-methyltetrols	25	3.5~83
C ₅ alkene triols	0.62	0.14~1.51
2-methylglyceric acid	0.09	n.d~0.18
Σ isoprene oxidation products	26	4.3~84
3-hydroxyglutaric acid	–	–
norpinic acid	–	–
3-methyl-1,2,3-butanetricarboxylic acid	–	–
Σ α -pinene oxidation products	–	–
malic acid	29	4.1~62
Arabitol	2.8	0.92~7.7
Mannitol	5.0	2.0~12
Fructose	8.8	1.3~37
Glucose	6.2	2.0~19
Threitol	0.21	n.d~0.51
Erythritol	3.1	0.22~18
<i>SUM</i>	81	15~178
<i>Meteorological parameters</i>		
Temperature ($^{\circ}\text{C}$)	24.2	24~28
Wind speed (m/s)	0.57	0.26~0.90
Relative humidity (%)	73.5	60~82

* only one sample was analyzed, no concentration range is available;

– not detected, the detection limit was $\sim 50 \text{ pg}/\text{m}^3$ for pinic acid, $\sim 20 \text{ pg}/\text{m}^3$ for norpinic acid.

**Polar organic tracers
in PM_{2.5} aerosols
from Chinese forests**

W. Wang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Table 4. Average concentrations and concentration ranges of OC, EC, organic compounds, and trace gases, for day-time, night-time and 24-h PM_{2.5} samples collected in Hainan and meteorological data.

Species	24-h		Day-time		Night-time	
	Average	Conc. Range	Average	Conc. Range	Average	Conc. Range
Hainan (20–27 November 2006)						
	n=5		n=8		n=7	
OC ($\mu\text{g}/\text{m}^3$)	2.4	1.4~4.5	2.7	1.9~3.7	3.1	1.5~5.8
EC	0.20	0.12~0.38	0.23	0.17~0.32	0.20	0.12~0.38
<i>Organics compounds (ng/m³)</i>						
2-methylthreitol	12	4.7~20	17	8.7~28	12	2.8~20
2-methylerythritol	29	11~48	43	27~65	38	8.2~71
Σ 2-methyltetrols	41	15~67	60	36~86	50	11~92
C ₅ alkene triols	9.3	2.3~19	19	8.1~32	17	0.33~42
2-methylglyceric acid	0.74	0.21~1.2	0.66	0.11~1.2	0.74	0.26~2.4
Σ isoprene oxidation products	51	19~87	80	49~119	68	12~123
3-hydroxyglutaric acid	2.4	1.1~4.4	3.1	3.0~3.2	2.8	2.2~3.4
norpinic acid	5.0	0.38~8.7	17	16~17	0.29	*
3-methyl-1,2,3-butanetricarboxylic acid	2.1	0.43~4.7	–	–	–	–
Σ α -pinene oxidation products	9.4	6.5~15	20	19~20	2.9	2.2~3.4
malic acid	24	15~34	33	18~456	46	15~122
Arabitol	15	7.0~42	10	8.5~11	8.7	5.6~15
Mannitol	16	7.9~47	15	11~23	8.6	3.8~18
Fructose	12	4.6~43	10	6.9~12	4.7	1.4~14
Glucose	22	9.5~61	20	9.9~33	7.1	4.3~13
Threitol	0.89	0.30~2.9	0.55	0.39~0.80	1.0	0.11~2.3
Erythritol	3.6	2.0~5.7	9.9	5.8~18	2.4	0.62~4.0
2-hydroxyglutaric acid	4.6	2.0~9.6	6.8	5.8~7.8	3.1	1.2~6.3
SUM	154	94~132	189	160~219	149	57~120.
<i>Meteorological parameters</i>						
Temperature (°C)	22	20~25	24	21~27	23	20~26
Wind speed (m/s)	0.48	0.15~0.85	0.90	0.2~2.3	0.45	n.d~0.85
Relative humidity (%)	80	69~88	72	47~87	82	69~91

* only one sample was analyzed, no concentration range is available;

– not detected, the detection limit was $\sim 50 \text{ pg}/\text{m}^3$ for pinic acid, $\sim 20 \text{ pg}/\text{m}^3$ for norpinic acid.

Polar organic tracers in PM_{2.5} aerosols from Chinese forests

W. Wang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

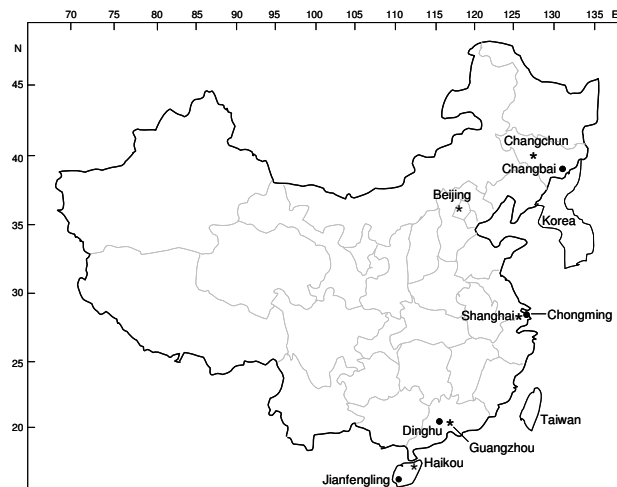
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Interactive Discussion



**Polar organic tracers
in PM_{2.5} aerosols
from Chinese forests**

W. Wang et al.

**Fig. 1.** Locations of sampling sites.[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

**Polar organic tracers
in PM_{2.5} aerosols
from Chinese forests**

W. Wang et al.

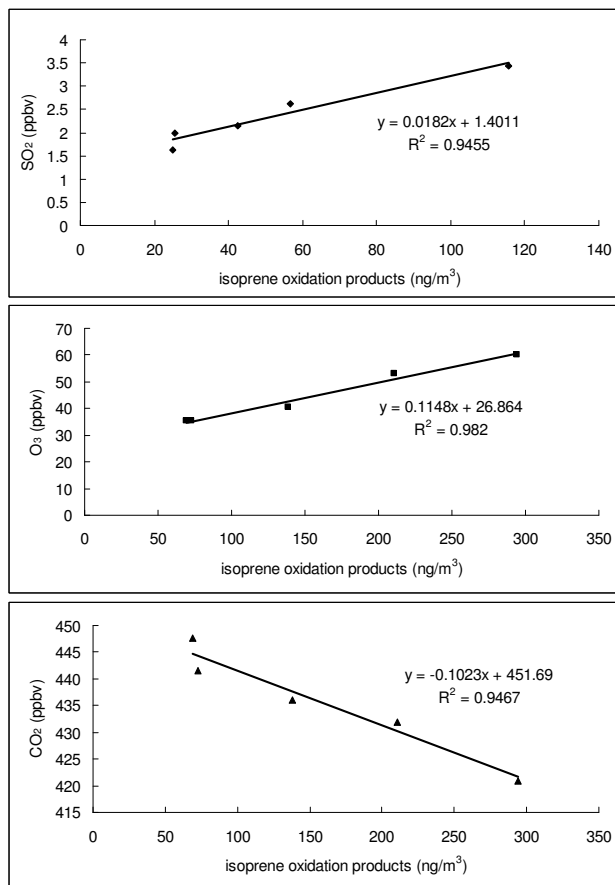


Fig. 2. Scatter plots of atmospheric concentration of SO₂ for 24-h (top), concentrations of O₃ (middle) and CO₂ (bottom) for the day-time versus the respective mass concentration of isoprene oxidation products (25–57 ng/m³ for 24-h, 69–294 ng/m³ for day-time sample) in Changbai.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

**Polar organic tracers
in PM_{2.5} aerosols
from Chinese forests**

W. Wang et al.

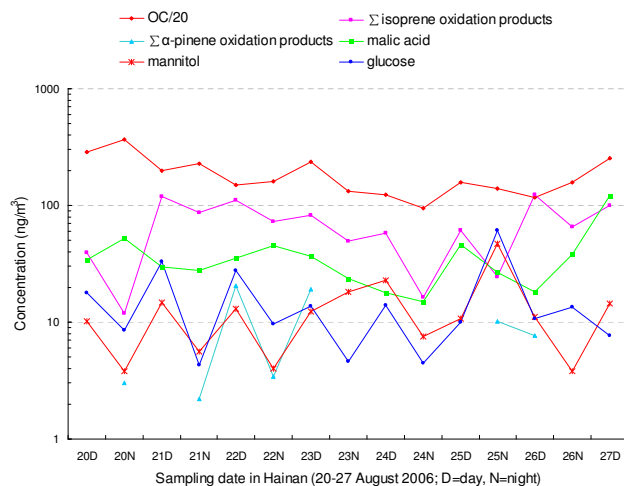


Fig. 3. Time trends for the concentrations of OC and selected compounds, i.e., isoprene oxidation products, α -pinene oxidation products, malic acid, mannitol and glucose for day/night samples collected from Hainan.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

**Polar organic tracers
in PM_{2.5} aerosols
from Chinese forests**

W. Wang et al.

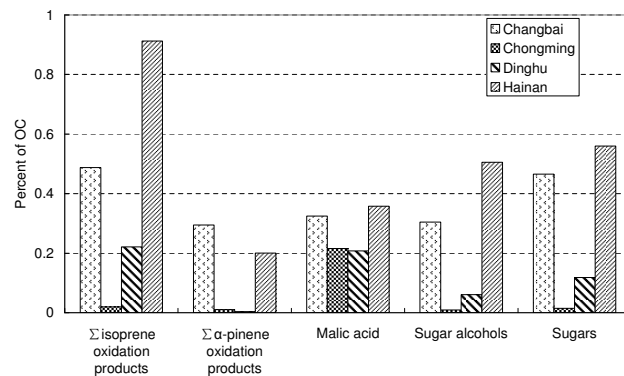


Fig. 4. Percentage of the OC attributable to the carbon in the organic compounds or groups of compounds for 24-h samples in Changbai, Chongming, Dinghu and Hainan.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)