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Seasonal variation of secondary organic aerosol in Nam Co, Central Tibetan Plateau

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Abstract

Secondary organic aerosol (SOA) affects the earth's radiation balance and global climate. High-elevation areas are sensitive to global climate change. However, at present, SOA origins and seasonal variations are understudied in remote high-elevation areas. In this study, particulate samples were collected from July 2012 to July 2013 at the remote Nam Co (NC) site, Central Tibetan Plateau and analyzed for SOA tracers from biogenic (isoprene, monoterpenes and β-caryophyllene) and anthropogenic (aromatics) precursors. Among these compounds, isoprene SOA (SOA_I) tracers represented the majority ($26.6 \pm 44.2 \, \mathrm{ng\,m^{-3}}$), followed by monoterpene SOA (SOA_M) tracers ($0.97 \pm 0.57 \, \mathrm{ng\,m^{-3}}$), aromatic SOA ($SOA_A$) tracer (2,3-dihydroxy-4-oxopentanoic acid, DHOPA, $0.25 \pm 0.18 \, \mathrm{ng\,m^{-3}}$) and β-caryophyllene SOA tracer (β-caryophyllenic acid, $0.09 \pm 0.10 \, \mathrm{ng\,m^{-3}}$). SOA_I tracers exhibited high concentrations in the summer and low levels in the winter. The similar temperature dependence of SOA_I tracers and isoprene emission suggested that the seasonal variation of SOA_I at the NC site was mainly influenced by isoprene emission. The ratio of high-NO_x to low-NO_x products of isoprene (2-methylglyceric acid to 2-methyltetrols) was the highest in the winter and the lowest in the summer, due to the influence of temperature and relative humidity. The seasonal variation of SOA_M tracers was impacted by monoterpenes emission and tracers partitioning. The similar temperature dependence of SOA_M tracers and monoterpenes emission was only observed during winter to spring. SOA_M tracer levels did not elevate with increased temperature in the summer, probably resulting from the counteraction of temperature effects on gas/particle partitioning and monoterpenes emission. The concentrations of DHOPA were 1–2 orders of magnitude lower than those reported in the urban regions of the world. Due to the transport of air pollutants from the adjacent Bangladesh and the eastern India, DHOPA presented relatively higher levels in the summer. In the winter when air masses mainly came from the northwestern India, mass fractions of DHOPA in total tracers increased, although its concentrations declined. The SOA-tracer method was applied to estimated secondary organic carbon (SOC) from

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these four precursors. The annual average of SOC was $0.22 \pm 0.29\,\mu g\,C\,m^{-3}$, with the biogenic SOC (sum of isoprene, monoterpenes and β-caryophyllene) accounting for 75 %. In the summer, isoprene was the major precursor with its SOC contributions of 81 %. In the winter when the emission of biogenic precursors largely dropped, the contributions of aromatic SOC increased. Our study implies that anthropogenic pollutants emitted in the Indian subcontinent could transport to the TP and have impact on SOC over the remote NC.

1 Introduction

Organic aerosol affects the earth's radiation balance and global climate. As a large fraction of organic aerosol, secondary organic aerosol (SOA) is produced by homogenous (Claeys et al., 2004) and heterogeneous (Jang et al., 2002) reactions of volatile organic compounds (VOCs) as well as aging of organic aerosol (Robinson et al., 2007; Donahue et al., 2012). The global emission of biogenic VOCs (BVOCs), such as isoprene and monoterpenes (Guenther et al., 1995) were estimated to be one order of magnitude higher than those of anthropogenic sources (Piccot et al., 1992). Thus, global SOA is believed to be largely from BVOCs.

SOA tracers from specific VOCs can provide insight on processes and sources influencing SOA formation and spatiotemporal distribution. The identification of the isoprene SOA (SOA_I) tracers, 2-methyltetrols (Claeys et al., 2004) revealed the importance of SOA_I in global SOA burden. The further studies in high-NO_x and low-NO_x products of isoprene intermediates (e.g. methacrylic acid epoxide and isoprene epoxydiols) provided more details in the mechanisms of SOA_I formation under the influence of NO_x (Paulot et al., 2009; Froyd et al., 2010; Surratt et al., 2010; Lin et al., 2013). The identification of tracers from aromatic SOA (SOA_A) (Offenberg et al., 2007) offered a way to directly evaluate the variation of anthropogenic SOA, particularly in urban regions. In addition, specific tracers have been determined in monoterpene SOA (SOA_M) (Jaoui et al., 2005; Claeys et al., 2007) and β-caryophyllene SOA (SOA_C)

(Jaoui et al., 2007; van Eijck et al., 2013). Based on these SOA tracers, Kleindienst and coworkers further developed an SOA-tracer method to attribute SOA sources in the ambient air. Since it is difficult to directly measure SOA, the SOA-tracer method provides a valuable technique to estimate SOA in the ambient air and it has been widely used around the world (Hu et al., 2008; von Schneidemesser et al., 2009; Guo et al., 2012; Lewandowski et al., 2013; Ding et al., 2014).

High-elevation areas are sensitive to global climate change (Xua et al., 2009). Observation of aerosol concentrations and compositions at high elevation sites can provide insight into the influence of natural and anthropogenic aerosols on global climate. The Tibetan Plateau (TP), the largest and highest plateau, is at the juncture of large desert areas and the densely populated Indian subcontinent. Previous study found the northwesterly winds could bring dust from the western deserts to the TP and lead to high levels of geological aerosols at a site on the southeast TP (Zhao et al., 2013). Moreover, anthropogenic pollutants (e.g. sulfate, nitrate, potassium, element carbon, and heavy metals) emitted in the developing countries in South Asia could be transported to the TP by the southerly and southwesterly winds, especially during the summer monsoon season (Cong et al., 2007; Ming et al., 2010; Li et al., 2013; Zhao et al., 2013).

The observation at the remote central TP site, Nam Co (NC) discovered that the mean ratio of organic carbon (OC) to element carbon (EC) was 31.9 ± 31.1 during July 2006 to January 2007, implying the significant SOA contribution to OC (Ming et al., 2010). However, there are only three studies in SOA compositions within the TP. Li et al. (2013) reported biogenic SOA (BSOA) tracers during the summer of 2010 at Qinghai Lake in the northeastern part of the TP. Stone et al. (2012) measured BSOA tracers from August to October 2005 on the south slope of Himalayas in the southwestern part of the TP. Due to the limited samples, it was difficult to examine the seasonal variation of these BSOA tracers in the TP. Moreover, due to the lack of anthropogenic SOA tracers, it was not possible to examine anthropogenic SOA in the TP, although above discussions have demonstrated that air pollutants from South Asia could transport to the TP. Our recent study provided a snapshot of SOA tracers over China (includ-

ing the NC and Linzhi sites in the TP) during the summer of 2012 (Ding et al., 2014). In this study, the observation at the remote NC site extended to one year. Seasonal trends of SOA tracers from isoprene, monoterpene, β-caryophyllene and aromatics were determined in the TP. Furthermore, secondary organic carbon (SOC) was estimated by the SOA-tracer method to check the variations of SOA origins at the NC site. To our knowledge, it is the first time that the seasonal trends of SOA tracers and its origins are studied in the remote TP.

2 Experiment

2.1 Field sampling

Samples were collected at a remote site (4730 m a.s.l.) at the southeastern shore of Nam Co Lake in the central TP (Fig. 1). Nam Co Lake ($90°16'$–$91°03'$ E and $30°30'$–$30°55'$ N) is located in the Nyainqen Tanglha Mountain Range with a total area of 2017 km^2 (Zhou et al., 2013). The major vegetation in the Nam Co Lake Basin is the high cold alpine meadow.

Sampling was undertaken from July 2012 to July 2013. An Anderson sampler equipped with 9-stage cascade impactors and pre-baked quartz fiber filters (Whatman, baked at 450 °C for 8 h) was used to get size-segregated particle samples at an air flow rate of 28.3 L min^{-1}. The 50 % cutoff sizes are < 0.4, 0.4–0.7, 0.7–1.1, 1.1–2.1, 2.1–3.3, 3.3–4.7, 4.7–5.8, 5.8–9.0, and ≥ 9.0 μm, respectively. The flow rate was calibrated before and after each sampling episode using an airflow meter to ensure the sampler operated at the specified flow rate. One set of 9 size-fractionated filters were collected for 72 h every two weeks. Additionally, four sets of field blanks were collected in the same way as the ambient samples for 5 min when the sampler was turned off. All samples were wrapped with aluminum foil and stored at -18 °C before analysis.

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2.2 Chemical analysis

Each set of nine filters were combined together as one sample to meet the analysis requirement. Detailed information on the SOA tracer analysis is described elsewhere (Ding et al., 2014). Prior to solvent extraction, isotope-labeled standard mixtures were spiked into samples as internal standards. Samples were extracted twice by sonication with the mixed solvent dichloride methane (DCM)/hexane (1 : 1, v/v), then three times with the mixed solvent DCM/methanol (1 : 1, v/v). The extracts of each sample were combined, filtered and concentrated to ~ 2 mL. Then, the concentrated solution was divided into two parts for methylation and silylation, respectively.

The samples were analyzed by an a gas chromatography/mass spectrometer detector (GC/MSD, Agilent 7890/5975C) in the selected ion monitoring (SIM) mode with a 30 m HP-5 MS capillary column (i.d. 0.25 mm, 0.25 µm film thickness). Splitless injection of a 2 µL sample was performed. The GC temperature was initiated at 65 °C, held for 2 min, then increased to 290 °C at 5 °C min^{-1} and held for 20 min. Thirteen SOA tracers were quantified, including five SOA$_M$ tracers (cis-pinonic acid, pinic acid, 3-methyl-1,2,3-butanetricarboxylic acid, 3-hydroxyglutaric acid and 3-hydroxy-4,4-dimethylglutaric acid), six SOA$_I$ tracers (2-methylthreitol, 2-methylerythritol, 2-methylglyceric acid, cis-2-methyl-1,3,4-trihydroxy-1-butene, trans-2-methyl-1,3,4-trihydroxy-1-butene and 3-methyl-2,3,4-trihydroxy-1-butene), one SOA$_C$ tracer (β-caryophyllenic acid) and one SOA$_A$ tracer (2,3-dihydroxy-4-oxopentanoic acid, DHOPA). cis-Pinonic acid and pinic acid were quantified by authentic standards. Due to the lack of standards, the SOA$_I$ tracers were quantified using erythritol (Claeys et al., 2004; Ding et al., 2008). The other SOA$_M$ tracers were quantified using cis-pinonic acid. β-Caryophyllenic acid and DHOPA were quantified using octadecanoic acid and azelaic acid, respectively (Ding et al., 2012). The method detection limits (MDLs) for cis-pinonic acid, pinic acid, erythritol, octadecanoic acid and azelaic acid were 0.03, 0.05, 0.04, 0.03 and 0.07 ng m^{-3}, respectively, at a total volume of 122 m^3.

It should be noted that ketopinic acid was used as the surrogate for the quantification of all SOA tracers by Kleindienst et al. (2007); while different surrogates were used to quantify different SOA tracers in this study. The response factors of internal standard calibration for the 5 surrogates ranged from 0.98 (azelaic acid) to 1.78 (pinic acid), with the average of 1.38 and the relative standard deviation (RSD) of 23 %. Thus, the quantification uncertainty caused by using surrogate calibration should be within 23 %. The response factor of ketopinic acid was also calculated in this study. Its value was 1.27, which was consistent with the average of the five surrogates.

2.3 Quality assurance and quality control

Field and laboratory blanks were analyzed in the same manner as the field samples. These SOA tracers were not detected in the field or laboratory blanks. To evaluate the recoveries of the analytical method, six spiked samples (authentic standards spiked into solvent with pre-baked quartz filters) were analyzed. The recoveries were 101 ± 3 % for cis-pinonic acid, 70 ± 10 % for pinic acid, 65 ± 14 % for erythritol, 83 ± 7 % for octadecanoic acid, and 89 ± 9 % for azelaic acid. The relative differences for target compounds in paired duplicate samples ($n = 6$) were all below 15 %.

2.4 Backward trajectories

The air masses' transport during each sampling episode was investigated using Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT V4.9). Five-day backward trajectories (BTs) were analyzed during each sampling episode with 6 h step at the height of 500 m above ground level. Then cluster analysis was performed to present the mean trajectory of each cluster, based on all the trajectories during our campaign.

3 Results and discussions

3.1 Seasonal variations of SOA tracers

Since the NC site is located in the high elevation TP, the annual temperature was only $-1.64\,°C$ with the range of $-16.1\,°C$ in January to $10.2\,°C$ in July (Table 1). The annual relative humidity (RH) was 58 % with the peak in July (84 %) and the lowest in January (30 %). The sum of all tracers ranged from 0.78 to $185\,\mathrm{ng\,m^{-3}}$. Among these compounds, SOA_I tracers ($26.6 \pm 44.2\,\mathrm{ng\,m^{-3}}$) represented the majority, followed by SOA_M tracers ($0.97 \pm 0.57\,\mathrm{ng\,m^{-3}}$), DHOPA ($0.25 \pm 0.18\,\mathrm{ng\,m^{-3}}$) and β-caryophyllenic acid ($0.09 \pm 0.10\,\mathrm{ng\,m^{-3}}$). During the summer (July–September 2012 and June–July 2013), SOA_I tracers presented the majority (> 95 %). The mass fractions of SOA_M tracers in all compounds increased during the cold period (October 2012 to May 2013).

3.1.1 Isoprene SOA tracers

The total concentrations of SOA_I tracers (sum of six tracers) ranged from 0.36–$184\,\mathrm{ng\,m^{-3}}$. The levels of SOA_I tracers were 1–2 orders of magnitude higher than those over the global oceans and the Arctic (Table 2). Among the SOA_I traces, 2-methyltetrols (sum of 2-methylthreitol and 2-methylerythritol, MTLs) were the major components (72 %), with an annual average of $23.8 \pm 40.3\,\mathrm{ng\,m^{-3}}$ (0.18 to $165\,\mathrm{ng\,m^{-3}}$). The 2-methylglyceric acid (MGA) averaged $1.95 \pm 2.92\,\mathrm{ng\,m^{-3}}$ and C_5-alkenetriols (sum of cis-2-methyl-1,3,4-trihydroxy-1-butene, trans-2-methyl-1,3,4-trihydroxy-1-butene and 3-methyl-2,3,4-trihydroxy-1-butene) averaged $0.93 \pm 1.39\,\mathrm{ng\,m^{-3}}$. MTLs are produced through the particle-phase uptake of the epoxydiols that formed in the gas-phase photo-oxidation of isoprene under low-NO_x or NO_x free conditions (Paulot et al., 2009; Surratt et al., 2010). Since the remote TP is a low-NO_x environment, it is expected that the low-NO_x products, MTLs dominated over other SOA_I tracers. The majority of MTLs at the NC site was consistent with those observed within the TP (Stone et al., 2012; Li et al., 2013) and over most global oceans (Fu et al., 2011; Hu et al., 2013), but

different from those over the North Pacific Ocean and the Arctic where MGA was the major SOA_I tracer due to the significant influence of Siberian fires (Fu et al., 2011; Ding et al., 2013). The two MTL isomers exhibited a strong correlation with each other throughout the year ($R^2 = 0.996$, $p < 0.001$) with a slope of 3.7, indicating that the two isomers shared similar formation pathways.

Figure 2a presents a typical seasonal trend of SOA_I tracers that high concentrations all existed in the summer. From October 2012 to April 2013, temperature was below zero, the levels of SOA_I tracers dramatically decreased as low as $0.38\,\mathrm{ng\,m^{-3}}$ in January. The natural logarithm of SOA_I tracer levels exhibited a negative correlation with the reciprocal of temperature in Kelvin ($p < 0.01$, Fig. 3a).

Isoprene emission rate (E_I) depends on light and temperature (Guenther et al., 1993):

$$E_I = EF_I \times C_L \times C_T \tag{1}$$

where EF_I is the basal emission rate at $30\,°C$ leaf temperature and $1000\,\mathrm{\mu mol\,m^{-2}\,s^{-1}}$ PAR. C_L and C_T are the factors representing the influences of light and temperature, respectively. C_T can be estimated as:

$$C_T = \frac{\exp^{\frac{C_{T1}(T-T_s)}{RT_sT}}}{1 + \exp^{\frac{C_{T2}(T-T_m)}{RT_sT}}} \tag{2}$$

Then the natural logarithm of C_T is calculated as:

$$\ln C_T = \frac{C_{T1}}{RT_s}\left(1 - \frac{T_s}{T}\right) - \ln\left[1 + \exp^{\frac{C_{T2}}{RT_s}\left(1 - \frac{T_m}{T}\right)}\right] \tag{3}$$

where $R = 8.314\,\mathrm{J\,K^{-1}\,mol^{-1}}$, $C_{T1} = 95\,000\,\mathrm{J\,mol^{-1}}$, $C_{T2} = 230\,000\,\mathrm{J\,mol^{-1}}$, $T_s = 303\,K$, $T_m = 314\,K$, and T is the leaf temperature (Guenther et al., 1993). Under the condition of $T < T_m$, the latter part in Eq. (3) is close to zero and $\ln C_T$ is linearly correlated

with $1/T$. As shown in Fig. 3a, there is a negative correlation between $\ln C_T$ and $1/T$ within the temperature range at the NC site (-16.7 to $10.2\,°C$). The similar temperature dependence of SOA$_I$ tracers and C_T indicated that the seasonal variation of SOA$_I$ at the NC site was mainly influenced by isoprene emission. In the summer, high temperature and intense light could enhance isoprene emission and photo-reactions and favor SOA$_I$ formation. In the winter, isoprene emission significantly dropped due to the extremely low temperature. Thus, the tracers were only in trace amount at the NC site.

It is worth noting that the ratio of MGA to MTLs (MGA/MTLs) was negatively correlated with temperature (Fig. 4a) and RH (Fig. 4b). Based on chamber results, the formation mechanisms of MGA and MTLs are quite different. MGA is produced under high-NO$_x$ conditions; while MTLs are mainly formed under low-NO$_x$ or NO$_x$ free conditions (Surratt et al., 2010). Moreover, low RH (15–40 %) could enhance the formation of MGA in the particulate phase but not of MTLs (Zhang et al., 2011). In addition, high particle acidity would favor the formation of MTLs instead of MGA (Surratt et al., 2007). Although there are few data available in the TP, the aerosols are expected to be neutral at the remote NC site. Thus, the influence of acidity on MGA/MTLs should be not significant. Isoprene emission is apparently high in summer due to high temperature and light intensity, which could enhance the ratio of isoprene to NO$_x$ and favor MTLs formation at the NC site. Moreover, high RH ($\sim 70\,\%$) in the summer (Table 1) could not favor MGA formation. Thus, MGA/MTLs exhibited the lowest values (less than 0.1) in the summer samples (Fig. 4). In the winter, both temperature and RH dropped to the lowest of the whole year. Low temperature reduced isoprene emission and low RH favored MGA formation. Thus, MGA/MTLs increased up to 0.8 in the winter samples (Fig. 4).

3.1.2 Terpene SOA tracers

The total concentrations of SOA$_M$ tracers (sum of five tracers) ranged from 0.11–2.39 ng m^{-3}. The levels of the SOA$_M$ tracers were consistent with those over the global

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oceans and the Arctic (Table 2). Among these traces, cis-pinonic acid was the major compound (54 %), with an annual average of 0.49 ± 0.38 ng m^{-3}, followed by pinic acid (0.22 ± 0.32 ng m^{-3}), 3-methyl-1,2,3-butanetricarboxylic acid (0.18 ± 0.25 ng m^{-3}), 3-hydroxyglutaric acid (0.08 ± 0.06 ng m^{-3}) and 3-hydroxy-4,4-dimethylglutaric acid (below MDL in the most samples).

The monthly variation of SOA$_M$ tracers did not fully follow that of temperature (Fig. 2b). From July to November 2012 (period 1), temperature decreased to $-15\,^{\circ}$C; while SOA$_M$ tracer levels increased as high as 1.99 ng m^{-3}. After that, both temperature and SOA$_M$ tracers dropped to the lowest values in January 2013 and increased concurrently till April 2013 (period 2). During May to July 2013 (period 3), SOA$_M$ tracer levels exhibited slight variation, although temperature kept increasing.

The seasonal variation of SOA$_M$ tracers could be influenced by monoterpenes emission and gas/particle partitioning. Monoterpenes emission rate (E_M) is often assumed to be solely dependent on temperature (Guenther et al., 1993):

$$E_M = EF_M \times \gamma_T \tag{4}$$

$$\gamma_T = \exp^{\beta(T - T_s)} \tag{5}$$

where EF$_M$ is monoterpenes emission rate at a standard temperature T_s (303 K), γ_T is the activity factor by temperature, β is an empirical coefficient usually taken to be 0.09 K^{-1} (Guenther et al., 1993), T is the leaf temperature. Apparently, the natural logarithm of γ_T is positively correlated with temperature. On the contrary, increasing temperature would favor the evaporation of SOA$_M$ tracers from particle phase to gas phase; and decreasing temperature would favor the condensation of these tracers from gas phase to particle phase (Saathoff et al., 2009). Thus, it is complicated that the influence of temperature on SOA$_M$ tracer levels in particle phase.

During the period 1, decreasing temperature could reduce monoterpenes emission and reactions. However, SOA$_M$ tracer levels were increasing, probably due to the dominant influence of partitioning over emission. During the period 2, both SOA$_M$ tracer levels and temperature were increasing. The natural logarithm of SOA$_M$ tracer levels

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were positively correlated with temperature (Fig. 3b), which was similar to that between $\ln\gamma_T$ and temperature within the temperature range at the NC site (Fig. 3b). The similar temperature dependence of SOA_M tracers and γ_T indicated that the significant increase of SOA_M from winter to spring at the NC site was mainly influenced by monoterpenes emission. The increase of SOA_M tracer concentrations during spring was also observed in the southeastern United States (Ding et al., 2008), resulting from the enhancement of monoterpenes emission in spring (Kim, 2011). During the period 3, high temperature could enhance monoterpenes emission and tracers formation; while it could favor the evaporation of these tracers from particle phase into gas phase. Thus, the relative stable of SOA_M tracer concentrations during the period 3 might reflect the counteraction of temperature effects on monoterpenes emission/tracers formation and gas/particle partitioning.

The levels of SOA_C tracer, β-caryophyllenic acid were in the range of below MDL to 0.40 ng m^{-3}. As Fig. 2c shows, the levels elevated from July to November 2012 and dropped to below MDL in December 2012. Then, the concentrations increased from January to March 2013 and decreased from April to June 2013. β-Caryophyllenic acid was significantly correlated with SOA_M tracers ($p = 0.025$), indicating the seasonal variation of β-caryophyllenic acid was similar with that of the SOA_M tracers.

3.1.3 Aromatic SOA tracer

The levels of SOA_A tracer, DHOPA were in the range of below MDL to 0.61 ng m^{-3}. This anthropogenic tracer was not detected or reported in global remote areas (Table 2). Due to few human activity at the remote NC site, the highest concentration of DHOPA was 1–2 orders of magnitude lower than those (up to 52 ng m^{-3}) reported in the urban regions of United States (Lewandowski et al., 2013) and China (Ding et al., 2014). DHOPA exhibited the higher concentrations in the summer and declined in the winter (Fig. 2d). Since there is no anthropogenic source near the remote NC site, the SOA_A tracer should be not locally formed but mainly transported from upwind regions.

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The TP features a monsoon climate (Cong et al., 2007; Ming et al., 2010; Zhao et al., 2013). Figure 5a presents the average trajectory of each cluster during our sampling in the whole year. The air masses over the NC were primarily from Bangladesh, Nepal and the eastern India (cluster 1, 32 %), the northwestern India (Indo-Gangetic basin) (cluster 3–6, 55 %), and the Taklimakan Desert (cluster 2, 13 %) during the sampling period. In the summer, the prevailing southerly winds (cluster 1, Fig. 5b) passed through the urban areas in the Bangladesh and the eastern India and could bring air pollutants into the TP. Previous studies in the TP have witnessed the enrichment of anthropogenic metals (Cong et al., 2007) and the enhancement of carbonaceous aerosols (Ming et al., 2010; Zhao et al., 2013) under the influence of summer monsoon. Thus, the increase of DHOPA levels at the NC site in the summer was mainly due to the transport of air pollutants from the upwind Bangladesh and the eastern India.

In the winter, the air masses over the NC site were mainly originated from the northwestern India by the westerly winds (Fig. 5b). As compared with the summer samples, the winter samples underwent the longer distance transport. Moreover, extreme low temperature in the winter could reduce DHOPA formation. Therefore, the levels of DHOPA were lower in the winter. It is worth noting that the mass fractions of DHOPA in all tracers significantly elevated in the winter (less than 2 % in the summer but up to 10 % in January, Fig. 2d), although its levels reduced. As described in Eqs. (1) and (4), temperature is an important factor controlling BVOCs emission. The drop of temperature from summer (up to 10.2 °C) to winter (low to −16.7 °C) at the NC site would lead to the emission of isoprene and monoterpenes decreasing by 98 and 90 %, respectively. The elevated fractions of DHOPA in the winter samples suggested that the SOA contributions from aromatics would increase in the winter when BVOCs emission largely decreased.

3.2 Source apportionment

The SOA-tracer method developed by Kleindienst and co-workers was applied to attribute SOC at the NC site. The researchers performed chamber experiments to obtain

the mass fraction of the tracers in SOC (f_{SOC}) for individual precursor:

$$f_{SOC} = \frac{\sum_i [\text{tri}]}{[\text{SOC}]} \qquad (6)$$

where $\sum_i [\text{tri}]$ is the total concentrations of the tracers for a certain precursor; [SOC] is the mass concentration of SOC. With these f_{SOC} values and the measured SOA tracers in the ambient air, SOC from different precursors can be estimated in the atmosphere, with the assumption that the f_{SOC} values in the chamber are the same as those in the ambient air. There is some degree of uncertainty in the SOA-tracer method due to the quantification with a single surrogate calibration standard (ketopinic acid) and the simplification of applying SOA tracers and conversion factors to calculate SOC in the ambient samples (Kleindienst et al., 2007). However, this method has been widely applied to attribute SOC from different precursors and proven to be able to provide reasonable results in the United States (Kleindienst et al., 2007, 2010; Stone et al., 2012; Lewandowski et al., 2013), and China (Hu et al., 2008; Guo et al., 2012; Peng et al., 2013; Ding et al., 2014).

The f_{SOC} were reported as 0.155 ± 0.039, 0.023 ± 0.0046 and $0.00797 \pm 0.0026\,\mu g\,(\mu g\,C)^{-1}$ for isoprene (SOC_I), β-caryophyllene (SOC_C) and aromatics (SOC_A), respectively (Kleindienst et al., 2007). In this study, the same set of SOA tracers as reported by Kleindienst et al. (2007) were used for SOC estimation, including MGA and MTLs for SOC_I, β-caryophyllenic acid for SOC_C and DHOPA for SOC_A. For monoterpene SOC (SOC_M), nine tracers were involved in the source profile (Kleindienst et al., 2007). However, only five of the nine SOA_M tracers were measured in the current study. Wang et al. (2013) compared the results from model prediction with field observation in the Pearl River Delta and pointed out that the SOA-tracer method would underestimate SOA_M, probably due to the mismatch of tracer compositions in the field and the source profile (Ding et al., 2014). To minimize the uncertainty caused by the mismatch in tracer compositions, the f_{SOC} with the same five SOA_M tracers ($0.059\,\mu g\,(\mu g\,C)^{-1}$) was computed using the chamber data from another study by the

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same research group (Offenberg et al., 2007). The same f_{SOC} for SOA_M was also applied to estimate SOC_M in our previous study over China (Ding et al., 2014).

The uncertainty in the SOA-tracer method is induced from the analysis of organic tracers and the determination of the conversion factors. The uncertainties in the tracer analyses were within 23 % in this study. The uncertainties of f_{SOC} were reported to be 25 % for isoprene, 48 % for monoterpenes, 22 % for β-caryophyllene and 33 % for aromatics (Kleindienst et al., 2007; Lewandowski et al., 2013). Considering these factors, the uncertainties of SOC were calculated through error propagation. The RSD were 34 % for SOC_I, 53 % for SOC_M, 32 % for SOC_C, and 40 % for SOC_A. On average, the RSD of the reconstructed SOC (sum of the four precursors) was 27 %.

Figure 6 presents the monthly variations of the reconstructed SOC. SOC was high in the summer 2012 and declined from October to December. After that, it kept increasing from January to June. The total concentrations of SOC ranged from 0.02 to 0.69 μg C m^{-3} with an annual average of 0.22 ± 0.29 μg C m^{-3}. The available data of OC in total suspended particles at the NC site were reported in the range of 1.18 to 2.26 μg C m^{-3} during July 2006 to January 2007 (Ming et al., 2010). Since we did not measured OC in our size-segregated samples, the OC data reported by Ming et al. (2010) were used to calculate SOC fraction in OC (SOC/OC) from July to January. The calculated SOC/OC was average 38 % in the summer and up to 58 % in September, suggesting SOC was an important contributor to OC at the NC site during the summer (Ming et al., 2010). However, from fall to winter, the elevated OC and decreased SOC led to SOC/OC declining from 11 % (in October) to 1 % (in January), indicating SOA from the four precursors had minor contributions to the elevated OC. Since the air masses during fall to winter were mostly originated from the northwestern Indo-Gangetic basin (cluster 3–6 in Fig. 5), primary pollutants emitted there could transport to the TP and have significant impact on the air at the NC site. In addition, SOA from aqueous-phase reactions and primary OA aging could not be captured by the SOA-tracer method. Thus, the current results might underestimate the total amount

of SOC, which partly explained the low OC shares of SOC at the NC site during fall to winter.

Biogenic SOC (sum of SOC_I, SOC_M, and SOC_C) dominated over anthropogenic SOC (SOC_A) at the NC site, averagely accounting for 75 % of the estimated SOC. In the summer, SOC_I was the major contributor with the SOC shares of 81 %. From fall to spring, SOC_M became the major contributor, averagely contributing 38 % to SOC. Although SOC_A level reduced in the winter, SOC_A contribution elevated as high as 53 % in January 2013. The elevated OC and the higher SOC_A contribution in the winter samples (Fig. 6) implied that the transport of anthropogenic pollutants from the Indian subcontinent might have significant influence on carbonaceous aerosols over the remote NC during winter.

4 Conclusion

Seasonal trends of SOA tracers and its origins were studied in the remote TP for the first time. SOA_I tracers represented the majority among these compounds. The significant temperature dependence of SOA_I tracers suggested that the seasonal variation of SOA_I at the NC site was mainly influenced by isoprene emission. Due to the influence of temperature and relative humidity, the ratio of high-NO_x to low-NO_x products of isoprene (MGA/MTLs) was the highest in the winter and the lowest in the summer. The seasonal variation of SOA_M tracers was impacted by monoterpenes emission and tracers partitioning. Due to the transport of air pollutants from the Indian subcontinent, DHOPA presented relatively higher concentrations in the summer and increased mass fractions in the winter. The SOA-tracer method was applied to estimated SOC from these four precursors. The annual average of SOC was $0.22 \pm 0.29\,\mu g\,C\,m^{-3}$, with the biogenic SOC accounting for 75 %. In the summer, isoprene was the major precursor with its SOC shares of 81 %. In the winter when the emission of biogenic precursors largely declined, the contributions of SOC_A increased. At present, SOA origins and seasonal variations are unclear in the remote high-elevation TP. The remote TP is

connected to the densely populated Indian subcontinent. Our study implies that anthropogenic pollutants emitted there could transport to the TP and influence SOC over the remote NC.

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Table 1. SOA tracers at the NC site ($\mathrm{ng\,m^{-3}}$).

Month	Temp.	RH	SOA tracers				
	°C	%	Isoprene	Monoterpenes	β-Caryophyllene	Aromatics	Sum
2012 07	7.78	84	54.1 ± 22.9	0.45 ± 0.48	0.10 ± 0.13	0.37 ± 0.23	55.0 ± 22.5
2012 08	7.70	76	66.0 ± 69.3	0.46 ± 0.18	nd [b]	0.49 ± 0.03	67.0 ± 69.1
2012 09	5.92	66	100 ± 118	1.06 ± 0.43	0.08 ± 0.11	0.35 ± 0.36	102 ± 118
2012 10	−1.50	70	14.7 ± 19.0	1.79 ± 0.08	0.16 ± 0.01	0.22 ± 0.07	16.8 ± 18.9
2012 11	−14.9	63	2.04 ± 1.76	1.99 ± 0.56	0.20 ± 0.19	0.25 ± 0.15	4.48 ± 2.66
2012 12	−13.0	45	0.52	0.73	nd	nd	1.25
2013 01	−16.1	30	0.38 ± 0.02	0.30 ± 0.04	0.03 ± 0.01	0.08 ± 0.01	0.78 ± 0.01
2013 02	−9.69	49	0.86 ± 0.45	0.52 ± 0.25	0.09 ± 0.02	0.09 ± 0.01	1.55 ± 0.22
2013 03	−7.83	41	1.56 ± 1.15	0.74 ± 0.59	0.23 ± 0.25	0.12 ± 0.17	2.65 ± 2.15
2013 04	−3.42	52	2.82 ± 0.20	1.24 ± 0.15	0.15 ± 0.03	0.20 ± 0.03	4.40 ± 0.11
2013 05	3.77	54	10.1 ± 9.70	1.11 ± 0.13	0.06 ± 0.06	0.27 ± 0.19	11.5 ± 9.97
2013 06	7.25	55	54.1 ± 42.9	0.83 ± 0.18	0.03 ± 0.04	0.30 ± 0.02	55.3 ± 42.8
2013 07	10.2	69	41.9	1.41	0.07	0.49	43.9
Annual	−1.64	58	26.6 ± 44.2	0.97 ± 0.57	0.09 ± 0.10	0.25 ± 0.18	28.0 ± 44.2

[a] one standard deviation; [b] "nd" means not detected.

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Table 2. SOA tracers in remote places on the global range ($ng\,m^{-3}$).

Locations		Seasons	References	SOA tracers			
				Isoprene[a]	Monoterpenes[a]	β-Caryophyllene	Aromatics
Tibetan Plateau	Nam Co Lake	Whole year	This study	26.6(0.36–184)[b]	0.97(0.11–2.39)	0.09(nd–0.40)	0.25(nd–0.61)
	Qianghai Lake	Summer	Li et al. (2013)	2.50(0.13–7.15)	2.95(0.30–10.4)	0.87(0.05–2.41)	na[c]
	Himalayas	Summer-autumn	Stone et al. (2012)	30.7(5.5–105)	13.2(5.6–31.3)	1.6(1.1–2.3)	na
Arctic	Alert	Winter–Summer	Fu et al. (2009)	0.3(0.08–0.567)	1.6(0.138–5.3)	0.12(0.01–0.372)	na
Global oceans	Arctic Ocean	Summer	Fu et al. (2013)	4.0(0.16–31.8)	4.8(0.44–24.1)	0.017(0.005–0.048)	na
	Low- to mid-latitude	Fall–Spring	Fu et al. (2011)	3.6(0.11–22)	2.7(0.02–15)	0.32(0–2.5)	na
	Antarctic to Arctic	Summer	Hu et al. (2013)	8.5(0.018–36)	3.0(0.05–20)	na	na
	North Pacific and Arctic	Summer	Ding et al. (2013)	0.62(0.12–1.45)	0.06(0.01-.25)	0.002(nd–0.03)	nd[d]

[a] compositions are different in different studies.
[b] data range in brackets.
[c] "na" means not available.
[d] "nd" means not detected.

Figure 1. Nam Co site in the Tibetan Plateau, China.
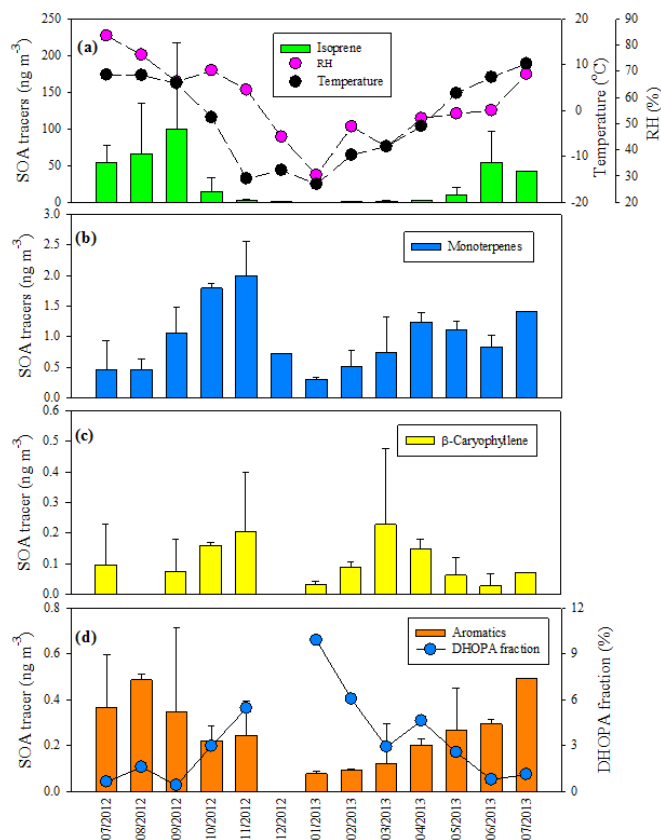
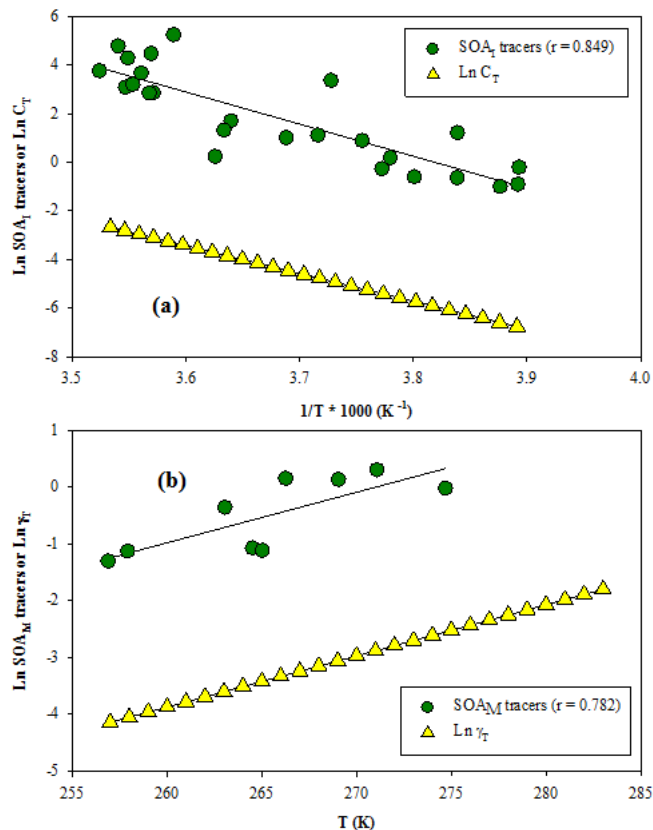**Figure 2.** Monthly variations of SOA tracers.

Figure 3. Temperature dependences of SOA$_I$ tracers in the whole year **(a)** and SOA$_M$ tracers during the period 2 **(b)**. All correlations are significant ($p < 0.001$).
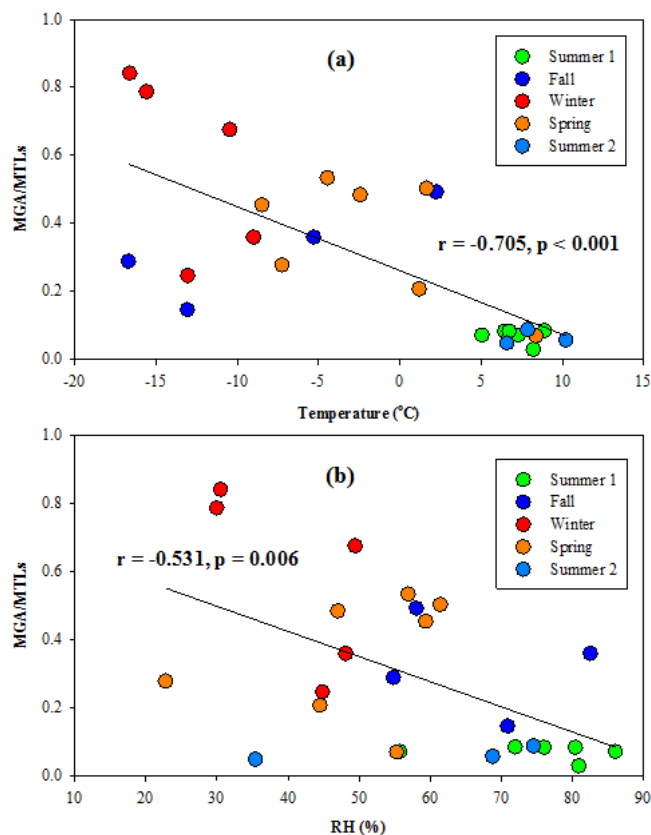
Figure 4. Correlations of MGA/MTL with temperature **(a)** and relative humidity **(b)**. Summer 1 is from July to September 2012, fall is from October to November 2012, winter is from December 2012 to February 2013, spring is from March to May 2013, summer 2 is from June to July 2013.
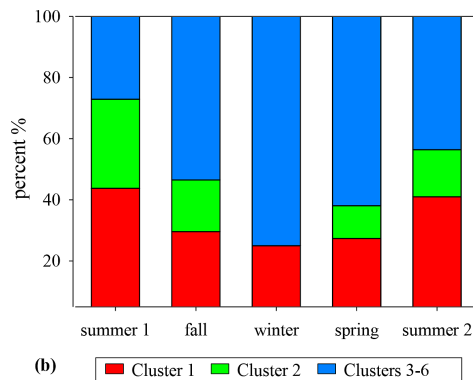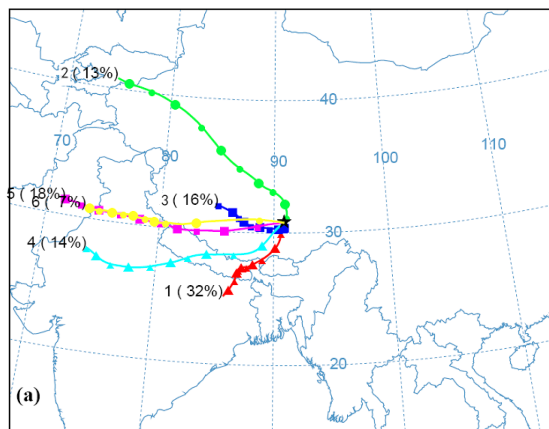
Figure 5. Cluster analyses of air masses at the NC site **(a)** and seasonal variations of clusters **(b)**, based on 5 day backward trajectories during the sampling period.

Figure 6. Seasonal variations of estimated SOC. OC data at the NC site during July 2006 to January 2007 were reported by Ming et al. (2010) and the err bar means one SD in each month.