Anonymous Referee #2

The manuscript presents new measurements of SOA tracers in the Central Tibetan Plateau and discusses seasonal variations in the absolute and relative contributions of biogenic and SOA tracers, particularly in the context of air mass origins. The manuscript requires major revisions prior to reconsideration for publication, namely in the broader discussion of potential local influences on SOA formation, the stability of the utilized tracers during transport, and the specificity and representativeness of a single toluene-SOA tracer to represent all of anthropogenic SOA. Analytically, differences between the methods utilized here and prior studies (e.g. Kleindienst et al. 2007) introduce complications and biases to the use of the SOA-tracer based source apportionment method. These biases, as well as others in quantification stemming from the use of surrogate quantification standards and an extraction protocol giving 65% recovery at times, need to be discussed and quantitatively assessed in order to develop realistic estimates of the errors in absolute quantification of SOA tracers.

Specific comments are provided here:
1. The title should be revised – "tracers" should be added after "secondary organic aerosol" in order to clarify that a select sub-set of SOA tracers were measured, and SOA in its entirety is not discussed. Also suggest removing "Nam Co" from the title to make it more concise.
Reply: The title has been revised as "Seasonal variation of secondary organic aerosol tracers in Central Tibetan Plateau"

2. The authors attribute SOA to long-range transport, but to not address the potential for SOA to form from local VOC precursors or combustion activities. The potential for release of biogenic VOC from nearby vegetation and NOx from local combustion sources (e.g. dung or biomass burning) must be addressed (Duo et al. 2015; Xiao et al. 2015).
Reply: Our study found that the seasonal variation of isoprene SOA (SOA_I) tracers was mainly influenced by the isoprene emission (C_T) which was estimated using local temperature. Considering the short lifetime of isoprene in the air (several hours), SOA_I should be mainly formed from local precursor. For monoterpene SOA (SOA_M), the ratio of *cis*-pinonic acid plus pinic acid to 3-methyl-1,2,3-butanetricarboxylic acid (P/M) indicated that SOA_M was generally fresh at the NC site (see the details in our reply to comment #12). Thus, SOA_M should be also mainly formed from local precursors. The local dung or biomass burning may be potential sources of aromatics, NOx, and even biogenic VOCs. However, the biomass burning tracer, levoglucosan not only exhibited different seasonal trend away from SOA tracers (Figure 1), but also presented poor correlations with all SOA tracers (p>0.05). These suggested that local dung or biomass burning was not the major sources of SOA during our sampling at the NC site.

For aromatic SOA (SOA_A), its tracer, DHOPA exhibited higher levels when air masses mainly came from the upwind Indian subcontinent (the Bangladesh and the northeastern India) where high population density and high levels of anthropogenic pollutants (AOT, CO, N_2O) were observed (see our response to the second comment by Reviewer #1). Considering there was few local anthropogenic source near the remote NC site, SOA_A should be not locally formed but mainly long-range transported from the upwind Indian subcontinent.

In the revised manuscript, we addressed that SOA_I and SOA_M were mainly formed from local precursors, and SOA_A was from long-range transport from the upwind Indian subcontinent. The impact of local biomass burning was also discussed (see below).

"Figure 3a presents a negative correlation between the natural logarithm of SOA_I tracer levels and the reciprocal of temperature in Kelvin (p<0.001). Moreover, the temperature dependence of SOA_I tracers was similar to that of C_T, and SOA_I tracers exhibited a significant positive correlation with C_T during our sampling at the NC site (Figure 3b). These results indicated that the seasonal variation of SOA_I at the NC site was mainly influenced by the isoprene

emission. Considering the short lifetime (several hours) of isoprene in the air, SOA_I should be mainly formed from local precursor."

"In this study, the ratio of P/M averaged 16.7 ± 20.9. Thus, SOA_M was generally fresh at the NC site and should be mainly formed from local precursors."

"Besides urban emissions from solvent and fossil fuel use, biomass burning is an important source of aromatics in many parts of the world (Lewis et al. 2013). The local dung or biomass burning (Duo et al. 2015; Xiao et al. 2015) may be potential sources of aromatics in the TP. Hence, DHOPA may come from the processing of biomass burning emissions. Figure 7 exhibits the monthly variation of biomass burning tracer, levoglucosan during our sampling. The concentrations of levoglucosan ranged from 0.82 ng m^{-3} (October 2012) to 4.55 ng m^{-3} (April 2013) with a mean of 1.87 ± 1.14 ng m^{-3}. Apparently, the monthly variation trend of levoglucosan was quite different from that of DHOPA. And there was no correlation between DHOPA and levoglucosan ($p>0.05$) (Figure S6). These indicated that DHOPA was not mainly from the processing of biomass burning emission at the NC site. Since there was few anthropogenic source near the remote NC site, the SOA_A tracer should be not locally formed but mainly transported from upwind regions."
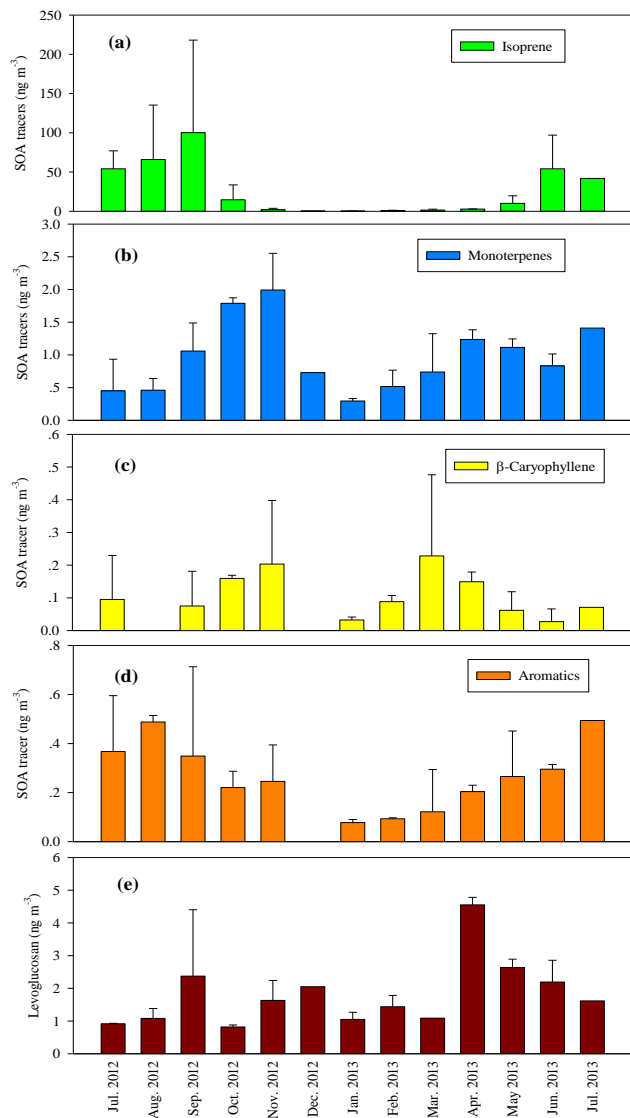

Figure 1 Monthly variations of SOA tracers (a-d) and levoglucosan (e)

3. Related, to what extent are the measured SOA tracers stable over the distance and time suggested for long-range transport? Prior studies have demonstrated extensive processing of organic aerosols in the region (Meng et al. 2013) as well as the loss of molecular tracers for organic aerosol during long-range transport (Stone et al. 2007).

Reply: The stability of SOA tracers during long-range transport is an interesting topic. Due to dilution, deposition and aging, the concentrations of SOA tracers should significantly decline during long-range transport. As we mentioned above, the biogenic SOA was mainly formed from local emissions. Thus, the concentrations of SOA_I tracers (up to 184 ng m^{-3}) at the NC site were comparable with those over continents (several to hundreds ng m^{-3}). On the contrary, since SOA_A was mainly transported from the upwind Indian subcontinent, even the highest concentration of DHOPA at the NC site was 1-2 orders of magnitude lower than those reported in the urban regions of the world. The global lifetimes of aerosols in the air are about 5 days (Bourgeois and Bey, 2011). Due to low temperature, the lifetimes of aerosols should be longer in cold regions, such as the Arctic and the Tibetan Plateau. At present, there is no result available in the lifetimes of these SOA tracers in the ambient air. We think the lifetimes of SOA tracers over the Tibetan Plateau should be comparable with those of aerosols. Given a lifetime of 5 days, the loss rate constant of tracers could be roughly estimated as 2.3×10^{-12} cm^3 molec^{-1} s^{-1} at OH levels of 1×10^6 molecules cm^{-3}. This is also the reason why we run back trajectory analysis only for 5 days.

Bourgeois, Q., and Bey, I.: Pollution transport efficiency toward the Arctic: Sensitivity to aerosol scavenging and source regions, J. Geophys. Res.-Atmos., 116, D08213, Doi: 10.1029/2010JD015096, 2011.

4. Section 2.2: Specify what type of ionization was used by the mass spectrometer
Reply: Electron impact (EI) ionization was used by the mass spectrometer. We have specified it in the revised manuscript "Thirteen SOA tracers were quantified by the GC-MS coupled with an electron impact (EI) ionization source"

5. Kleindienst et al. (2007) utilized chemical ionization, a soft ionization technique, to identify SOA tracers using molecular ions and high-m/z ratios, and analyzed SOA chamber samples in parallel to ambient samples to ensure consistency in mass fragmentation and gas chromatography (GC) retention times. Evidence in the form of retention data, observed MS fragments, and relative ratios of MS fragments are needed as evidence for the correct identification of the SOA tracer compounds. This information should be added as supplemental information.
Reply: Figure 2 presents the TIC of these SOA tracers with the retention time of each compound labeled. Figure 3-5 show the EI spectrum of each tracer. Figure 2-5 were added in the supplemental information file as Figure S1-S4 in the revised manuscript.
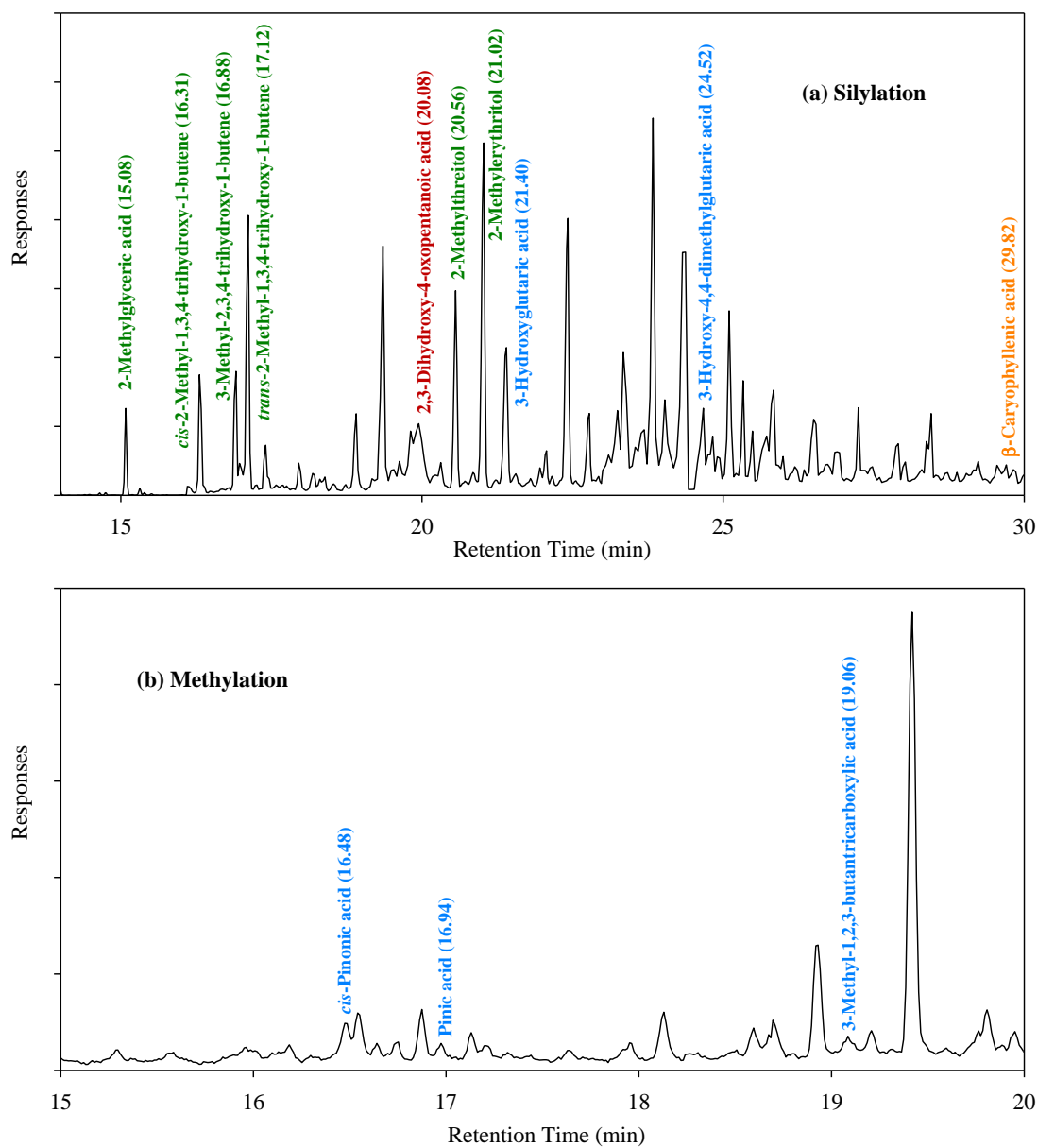
Figure 2 TIC of silylated (a) and methylated samples (b). Retention time of each tracer is labeled in brackets. Green, blue, orange and red represent SOA tracers from isoprene, monoterpenes, β-caryophyllene and aromatics, respectively.
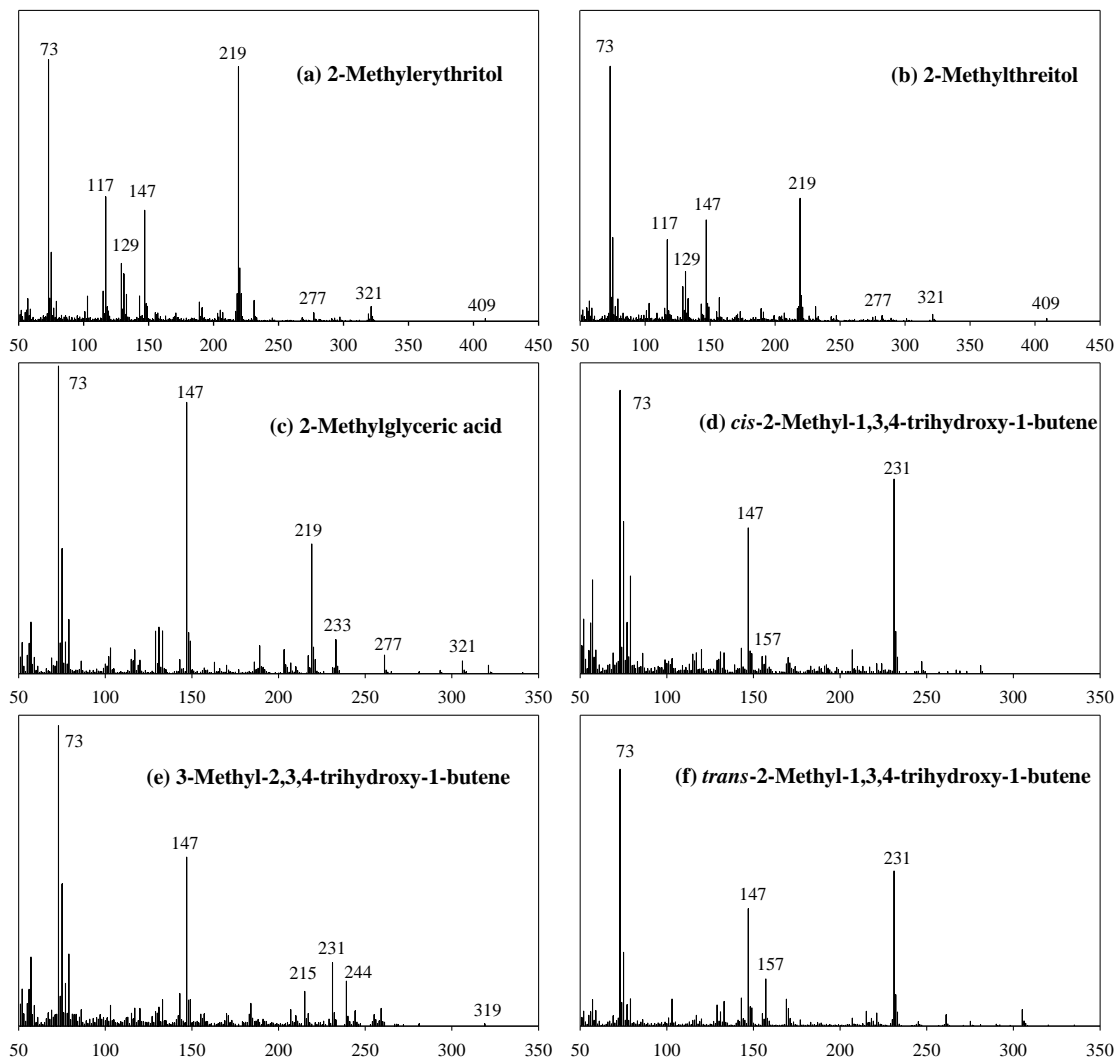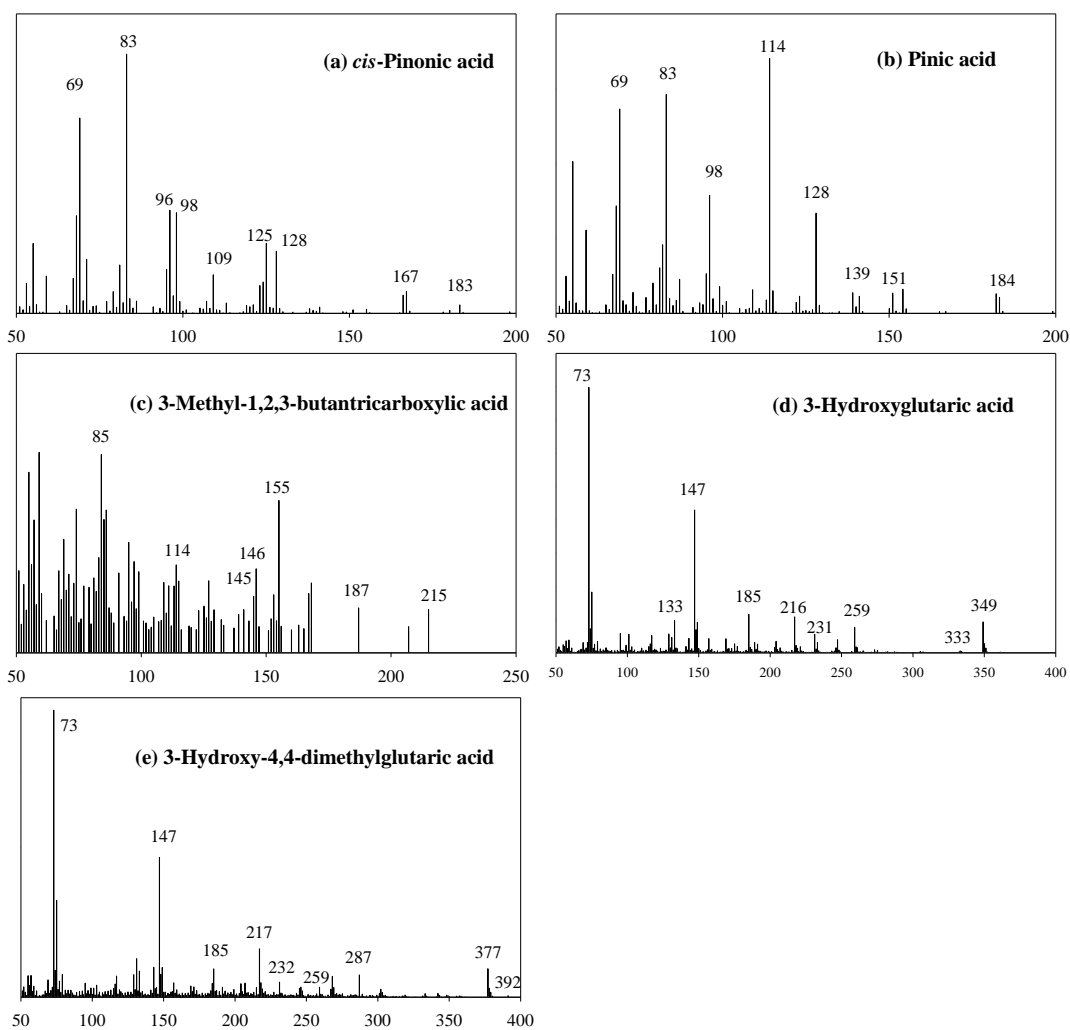
Figure 3 EI spectra of silylated isoprene SOA tracers

Figure 4 EI spectra of methylated (a-c) and silylated (d and e) monoterpene SOA tracers
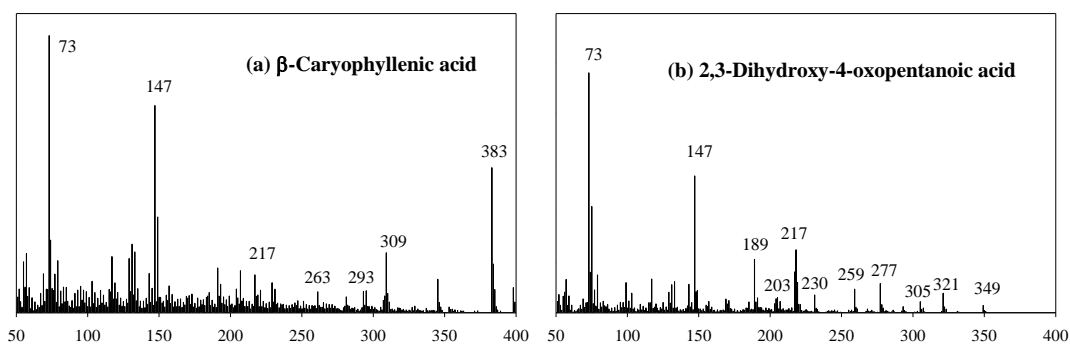

Figure 5 EI spectra of silylated SOA tracers from β-caryophyllene (a) and aromatics (b)

6. SOA tracers were quantified using surrogate standards, because analytical standards are not commercially-available for most of these compounds. The use of a surrogate standard introduces bias to the measurement, because it does not accurately represent the ionization and mass fragmentation of the target analyte. Moreover, the comparison of response factors of the five

surrogate standards (1-8 on page 7147) is irrelevant, because the issue is about the agreement of the response factors of the SOA tracers with the surrogates, not between the surrogates and each other. Consequently, the reported absolute concentrations of SOA tracers, are thus subject to large uncertainties, particularly when the structural match between the surrogate standard and analyte is poor (Stone et al. 2012). The manuscript should clearly state that presence and significance of this uncertainty and discuss the potential bias it may introduce to the results.

Reply: We appreciate the suggestion in uncertainty estimation for SOA tracer measurement. Stone et al., (2012) developed an empirical approach to estimate the error from surrogate quantification (E_Q) based on homologous series of atmospherically relevant compounds. These quantification errors (E_Q) are then propagated with the standard deviation of the field blank (E_{FB}) and error in spike recovery (E_R), and yield the measurement uncertainties (E_A) for SOA tracers. We have calculated the E_Q and E_A for SOA tracers using the method developed by Stone et al. (2012). As shown in Table 1, the errors from surrogate quantification (E_Q) ranged from 15% (2-methyltetrols) to 155% (β-caryophyllenic acid) in this study. Since SOA tracers were not detected in the field blanks, E_{FB} was 0 in this study. The spike recoveries of surrogate standards were used to estimate the E_R of tracers. The uncertainties in analyte measurement (E_A) were estimated in the range of 38% to 156%.

In the revised manuscript, we added a section "2.4 Estimation of measurement uncertainty" to discuss the uncertainty in tracer measurement (see below). Table 1 was added in the supplemental information file as Table S1.

"2.4 Estimation of measurement uncertainty

Since there is no commercial standard available for most SOA tracers (except *cis*-pinonic acid and pinic acid), the use of surrogate standards for quantification introduces additional error to measurement. Error in analyte measurement (E_A) is propagated from the standard deviation of the field blank (E_{FB}), error in spike recovery (E_R) and the error from surrogate quantification (E_Q):

$$E_A = \sqrt{E_{FB}{}^2 + E_R{}^2 + E_Q{}^2} \tag{1}$$

Since SOA tracers were not detected in the field blanks, E_{FB} was 0 in this study. The spike recoveries of surrogate standards were used to estimate the E_R of tracers which ranged from 1% (*cis*-pinonic acid) to 35% (erythritol). Stone et al. (2012) developed an empirical approach to estimate E_Q based on homologous series of atmospherically relevant compounds. The relative error introduced by each carbon atom (E_n) was estimated to be 15 %, each oxygenated functional group (E_f) to be 10% and alkenes (E_d) to be 60%. The errors introduced from surrogate quantification are treated as additive and are calculated as:

$$E_Q = E_n \Delta n + E_f \Delta f + E_d \Delta d \tag{2}$$

where Δn is the difference in carbon atom number between a surrogate and an analyte, Δf is the difference in oxygen-containing functional group between a surrogate and an analyte, Δd is the difference in alkene functionality between a surrogate and an analyte.

Table S1 shows the estimated uncertainties in tracer measurement. The errors from surrogate quantification (E_Q) ranged from 15% (2-methyltetrols) to 155% (β-caryophyllenic acid) in this study. Propagated with the error in recovery, the uncertainties in analyte measurement (E_A) were estimated in the range of 38% to 156%."

Table 1 Estimation of measurement uncertainty

Tracers	Tracer formula	Surrogates	Surrogate formula	E_Q (%)	E_R^a (%)	E_A (%)
cis-Pinonic acid	$C_{10}H_{16}O_3$	*cis*-Pinonic acid			1	
Pinic acid	$C_9H_{14}O_4$	Pinic acid			30	
3-Methyl-1,2,3-butantricarboxylic acid	$C_8H_{12}O_6$	*cis*-Pinonic acid	$C_{10}H_{16}O_3$	60	1	60
3-Hydroxyglutaric acid	$C_5H_8O_5$	*cis*-Pinonic acid	$C_{10}H_{16}O_3$	95	1	95
3-Hydroxy-4,4-dimethylglutaric acid	$C_7H_{12}O_5$	*cis*-Pinonic acid	$C_{10}H_{16}O_3$	65	1	65
cis-2-Methyl-1,3,4-trihydroxy-1-butene	$C_5H_{10}O_3$	Erythritol	$C_4H_{10}O_4$	85	35	92
3-Methyl-2,3,4-trihydroxy-1-butene	$C_5H_{10}O_3$	Erythritol	$C_4H_{10}O_4$	85	35	92
trans-2-Methyl-1,3,4-trihydroxy-1-butene	$C_5H_{10}O_3$	Erythritol	$C_4H_{10}O_4$	85	35	92
2-Methylglyceric acid	$C_4H_8O_4$	Erythritol	$C_4H_{10}O_4$	20	35	40
2-Methylthreitol	$C_5H_{12}O_4$	Erythritol	$C_4H_{10}O_4$	15	35	38
2-Methylerythritol	$C_5H_{12}O_4$	Erythritol	$C_4H_{10}O_4$	15	35	38
β-Caryophyllenic acid	$C_{13}H_{20}O_4$	Octadecanoic acid	$C_{18}H_{36}O_2$	155	17	156
2,3-Dihydroxy-4-oxopentanoic acid	$C_5H_8O_5$	Azelaic acid	$C_9H_{16}O_4$	90	11	91

[a] E_R is the difference between 100% and mean recovery of each surrogate standard.

7. Major issues arise when deviating from the quantification method of Kleindienst et al. (2007) for SOA tracers and using the SOA-tracer method for source apportionment. Namely, the f-SOC values (page 7154 line 15) were developed using the 5-ion quantification approach with a single-point calibration with ketopinic acid as the quantification standard and chemical ionization in the MS source. In this work, the authors have changed the MS detection method, internal standard, and surrogate standards, such that the f-SOC values cannot be directly applied. A large degree of bias is expectedly introduced, but is not quantifiable. The authors need to be realistic about the magnitude of error that this could introduce, which is likely on the order of 5-10 times different, rather than 23%.

Reply: As discuss above, we re-calculated the uncertainties in SOA tracer measurement and then re-estimated the uncertainties in SOC apportionment. Based on the E_A values in Table 1, the uncertainties in tracer analyses were up to 40% for SOA$_I$ (only MGA and MTLs involved for SOC estimation), up to 95% for SOA$_M$, 156% for SOA$_C$, and 91% for SOA$_A$. The uncertainties of f_{SOC} were reported to be 25% for isoprene, 48% for monoterpenes, 22% for β-caryophyllene and 33% for aromatics. Considering these factors, the uncertainties of SOC apportionment were calculated through error propagation. The RSD were 47% for SOC$_I$, 106% for SOC$_M$, 157% for SOC$_C$, and 96% for SOC$_A$. On average, the RSD of the reconstructed SOC (sum of the four precursors) was 51±11%.

In the revised manuscript, we addressed these as "The uncertainty in the SOA-tracer method is induced from the analysis of organic tracers and the determination of the conversion factors. Based on the E_A values in Table S1, the uncertainties in the tracer analyses were within 40% for SOA$_I$ (only MGA and MTLs involved for SOC estimation), 95% for SOA$_M$, 156% for SOA$_C$, and 91% for SOA$_A$. 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 apportionment were calculated through error propagation. The RSD were 47% for SOC$_I$, 106% for SOC$_M$, 157%

for SOC_C, and 96% for SOC_A. On average, the RSD of the reconstructed SOC (sum of the four precursors) was $51 \pm 11\%$."

8. Revision with respect to uncertainty in SOA tracer measurements is needed in section 3.2, page 7155 line 3.
Reply: In the revised manuscript, we have re-calculated the uncertainties of SOC apportionment with respect to uncertainty in SOA tracer measurement. The RSD were 47% for SOC_I, 106% for SOC_M, 157% for SOC_C, and 96% for SOC_A. On average, the RSD of the reconstructed SOC (sum of the four precursors) was $51 \pm 11\%$. (See discussions above)

9. Clarify the "paired duplicate samples" described in section 2.3 line 16. Are these duplicate samples of ambient aerosol that were collected in parallel? Or were these extracts that were split and analyzed twice?
Reply: These duplicate samples were ambient aerosol that were collected in parallel. We clarify it in the revised manuscript "The relative differences for target compounds in samples collected in parallel (n=6) were all below 15%."

10. Were the absolute concentrations of SOA tracers corrected for the less than optimal recoveries reported in section 2.3? Or is this another source of error in the ambient measurements?
Reply: No, the absolute concentrations of SOA tracers did not correct using recoveries. The recoveries of target compounds ranged from 65% (erythritol) to 101% (*cis*-pinonic acid), suggesting that the errors in analyte recovery should be within 35% in this study. In the revised manuscript, the recovery data were used to calculate the errors in analyte recovery (E_R) for SOA tracers (See Table 1). Then, uncertainties in SOA tracer measurement (E_A) were estimated through error propagation from E_R and the error from surrogate quantification (E_Q).

11. The sentence on page 7149 (lines 9-10) should be moved to follow the description of the model.
Reply: In the revised manuscript, we moved the sentence to follow the description of the model.

12. The authors should work towards developing a deeper discussion of the monoterpene SOA tracers using knowledge of first and multi-generation oxidation products of monoterpenes (Glasius et al. 2000; Jaoui et al. 2005; Szmigielski et al. 2007) and reaction pathways (Eddingsaas et al. 2012) as has been done for isoprene.
Reply: We appreciate the suggestion. Previous study proposed that *cis*-pinonic acid and pinic acid were the first-generation products of SOA_M, and only formed under low-NOx conditions (Eddingsaas et al. 2012). The dominance of *cis*-pinonic acid and pinic acid among SOA_M tracers at the remote NC site indicated that SOA_M there was mainly formed under low-NOx conditions. Moreover, *cis*-pinonic acid and pinic acid (P) could be further photo-degraded to higher-generation products, e.g. 3-methyl-1,2,3-butanetricarboxylic acid (M) (Glasius et al. 2000; Jaoui et al. 2005; Szmigielski, et al., 2007). And the ratio of *cis*-pinonic acid plus pinic acid to 3-methyl-1,2,3-butanetricarboxylic acid (P/M) could be applied to trace the aging of SOA_M (Ding et al., 2011; Gómez-González et al., 2012). In the fresh chamber produced α-pinene SOA samples, the ratios of P/M were reported in the range of 1.51 to 3.21 (Offenberg, et al., 2007). In this study, the ratio of P/M averaged 16.7 ± 20.9. Thus, SOA_M was generally fresh at the NC site. Figure 6 presents a negative correlation between P/M and temperature (r=-0.560, p=0.008). Since temperature has positive influence on photo-reaction rates, the higher temperature during the summer could accelerate the photochemistry in the air and result in P to M conversion being more efficient. Thus, SOA_M in the summer was more aged than that in the winter.

In the revised manuscript, we added a figure (Figure 6) to show the negative correlation

between P/M and temperature, and addressed the discussions above "Previous study proposed that *cis*-pinonic acid and pinic acid (P) were the first-generation products of SOA$_M$ and only formed under low-NOx conditions (Eddingsaas et al. 2012). The dominance of *cis*-pinonic acid and pinic acid among SOA$_M$ tracers at the remote NC site indicated that SOA$_M$ there was mainly formed under low-NOx conditions. Moreover, *cis*-pinonic acid and pinic acid could be further photo-degraded to higher-generation products, e.g. 3-methyl-1,2,3-butanetricarboxylic acid (M) (Glasius et al. 2000; Jaoui et al. 2005; Szmigielski, et al., 2007). And the ratio of *cis*-pinonic acid plus pinic acid to 3-methyl-1,2,3-butanetricarboxylic acid (P/M) could be applied to trace the aging of SOA$_M$ (Ding et al., 2011; Gómez-González et al., 2012). In the fresh chamber produced α-pinene SOA samples, the ratios of P/M were reported in the range of 1.51 to 3.21 (Offenberg, et al., 2007). In this study, the ratio of P/M averaged 16.7 ± 20.9. Thus, SOA$_M$ was generally fresh at the NC site and should be mainly formed from local precursors. Figure 6 presents a negative correlation between P/M and temperature (r=-0.560, p=0.008). Higher P/M ratios were observed in the fall and the winter, and lower P/M ratios occurred in the spring and the summer. Since temperature has positive influence on photo-reaction rates, the higher temperature during the summer could accelerate the photochemistry in the air and result in P to M conversion being more efficient. Thus, SOA$_M$ in the summer was more aged than that in the winter."
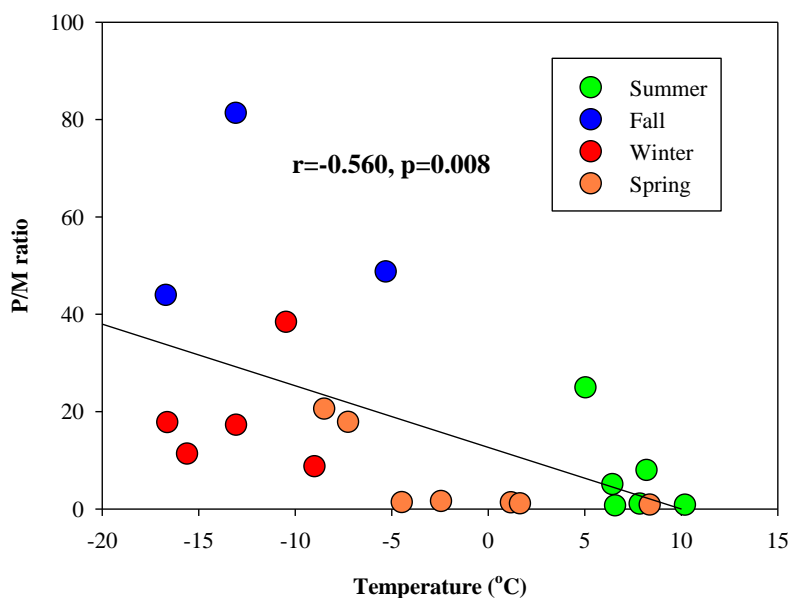

Figure 6 Negative correlation between P/M ratio and temperature

Ding, X., Wang, X., and Zheng, M.: The influence of temperature and aerosol acidity on biogenic secondary organic aerosol tracers: Observations at a rural site in the central Pearl River Delta region, South China, Atmos. Environ., 45, 1303-1311, 2011.
Eddingsaas, N. C., Loza, C. L., Yee, L. D., Chan, M., Schilling, K. A., Chhabra, P. S., Seinfeld, J. H., and Wennberg, P. O.: α-pinene photooxidation under controlled chemical conditions – Part 2: SOA yield and composition in low- and high-NOx environments, Atmos. Chem. Phys., 12, 7413-7427, 2012.
Gómez-González, Y., Wang, W., Vermeylen, R., Chi, X., Neirynck, J., Janssens, I. A., Maenhaut, W., and Claeys, M.: Chemical characterisation of atmospheric aerosols during a 2007 summer field campaign at Brasschaat, Belgium: sources and source processes of biogenic secondary organic aerosol, Atmos. Chem. Phys., 12, 125-138, 2012.
Glasius, M., Lahaniati, M., Calogirou, A., Di Bella, D., Jensen, N. R., Hjorth, J., Kotzias, D., and Larsen, B. R.: Carboxylic acids in secondary aerosols from oxidation of cyclic monoterpenes by

ozone, Environ. Sci. Technol., 34, 1001-1010, 2000.

Jaoui, M., Kleindienst, T. E., Lewandowski, M., Offenberg, J. H., and Edney, E. O.: Identification and quantification of aerosol polar oxygenated compounds bearing carboxylic or hydroxyl groups. 2. Organic tracer compounds from monoterpenes, Environ. Sci. Technol., 39, 5661-5673, 2005.

Offenberg, J. H., Lewis, C. W., Lewandowski, M., Jaoui, M., Kleindienst, T. E., and Edney, E. O.: Contributions of toluene and α-pinene to SOA formed in an irradiated toluene/α-pinene/NOx/ air mixture: Comparison of results using ^{14}C content and SOA organic tracer methods, Environ. Sci. Technol., 41, 3972-3976, 2007.

Szmigielski, R., Surratt, J. D., Gómez-González, Y., Veken, P. V. d., Kourtchev, I., Vermeylen, R., Blockhuys, F., Jaoui, M., Kleindienst, T. E., Lewandowski, M., Offenberg, J. H., Edney, E. O., Seinfeld, J. H., Maenhaut, W., and Claeys, M.: 3-Methyl-1,2,3-butanetricarboxylic acid: An atmospheric tracer for terpene secondary organic aerosol, Geophys. Res. Lett., 34, L24811, 10.1029/2007GL031338, 2007.

13. The conclusion that biogenic SOC "dominated over anthropogenic SOC" is not robust, in part because only a single organic molecule is being used as a tracer of anthropogenic VOC – DHOPA. The specificity of DHOPA to anthropogenic sources is not fully established (Kleindienst et al. 2004). For example, biomass burning is a major source of toluene in many parts of the world (Lewis et al. 2013). Hence, DHOPA may be an indicator of the processing of biomass burning emissions, and not a measure of urban pollutants from solvent and fossil fuel use. The limitations of using a single, and potentially non-specific tracer for anthropogenic SOA must be discussed and the conclusions restated.

Reply: Our data suggested that biogenic SOC dominated over aromatic SOC (SOC$_A$). DHOPA has been widely used to access the aromatic SOA in the United States (Kleindienst et al. 2007, Stone et al. 2009, Lewandowski et al. 2013), and China (Hu et al. 2008, Guo et al. 2012, Peng et al. 2013, Ding et al. 2014). Since aromatics are mainly emitted from anthropogenic sources, and are major anthropogenic SOA precursors, we think aromatic SOA can reflect the majority of anthropogenic SOA. We admit there are limitations using a single tracer for anthropogenic SOA. In the revised manuscript, we changed the conclusion "biogenic SOC dominated over anthropogenic SOC" to "biogenic SOC dominated over SOC$_A$"

We did measure biomass burning tracer, levoglucosan in our samples. As shown in Figure 7, the monthly variation trend of levoglucosan was quite different from that of DHOPA. And there was no correlation between DHOPA and levoglucosan (p>0.05) at the NC site (Figure 8). These indicated that DHOPA was not mainly from the processing of biomass burning emission at the NC site. As discussed in the ACPD manuscript, the higher levels of DHOPA existed when air masses mainly came from the upwind Indian subcontinent (the Bangladesh and the northeastern India) where high population density and high levels of anthropogenic pollutants (AOD, CO, and N$_2$O) were observed (See our response to the second comment by Reviewer #1). Thus, we believe DHOPA observed in this study should be not mainly from local biomass burning but from long-range transport.

In the revised manuscript, we addressed these as "Besides urban emissions from solvent and fossil fuel use, biomass burning is an important source of aromatics in many parts of the world (Lewis et al. 2013). The local dung or biomass burning (Duo et al. 2015; Xiao et al. 2015) may be potential sources of aromatics in the TP. Hence, DHOPA may come from the processing of biomass burning emissions. Figure 7 exhibits the monthly variation of biomass burning tracer, levoglucosan during our sampling. The concentrations of levoglucosan ranged from 0.82 ng m^{-3} (October 2012) to 4.55 ng m^{-3} (April 2013) with a mean of 1.87±1.14 ng m^{-3}. Apparently, the monthly variation trend of levoglucosan was quite different from that of DHOPA. And there was no correlation between DHOPA and levoglucosan (p>0.05) (Figure S6). These indicated that DHOPA was not mainly from the processing of biomass burning emission at the NC site. Since

there was few anthropogenic source near the remote NC site, the SOA$_A$ tracer should be not locally formed but mainly transported from upwind regions." The figure of monthly variation of legoglucosan and the scatter plot of DHOPA and levoglucosan were added in the revised manuscript (Figure 7) and the supplemental information file (Figure S6), respectively.
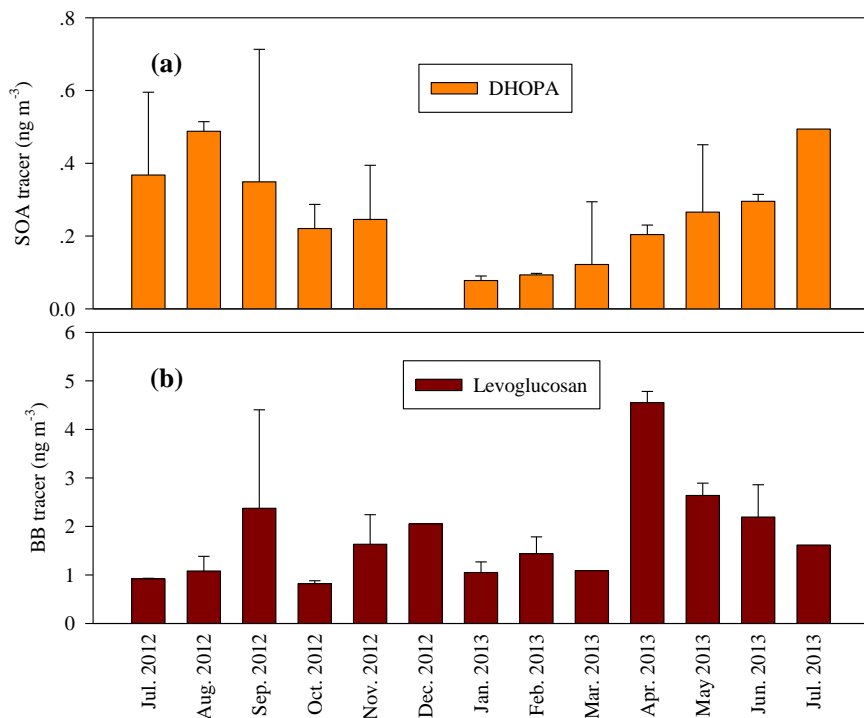

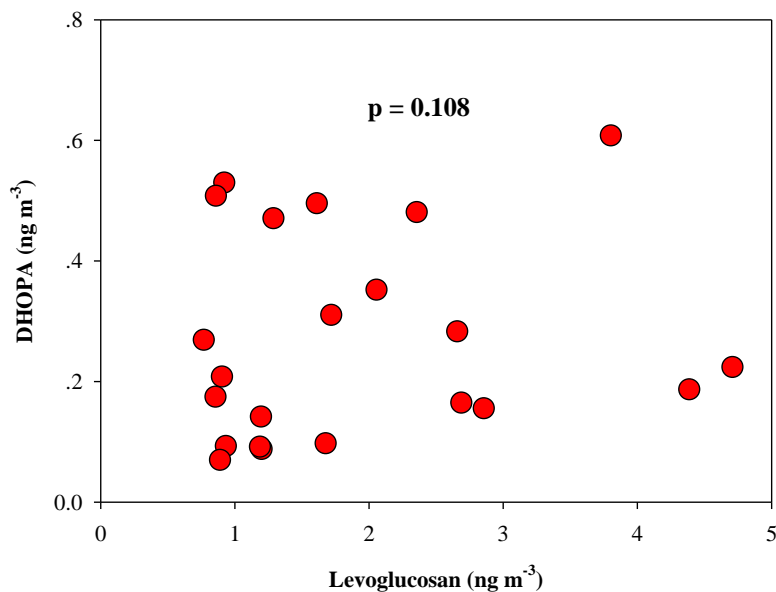Figure 7 Monthly variations of DHOPA (a) and levoglucosan (b)

Figure 8 Scatter plot of DHOPA and levoglucosan

Kleindienst, T. E., Jaoui, M., Lewandowski, M., Offenberg, J. H., Lewis, C. W., Bhave, P. V., and Edney, E. O.: Estimates of the contributions of biogenic and anthropogenic hydrocarbons to secondary organic aerosol at a southeastern US location, Atmos. Environ., 41, 8288-8300, 2007.

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14. Clarification needed on page 7154 line 11-12 – What specifically has been done to show that the SOA tracer approach provides "reasonable results"?

Reply: Previous studies have compared the estimated SOC by SOA-tracer method and other techniques. Lewandowski et al. (2008) found that the measured OC in the midwestern United States could be fully explained by primary OC from chemical mass balance (CMB) model plus SOC from the SOA-tracer method, suggesting that the secondary organic tracer technique could be a valuable method for SOC estimation. Kleindienst et al. (2010) further compared the estimated SOC by the SOA-tracer method and other four independent methods (multiple regressions, CMB, carbon isotope and EC-tracer) in the southeastern United States, and found that these five methods matched well. Our previous study in the Pearl River Delta, south China found SOC levels estimated by the SOA-tracer method were not only consistent with but also correlated well with those by EC-tracer method in summer, (Ding et al., 2012). The SOC apportionment results were also comparable between the SOA-tracer method and the positive matrix factorization (PMF) model in Hong Kong (Hu et al. 2010). All these demonstrate that the SOA tracer approach can provide reasonable results.

In the revised manuscript, we added these statements to show that the SOA tracer approach provides reasonable results. "Lewandowski et al. (2008) found that the measured OC in the midwestern United States could be fully explained by primary OC from chemical mass balance (CMB) model plus SOC from the SOA-tracer method, suggesting that the secondary organic tracer technique could be a valuable method for SOC estimation. Kleindienst et al. (2010) further compared the estimated SOC by the SOA-tracer method and other four independent methods (multiple regressions, CMB, carbon isotope and EC-tracer) in the southeastern United States and found that these five methods matched well. Our previous study in the Pearl River Delta found SOC levels estimated by the SOA-tracer method were not only consistent with but also correlated well with those by EC-tracer method in summer, (Ding et al., 2012). The SOC apportionment results were also comparable between the SOA-tracer method and positive matrix factorization (PMF) model in Hong Kong (Hu et al. 2010)."

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15. Page 7142 line 18: replace "emission and tracers partitioning." With "emission and gas-particle partitioning."
Reply: Replaced

16. Page 7142, line 28: revise to "to estimate secondary"
Reply: Revised

17. Page 7155, line 17 "not measure OC"
Reply: Revised

18. Table 1 – improve the "Month column" The six numbers are not easily interpreted.
Suggest writing out the month and year, e.g. July 2012
Reply: Revised as suggested

19. Figure 2 – suggest replacing numerical dates on the x-axis with "July 2012" to improve readability.
Reply: Revised as suggested

20. Table 1 – Indicate that temp and RH are monthly averages.
Reply: We indicated that "temperature and RH are monthly averages" as the note of Table 1.

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