

## ***Interactive comment on “Characterization of organic nitrogen in aerosols at a forest site in the southern Appalachian Mountains” by Xi Chen et al.***

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Received and published: 9 April 2018

Point to point reply to reviewer 2's comments: This paper presents the analytical results of water-soluble organic nitrogen (WSON) for both bulk and related molecular compounds in PM 2.5 filter samples collected at a remote montane forest site in the U.S. The authors present the season variation of WSON and related organic molecular compounds to characterize aerosol WSON and investigate its possible sources. Combination of bulk WSON and molecular tracer compounds related to WSON and WSOC obtained in the forest environment provides new insights into our understanding on aerosol WSON particularly from terrestrial biogenic sources. While the data presented

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are valuable, there are some important issues that need to be worked out and clarified before I recommend its publication in ACP.

Specific comments Comment (1) One of my concern is on the interpretation for the positive correlation between biogenic SOA tracers and ambient temperature (Lines 470-476, 562-565). The authors conclude that such a relationship indicates temperature dependence of “oxidation.” It may be true to some extents, but how about the temperature dependence of VOC emissions? Most of terpenes generally show temperature dependence of emission, which can also explain the correlation shown in this manuscript.

Response: The positive correlation referred to here is not between the abundance of the biogenic SOA tracers with temperature, rather we are using the relative abundance (concentration ratios) with temperature instead. It is stated in the text “To assess the extent of aging, concentration ratios of higher generation oxidation products (C<sub>8</sub>H<sub>12</sub>O<sub>6</sub>, m/z 203 and C<sub>8</sub>H<sub>12</sub>O<sub>5</sub>, m/z 187) to early oxidation fresh SOA products (C<sub>8</sub>H<sub>12</sub>O<sub>4</sub>, m/z 171 and C<sub>10</sub>H<sub>16</sub>O<sub>6</sub>, m/z 231) are calculated.” We do agree that most terpene emissions are temperature dependent, but we think the relative abundance of higher generation products to early oxidation products probably will cancel out such effects.

Comment (2) The authors use the term “aged biogenic SOA” (e.g., Lines. 560-565 and others) in the text. Please add more discussion about specific time scale on this aging (hours, days?). This should be discussed relative to the time scale of transport (e.g., vertical mixing within the forest canopy or between the canopy and the above atmosphere, horizontal transport, etc.).

Response: Under atmospherically relevant conditions, the lifetime of  $\alpha$ -pinene SOA was reported as several days (4-7days) as a result of heterogeneous and condensed phase oxidation processes (Epstein et al., 2014). In addition, the precipitation frequency during spring and summer at the Coweeta site was quite high (as in days) and we think the aged biogenic SOA probably had a time scale of aging of days be-

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fore scavenged by the precipitation. Figure 1C also shows such scavenging effect after precipitation on WSOC (a drop in WSOC right after precipitation occasions). A brief discussion on this topic is now added to the text at Line 495 “Based on typical chemical lifetime of biogenic SOA by OH oxidation and the precipitation frequency at the Coweeta site, biogenic SOA at Coweeta probably had an atmospheric lifetime of several days before depletion by oxidation processes (Epstein et al., 2014) and/or scavenging by precipitation.”

Comment (3) Nitro-aromatics: In section 3.3., the authors conclude that the contribution of nitroaromatics to WSON was generally “small,” whereas they state potential importance of nitro-aromatics to the atmospheric N deposition budget (L.507-509) in section 3.6. These statements do not seem to be consistent and confusing.

Response: L507-509 “Nitro-aromatics and nitrooxy-organosulfates were estimated to account for as much as 28% of WSON, which reflected the abundance and potential importance of these groups of species to the atmospheric N deposition budget.” Here we are referring to the contribution of Nitro-aromatics and nitrooxy-organosulfates combined to N deposition budget, not nitro-aromatics alone. The sentence is now revised to “Nitro-aromatics and nitrooxy-organosulfates combined were estimated to account for as much as 28% of WSON, which reflected the abundance and potential importance of these groups of species to the atmospheric N deposition budget.”

Comment (4) Lines 383-384: If the event cannot be attributed to local burning, then what is the most likely origin (source location)? “Long range transport” is not enough to explain the source of the observed particles in this event.

Response: The following analysis and discussion is now included at line 398: Clustering of backward trajectories (120hr duration for individual trajectories; 48 total trajectories covering the two-day event) suggests that northeast Georgia (shown in supplemental information Figure S5) is the most likely origin of the biomass burning event observed on October 24th and 25th.

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Comment (5) Section 2.3: The authors should describe the measurement uncertainties for each analysis. This is particularly important for the analysis of WSON, whose measurement uncertainty includes propagation of errors of WSTN, NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup>, : : .

Response: Yes, the measurement uncertainty of WSON includes propagation of errors of WSTN, NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup> and NO<sub>2</sub><sup>-</sup>. We have included the estimated measurement uncertainties in the text at Line 332: “The measurement uncertainty of WSON was estimated to be  $\pm 30\%$  from error propagation of WSTN (2%), NH<sub>4</sub><sup>+</sup> (1%), NO<sub>3</sub><sup>-</sup> (1%) and NO<sub>2</sub><sup>-</sup> (1%).”

Comment (6) Figure 6: I think that the author should show time series of integrated factor contributions vs. the measured WSOC concentrations to show how well the PMF reproduced the measurements. Then the authors should show fractional contribution of each factor to WSOC in the time series as they discuss it in the text.

Response: A linear regression plot for integrated factor contributions with measured WSOC is now included in the supplemental information as Figure S6. Linear regression coefficients are also provided in the figure. In Figure 6, the mean fractional contributions of each factor to WSOC are now included.

Comment (7) The authors use the term “N/C ratio” in the manuscript: Lines 42,43, 517, 524, 592,and 593. Should this term be “(WS)ON/OC ratio?” “N/C” includes inorganic N and elemental C.

Response: Yes, we are referring to WSON/WSOC ratio. We have revised the term “N/C ratio” throughout the manuscript to “WSON/WSOC ratio”.

Comment (8) Lines 522-525: The identified-ON/WSON ratios also show a seasonal difference (Table 4). Can the authors add a few more statement on this difference in terms of unidentified compounds?

Response: The following discussion is included in the text regarding seasonal differences observed for identified-ON/WSON: “Moreover, identified ON/WSON ratio was

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estimated to be 1.0, 2.0 and 4.4 for spring, summer and fall, respectively. Such differences further suggest much more unidentified WSON compounds exist in spring when organic N was most enriched from biological processes.”

Minor comments Comment (9) Abstract: The authors should specify that the sampled aerosols are PM2.5.

Response: Yes, PM2.5 is specified now in the abstract on line 24.

Comment (10) L.312: Please define “[O3]” here.

Response: [O3] definition is added as ozone concentration.

Comment (11) L.394: Correct “extreme” to “extremely.”

Response: “extreme” is changed to “extremely”.

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Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2018-34>, 2018.

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