

We appreciate the referee's valuable comments on our work. Our responses to the specific comments are given below.

Responses to the comments of Referee#2:

Comment 1: In the abstract and the throughout the text it needs to be made clear what aerosol size range these results pertain to; specifically the WSOC reported is TSP. (An actual estimate of the upper size limit of the filter sampling system would be very useful). This is important since it tends to explain why the authors see significant contributions from both secondary and primary sources, whereas one may expect significant less contributions if the focus was say, fine WSOC. As a further example, the statement (page 30781 lines 26. . .) Quoted from the paper: In summary, the present results indicate that at this forest site, the SOA formations associated mainly with α -/ β -pinene oxidation and the primary emissions from biological sources contribute almost equally to the peak of WSOC in the growing season of early summer as well as in autumn (end quote). It needs to be specifically stated that TSP WSOC is being discussed.

***Reply 1:* We agree that it is important to clarify the size range of WSOC being discussed. As suggested by the referee, we have added specific statements to the Abstract and Conclusions, as well as in the text, indicating that WSOC in this study was measured and discussed in terms of TSP. With regard to the upper size limit for the sampling, it varies with wind speed and wind direction. High-volume samplers do not provide definitive particle size information. Instead, an effective cutoff point of 30- μ m in aerodynamic diameter is frequently assigned to a standard high-volume sampler.**

Comment 2: Also, when reading the abstract my impression of the canopy floor is one barren of living vegetation and composed mainly of decaying plant matter. In this forest, it appears there is significant mass of living photosynthetic plants at or near the surface (page 30776 lines 8-10). The last line of the Conclusions provides this information in a nice format and I suggest should be included in the abstract.

***Reply 2:* We have already mentioned in the Abstract that “the forest floor, including ground vegetation and soil, acts as a significant source of WSOC,” as provided in the Conclusions section. Because further details about the forest floor are already given in section 2.1., we do not think that an additional description is needed in the Abstract.**

Comment 3: A few more lines describing the NEE method (Section 2.3) would be useful.

For example, details on the CO₂ measurement to determine eddy covariance fluxes, how was the change in CO₂ storage in air mass from ground to eddy flux measurement determined. Although more details are provided in the reference, the reader needs to know more on what is essentially a mass balance analysis, as it is an important component of this paper.

Reply 3: We agree that the NEE method is an important component of our paper. Based on this comment, we have added more information about the NEE method, including the CO₂ measurements, in the revised manuscript:

“The eddy-covariance fluxes were obtained in a flux tower using a three-dimensional (3-D) sonic anemothermometer and an infrared CO₂/H₂O analyzer (Li-6262; LiCor, Lincoln, NE, USA) (Nakai et al., 2003). To quantify the storage of CO₂ below the altitude level of the eddy measurement, vertical profiles of CO₂ concentrations were measured at five altitude levels (2.7, 10.5, 16.3, 20.1, and 29.6 m above the ground surface) via sampling inlets mounted on the tower. Inside a shelter at the bottom of the tower, the sample air from each altitude was introduced into the CO₂/H₂O analyzer by rotation using an automatically controlled solenoid valve manifold. In the present study, an upward CO₂ flux is considered to be positive, which means that a negative NEE value indicates net uptake of CO₂ by the forest ecosystem. Further details of the NEE derived here are given elsewhere (Nakai et al., 2003).”

Comment 4: The Method section also needs a clear description of the principle behind the $\delta^{13}\text{C}_{\text{WSOC}}$ measurements; what does it tell you and why or how does this measurement provide this information.

Reply 4: As suggested, we have added a description of the $\delta^{13}\text{C}_{\text{WSOC}}$ measurements and have moved this information to the Introduction:

“The stable carbon isotope ratio ($\delta^{13}\text{C}$) is a powerful tool for source determination based on the distinctive signals of different aerosol carbon fractions. For example, C₃ plants, which use the Calvin-Benson cycle as a metabolic pathway for carbon fixation in photosynthesis, have $\delta^{13}\text{C}$ values in the range of -23 to -30‰ . All trees and most shrubs, grasses, and sedges in mid-latitude and boreal regions belong to the C₃ class of plants. The most common application of carbon isotopic measurements for aerosols is for the measurement of total carbon (TC) (e.g., Cachier et al., 1986; Turekian et al., 2003). In contrast, very few studies have used the $\delta^{13}\text{C}$ of WSOC for source apportionment (Fisseha et al., 2009; Kirillova et al., 2010). The WSOC-specific $\delta^{13}\text{C}$ analysis, combined with biogenic molecular markers, allows for source apportionment of aerosol WSOC in forested areas.”

Comment 5: Could not find any discussion of the MSA measurement?

Reply 5: We have added discussions of the importance and methods of the MSA measurement to the Introduction and Experimental sections, respectively.

In the Introduction:

“Moreover, methanesulfonic acid (MSA) was found at the forest site, and its possible sources are discussed. MSA is formed by the photooxidation of dimethylsulfide (DMS) and is typically found in marine-biologically influenced aerosols; measurements of MSA in terrestrial regions are limited (e.g., Lukács et al., 2009).”

In the Experimental section:

“For determination of inorganic ions, another filter cut was extracted with Milli-Q water. The total extract was filtrated through a membrane disc filter, and major anions and cations as well as MSA were determined using a Metrohm ion chromatograph (Model 761 compact IC; Metrohm, Herisau, Switzerland) (Miyazaki et al., 2009).”

Comment 6: No discussion on issues, if any, with measurement blanks, possible sampling artifact issues with integrated filters; are any expected, is this measurement include semivolatile components, is any semi-volatile WSOC expected.

Reply 6: We have added descriptions of our measurement blanks and possible sampling artifacts to the revised manuscript, as follows:

“The WSOC value for a filter punch of a field blank corresponds to ~7% of the average WSOC concentration of the ambient samples. All WSOC data presented here have been corrected against field blanks. Interferences may possibly occur due to the following interactions with vapor-phase compounds: adsorption of VOCs that contribute to the aerosol measurements (positive artifact) and volatilization of semi-VOCs during the sampling (negative artifact).”

Comment 7: In figure 2 a very distinct peak in WSOC is seen in May-June 2010, and with additional data, is attributed to the growing season. However, measurements were made at the beginning of the study (June 1, 2009), but which appear not to show this large peak. Can the authors be sure that the may 2010 are a consistent feature of this forests seasonal trends? The same is true of MSA, it appears unique to 2010 since a similar large peak was not observed in 2009. Thus the discussion of section 3.3 seems to only apply to the rather unique year of 2010. Some discussion on this should be included.

Reply 7: In fact, we started the first sampling at the end of June 2009, and we do not have sufficient data to discuss WSOC in the 2009 growing season. However, the increase in the concentrations of WSOC and MSA in autumn was apparent in both 2009 and 2010. Moreover, $\delta^{13}\text{C}_{\text{WSOC}}$ exhibited similar seasonal cycles in both years. Therefore, we believe that the seasonal trends of WSOC and other parameters are not unique to the year 2010. We agree that in order to discuss the representativeness of the data at this site, more data are needed at least over a time scale of a few years. Taking the referee's comment into account, we have mentioned this point in the revised manuscript:

“More data including both BSOA tracers and VOCs near the forest floor over a time scale of a few years are needed to discuss the representativeness of the data at this site in future study.”

Comment 8: In the factor analysis, as summarized in Figure 4, and throughout the paper when seasonal trends are discussed, are all the data used? That is there are some seasons in which sampling occurred twice and some seasons where sampling occurred only once.

Reply 8: In the PMF analysis, we used approximately half of the total data because of the complexity of the tracer analysis. Nevertheless, the sampling data used for the PMF analysis spanned a 2-week period of each month, and we believe that these data were representative in terms of each season.

References

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