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#4:

Comment 1: Include some references in the first paragraph of the introduction.

Reply 1: We have added the following references to the Introduction, as suggested: Kavouras et al., 1998; Kulmala et al., 2004; Tunved et al., 2006; Peñuelas and Staudt, 2010; Mahowald, 2011; and Bonan, 2008.

Comment 2: Page 30777 (lines 12-16): the paragraph "The stable carbon isotopic ratio (δ 13C) is a powerful tool for source determination based on the distinctive signals of different aerosol carbon fractions. The most common carbon isotopic application for aerosol is for total carbon (TC) (e.g., Cachier et al., 1986; Turekian et al., 2003). In contrast, very few studies have used the δ 13C of WSOC for source apportionment (Fisseha et al., 2009; Kirillova et al., 2010)." Should be under the introduction not in experimental part

Reply 2: In response to the referee's comment, the paragraph with some modification has been moved to the Introduction as follows:

"The stable carbon isotope ratio (δ^{13} 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 δ^{13} 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 δ^{13} C of WSOC for source apportionment (Fisseha et al., 2009; Kirillova et al., 2010). The WSOC-specific δ^{13} C analysis combined with biogenic molecular markers allows a source apportionment for aerosol WSOC in forested areas."

Comment 3: Page 30777, line 21-22: a short description on how the extracted WSOC was concentrated to 40ml would be useful because this process can potentially affect the stable carbon isotope ratio of water soluble organic carbon.

Reply 3: We have added a description of the method used to concentrate the extracted WSOC to the revised manuscript:

"The extracted samples were concentrated by rotary evaporation, and 40 µl of each sample were transferred to be absorbed onto 10 mg of pre-combusted Chromosorb in a pre-cleaned tin cup."

Comment 4: Are there any blank measurements? If yes, please provide blank values.

Reply 4: We have provided a blank value for determining the WSOC concentration: "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."

We could not measure a blank value for $\delta^{13}C_{WSOC}$ because the carbon mass on the blank filter was too small to be detected by the instrument.

Comment 5: Page 30779, line 5-9: the value reported on Kirillova et al., 2010 is for commercial cis-pinonic acid and sucrose. Therefore, it should not be used as a reference for the stable isotopic composition of a tracer compound. The discussion on the stable isotopic composition of the water soluble compounds also gave me an impression that the authors are making an assumption that there is no difference in the δ 13C of primary and secondary organic aerosols. Although I am aware of the scarcity of data in this subject, it should be addressed to encourage future work.

Reply 5: Although cis-pinonic acid and sucrose are commercial standards, Kirillova et al. (2010) reported that radiocarbon (Δ^{14} C) analysis indicates that they lack any significant contribution from fossil carbon (i.e., they are indeed modern). Our discussion does not fully depend on the commercial standards described by Kirillova et al. (2010). The δ^{13} C of WSOC originating from C3 plants and plant wax n-alcohols in ambient aerosols (Conte and Weber, 2002; Kirillova et al., 2010) also supports a dominant input of precursor molecules from C3 plants to WSOC. Therefore, we believe that commercial cis-pinonic acid and sucrose are still useful in our discussion and have decided to keep them.

The value of δ^{13} C alone cannot differentiate between primary and secondary WSOC in our data. In response to the comment, we have clarified this point in the revised manuscript:

"Although $\delta^{13}C_{WSOC}$ alone cannot differentiate between primary and secondary WSOC in our data, the results indicate a dominant input of precursor molecules from C3 plants to WSOC from May to October."

Comment 6: Some anthropogenic sources also have a $\delta 13C$ value similar to C3 plants (Widory et al., 2004). Therefore, it would be very helpful to included some discussion why

the authors believe the δ 13C value of the WSOC during the growing season is an indicative of precursor molecule from C3 plant and not influenced by the anthropogenic sources

Reply 6: We measured several anthropogenic tracers such as hopan and found that their concentrations in the growing season were substantially low (some were below the detection limit). In addition, the predominant local wind direction and speed in this season indicated that the sampled aerosols originated mostly from the forested area, as discussed in section 2.1. We believe that the low concentrations of the anthropogenic tracers together with the local wind data support the idea that the δ^{13} C values in the growing season are attributable mainly to C3 plants, rather than anthropogenic sources. We have added the following discussion to the text: "It should be noted that some anthropogenic sources have δ^{13} C values similar to those of C3 plants in aerosols (e.g., Widory et al., 2004). However, an insignificant contribution of anthropogenic sources to the observed aerosols in the growing season is suggested by the substantially low concentrations of anthropogenic tracers (such as hopan some of which were not detected). Moreover, the predominant local wind direction and speed in this season indicated that the sampled aerosols originated mostly from the forested area, as discussed in section 2.1. The low concentrations of anthropogenic tracers together with the local wind data support the idea that the $\delta^{13}C$ values in the growing season are attributable mostly to C3 plants, rather than anthropogenic sources."

Technical

Comment 7: In the title, replace "Isotopic ratios" with either "isotopic composition" or "isotope ratio".

Reply 7: As suggested, "isotopic ratios" is replaced by "isotopic composition."

References

Bonan, G.: Forests and climate change: Forcings, feedbacks, and the climate benefits of forests, Science, 320, 1444–1449, 2008.

Cachier, H., Buat-Ménard, M. P., Fontugne, M., and Chesselet, R.: Long-range transport of continentally-derived particulate carbon in the marine atmosphere: Evidence from stable carbon isotope studies, Tellus, Ser. B, 38, 161–177, 1986.

Conte, M. H., and Weber, J. C.: Plant biomarkers in aerosols record isotopic discrimination of terrestrial photosynthesis, Nature, 417, 639–641, 2002.

Fisseha, R., Saurer, M., Jäggi, M., Siegwolf, R., T., W., Dommen, J., Szidat, S., Samburova, V., and Baltensperger, U.: Determination of primary and secondary sources of organic acids and carbonaceous aerosols using stable carbon isotopes, Atmos. Environ., 43, 431–437, 2009.

Kavouras, I. G., Mihalopoulos, N., and Stephanou, E. G.: Formation of atmospheric particles from organic acids produced by forests, Nature, 395, 683–686, 1998.

Kirillova, E. N., Sheesley, R. J., Andersson, A., and Gustafsson, Ö: Natural abundance ¹³C and ¹⁴C analysis of water-soluble organic carbon in atmospheric aerosols, Anal. Chem., 82, 7973–7978, 2010.

Kulmala, M., Suni, T., Lehtinen, K. E. J., Dal Maso, M., Boy, M., Reissell, A., Rannik, Ü., Aalto, P., Keronen, P., Hakola, H., Bäck, J., Hoffmann, T., Vesala, T., and Hari, P.: A new feedback mechanism linking forests, aerosols, and climate, Atmos. Chem. Phys., 4, 557–562, 2004.

Mahowald, N.: Aerosol indirect effect on biogeochemical cycles and climate, Science, 334, 794–796, 2011.

Peñuelas, J. and Staudt, M.: BVOCs and global change, Trends in Plant Sci., 15, 133–144, 2010.

Tunved, P., Hansson, H.-C., Kerminen, V.-M., Ström, J., Dal Maso, M., Lihavainen, H., Viisanen, Y., Aalto, P. P., Komppula, M., and Kulmala, M.: High natural aerosol loading over boreal forests, Science, 312, 261-263, 2006.

Turekian, V. C., Macko, S. A., and Keene, W. C.: Concentrations, isotopic compositions, and sources of size-resolved, particulate organic carbon and oxalate in near-surface marine air at Bermuda during spring, J. Geophys. Res., 108(D5), 4157, doi:10.1029/2002JD002053, 2003.

Widory, D.: Combustibles, fuels and their combustion products: A view through carbon isotopes, Combust. Theory Modell., 10, 831–841, 2006.