Interactive comment on “Seasonal variations of stable carbon isotopic ratios and biogenic tracer compounds of water-soluble organic aerosols in a deciduous forest” by Y. Miyazaki et al.

Anonymous Referee #3

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The paper describes the use of stable carbon isotope analysis to determine the sources of water soluble organic aerosols (WSOC) in a forest canopy. A seasonal study of WSOC concentration and δ^{13}C_{WSOC} covering a period of 18 months is presented for the first time. Concentration and isotopic ratio measurements are related to the net carbon dioxide exchange between the studied ecosystem and atmosphere, showing in the vegetation growing period highest CO_2 uptake together with high input of precursor molecules from C3 plants to the measured WSOC. A positive matrix factorization analysis was carried out using tracer compounds for primary emitted and secondary formed organic aerosols, as well as inorganic tracers, resulting in five factors. Investigating the seasonal relative contribution of each factor to WSOC, the authors concluded that veg-
etation growing season is important for primary and secondary formation of WSOC. Further, in midsummer biogenic secondary organic aerosols formation is most important WSOC source. Finally, vertical concentration gradients of WSOC and tracers show that the forest floor is a significant source of WSOC.

The use of stable isotopes analyses of ambient samples to determine sources and extent of the atmospheric processing of aerosols is an innovative method. The paper is suitable for publication in the journal after considering following points:

General:

Page 30784 (lines 20-21): The phrase ‘similar to a typical range of marine sources... (Miyazaki et al., 2011)’ should be more clearly expressed. Does this value -20 to -22 per mille represent the $\delta^{13}C_{WSOC}$ or the isotopic ratio of all water soluble compounds, including inorganics (in the chemical analysis part of Miyazaki et al. (2011) no acidification of the samples is described). In the latter case, the comparison needs some more discussion.

Page 30784 (lines 26-27): Sentence ‘However, reported ... (Rudolph et al., 2003)’ should be rephrased. The ‘processes’ should be separately commented (formation and loss processes of WSOC, partitioning of WSOC to aerosol particles, see Fisseha et al. 2009). Is the reference Rudolph et al. 2003 the best one for this discussion on $\delta^{13}C_{WSOC}$, at all?

Figure 5: The authors should consider renouncing to the lines connecting the measurements in 2 and 15m height. This gives the impression that the profiles are linear (which is not necessary the case).

The importance of MSA measurements should be shortly mentioned in the introduction.

Page 30778 (line 4): ‘larger’ should be replaced by e.g. ‘existing’. The N to W fractions are very small.
Technical:

Page 30776 (line 4): Use 'contain' instead of 'constitute'.
Page 30778 (line 15): NEE = net ecosystem exchange.
Page 30784 (line 18): Instead of 'In winter, a factor in which...' use 'In winter, factor5, in which...'
Generally, 'forest floor' should be used instead of 'canopy floor'.

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