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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Measurement report: Source characteristics of water-soluble organic carbon in $PM_{2.5}$ at two sites in Japan, as assessed by long-term observation and stable carbon isotope ratio" *by* Nana Suto and Hiroto Kawashima

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The manuscript entitled "Measurement report: Source characteristics of water-soluble organic carbon in PM2.5 at two sites in Japan, as assessed by long-term observation and stable carbon isotope ratio" by N. Suto and H. Kawashima represents investigation of aerosol particles through the prism of carbon stable isotopes, EC, OC and ion analysis. The sample collection was performed during two years, which allowed for the authors to catch the seasonal changes in some aerosol parameters.

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Discussion paper

In general, the manuscript is a bit chaotic, sometimes pointing at not relevant issues. Also, I think Authors have not exploited the benefits of long time samples collection and analysis.

I would like to address the issues that I am concerned on:

Lines 25-30 In the introduction part, the most recent reference is from the year 2011.

Line 35. Why authors mention about PMF? They are not using PMF in this manuscript? What it means “it is necessary to identify the characterisation of source artificially”?

Line 45. In general, the isotope ratio literature review part is too narrow in this manuscript. I propose to make overview on most recent references, which deals with aerosols and stable isotopes in different areas, which are relevant for this manuscript: transportation, natural terrestrial and marine (also oceanic) sources, fires etc. Suggestions: Zhang W. et al., 2019, ACP 19, 11071-11087 (δ13C of WSOC in China); Fisseha R. et al 2006, Atmos. Env. 43 431-437 (δ13C values in the different aerosol fractions in Zurich, Switzerland); Kirillova E.N. et al. 2010 Anal. Chem. 82 7973-7978; Lang S.Q. et al. 2012 Rapid Commun Mass Sp. 26 9-16; Suto N. and Kawashima H. Rapid Commun Mass Sp. 32 1668-1674 (In this manuscript the same Authors present the own developed method for δ13C in WSOC). The last citation is used later, but I propose to cite it here also, as it is relevant to this part.

Section 2.1. I see that present study two sampling points were separated by a distance of 370 km . Both Tsukuba and Yurihonjo has a Humid continental climate, with different annual mean temperature of about 2°C. Tsukuba is close to Ocean, while Yurihonjo is close to Sea of Japan. I assume that air mass back trajectories are important when comparing two sampling points in this case. I assume that can be problematic to compare suburban or rural places, as they can be influenced by totally different atmosphere (and aerosol sources) conditions. In the rest of the text I’m missing marine source evaluation (except nss-K+).

Line 95. I did not understand how long sampling for one filter was performed, does all filters were loaded periodically? Maybe worth to add as supplementary filter data (time, weight of loaded aerosol). How Authors ensured that the filter not lost mass itself, as quartz filters break easily, and making gravimetric measurements can be complicated. Does Authors used equilibration for the unexposed and exposed filter to account for humidity?

Section 2.2 can be improved. I miss a clear step-by-step description of analytical procedures: Line 105: "...the carbon was converted to CO2 via an oxidation catalyst in the reduction tube of EA" - I think is incorrect and must be clarified. Does oxygen was added? Does nitrogen was seperated from CO2? What about blank measurements, does it gave any signal? In this section, is no clear separation, which part of the section corresponds to total carbon measurements, which part to WSOC?

Line 170. Authors state that snow is responsible for low PM2.5 concentration in Yurihonjo. My question, does land in winter emits significant amount of aerosol, even not covered by snow? What about heating activities, long range transport etc. I want to say that Authors must to think generally what they want to deliver to the reader, and maybe skip parts which are not relevant to the main purpose of the article (for example about most polluted megacities, as here megacities are not investigated). Yurihonjo can't be compared to a megacity, as by definition it is an urban site (last line in 165).

Line 260-265. The assumption that rice residues burning is the main source of WSOC in Tsukuma is nice assumption, but I was not convinced by this statement. Usually, in urban sites weak seasonal trend in carbon delta values is due to transportation influence. Here, Authors did not evaluate the δ13C of fossil fuel emissions. In addition, assuming that crop combustion is the main source of WSOC, we must to see correlation with delta values. No correlation between WSOC concentrations and delta values means that does not exist one dominating aerosol source. In other words, if in Tsukuma main source would be crop burning, we must observe correlation between WSOC and delta values. Or, delta values are affected by atmospheric processing, but

then air mass back trajectory analysis (even with Solar intensity) must be performed to understand the resident time of the aerosol.

Line 290. Speculations about long-range transport must be verified by air mass trajectory analysis. Also, Authors can use other delta signatures (various crops, land and forest fires, transportation and fuel burning emissions, shipping emissions etc.), not only rise burning.

Line 310. Kawashima and Murakami 2014 report that in roadside samples VOC ranged from -29.6 to 23.5 permill (the same range for ambient samples). It makes difficulty interpreting vehicular emissions from a stable isotopes perspective. I'm not sure if only Toluene and Xylene are responsible for WSOC in Yurihonjo, so 5.8 permill subtraction must be made carefully (Authors note themselves that anthropogenic emissions of VOC are smaller than that biogenic).

Figure 2. I not found in the manuscript explanation, what causes enriched delta values in the winter in season 2018/2019 compared to 2017/2018 at both locations?

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