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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Radiocarbon analysis of elemental and organic carbon in Switzerland during winter-smog episodes from 2008 to 2012 – Part 1: Source apportionment and spatial variability" *by* P. Zotter et al.

P. Zotter et al.

Peter.Zotter@psi.ch

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We thank the reviewer for the comments on our paper. To guide the review process we have copied the reviewers' comments in italics. We have responded to all the referee comments and done the modifications accordingly (**in bold in the text**).

This is a very interesting manuscript, with the aim of identifying the sources of carbonaceous aerosol during wintertime smog episodes in Switzerland. The main result is that wood burning consistently makes a major contribution to wintertime smog in

Switzerland, over a large region and long time period, even in major urban centers. There was previous evidence for this, but only for campaigns of limited regional and temporal scope, where it was difficult to prove that this is indeed a large-scale problem. The study is very thoroughly conducted, with careful corrections of artifacts and a good error analysis. The results are clearly presented in the figures and generally support the conclusions very well. Only the descriptions of the results is sometimes a bit cumbersome, and it is easy to lose track what the many numbers in the text are referring to. I therefore recommend acceptance with a few minor revisions, listed below.

We thank the reviewer for the positive comments. In the following we will respond to each comment separately.

R2.1. Page 15608, line 9 -20: All the individual p values make this very tedious to read. It is sufficient to state at the beginning or the end of the paragraph that all the differences discussed here are significant at the 95% confidence level.

We added the following in lines 433-434 in the revised manuscript

"...(t-test significant at 95%**, in general throughout the manuscript we always used a t-test with p = 0.05 to test the statistical significance of differences between stations north and south of the Alps**)."

and therefore deleted all expressions "(t-test significant at 95%, p = xxx)" occurring later in the text (see line 434, 437, 440-441, 483, 524 in the revised manuscript)

R2.2. Page 15608, line 16 – 23: This sentence is too long.

We split this sentence now into 2 and since "(t test, significant at 95%, p = $9.2\times10-4$ for NO3-, p = $2.0\times10-20$ for SO42- and p = $1.0\times10-7$ for NH4+)" was deleted it is also better readable now (see lines 439-441 in the revised manuscript)

R2.3. Page 15610, line 10ff: Four stations were measured only for one winter, consequently only 5 data points are used for the whisker plot, including 10th and 90th percentile. This is statistically not very meaningful. I suggest to simply use a mean and

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standard deviation for these 4 stations in Figures 2 and 3. This also has the advantage that the stations with only few data points are immediately recognizable in the figures.

We follow the suggestion of the reviewer and show only the mean and standard deviation for the 4 stations where only filters from 1 winter were analyzed (see new Figure 2 and Figure 3 in the revised manuscript and new Figure S3 and Figure S4 in the revised supporting information). We also added the following to the corresponding figure captions:

"...**Only averages ± standard deviations are displayed for stations from which only filters from one winter were analyzed**..."

R2.4 Page 15613, line 19-21 and line 23-24: This is an example of the writing style that made reading of the results section unnecessarily difficult. Four numbers are given in succession and only at the end of the sentence it is clear which ones refer to OC and EC. It would have been much more clean to write ". . . 58-71% and 1.5 – 5.2 ugCm-3 for OCNF and . . . for ECNF". This is just a small difference in sentence construction but makes a big difference for the reader, e.g. I had to reread the original sentence at least once to get the numbers straight in my head. Please throughout the manuscript, use the construction with "respectively" as sparingly as possible. (Only of it makes the sentence significantly shorter, or if there are not too many numbers in the text.) This will make the results section much more easy to read.

According to the suggestion of the reviewer we adapted the text and removed "respectively" many times in the result section.

See lines 438-439, 501-502, 510-515, 528, 538-541, 567-569, 576-577, 579-580, 590, 645-646 and 670-671 in the revised manuscript.

R2.5. Page 15613, line 25ff: You are packing three separate conclusions (sources, SOC formation, and regional air pollution) in one sentence. Better to use one sentence per conclusion. In my opinion the point about SOC formation is not self evident, since

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SOC formation and possible influence on OCNF was not discussed before. Please elaborate this point a bit more.

The fact that OCNF can also be partly SOC formed from non-fossil VOCs was discussed several times before this section (see below).

Page 15594 lines 25-28 and Page 15595 line 1: "Meanwhile, OC may be either primary OC (POC) directly emitted in the atmosphere or secondary OC (SOC) formed in the atmosphere through the oxidation of volatile organic compounds (VOCs) from both fossil (coal combustion, industrial and vehicle emissions) and non-fossil (e.g. biomass burning and biogenic emissions as well as cooking) sources. . ."

Page 15595 lines 7-11: "The most detailed information about different sources can be achieved when 14C measurements are performed on OC and EC separately, since EC originates exclusively from biomass burning and fossil fuel combustion. By contrast, the apportionment of OC into these two sources using this methodology is less straightforward due to the complex primary and secondary sources of this fraction."

Page 15610 lines 10-12: "As discussed above, non-fossil OC may include, POC and SOC from biomass burning and cooking emissions, as well as primary biological particles and biogenic SOC."

However, we agree with the reviewer that this sentence is quite long. In addition, we agree that we didn't point out that SOC formation can be highly variable. Therefore, we changed lines 25 to 29 on page 15613 to (see lines 580-588 in the revised manuscript):

"Together with the low station-to-station differences, this suggests that non-fossil sources very consistently influence stations on the Swiss Plateau. **Furthermore, as discussed above, OCNF can be influenced by SOC formation which can be highly variable. However, the low OCNF station-to-station and day-to-day variability points to a similar** degree of atmospheric processing and SOC formation for the chosen days **in this region of Switzerland. Last, the low absolute and relative ECNF**

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and OCNF IQRs at the individual stations and station-to-station differences also indicate that locations on the Swiss Plateau are rather influenced by regional (still mainly within Switzerland) air pollution."

R2.6. Page 15614, line 9: "contributions of OC, or "concentrations of OC"?

We changed the text (see lines 597-598 in the revised manuscript) from "...absolute...contributions..." to "...**concentrations**..."

R2.7. Page 15618, line 21ff: see comment 4, this is really not clear

We added now the different biomass burning ratios from literature to Table 3 and in the text in section 3.3.3 we therefore removed most of the numbers (and therefore also most of the expressions "respectively") and just refer to Table 3 which makes this section now more readable.

R2.8. Page 15619 line 10ff: If OCbb/ECNF ratios are similar North and South of the Alps is this not somewhat contradictory to the conclusion that more efficient burners are used in the North? Is there evidence that more efficient burners have a similar OC/EC ratio as less efficient burners?

We agree with the reviewer that OC/EC wood burning emission ratios are a function of burning conditions. However, in new and more efficient log wood and pellet burners both, the OC and EC emissions are very low compared to old burners (see e.g. Heringa et al., 2011). From our data we cannot completely rule out different OC/EC emission ratios in both regions. The ambient OCNF/ECNF ratios in the north and the south could be similar as the possible higher OCNF concentrations in the south from primary biomass burning emissions might be compensated by a larger non-fossil SOC fraction in the north. Therefore, we added the following in the revised manuscript (see lines 665-667 in the revised manuscript)

"**However, with our data we cannot completely rule out different wood burning OC/EC emission ratios in both regions of Switzerland as higher primary wood**

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burning OC emissions in the south could be compensated by a larger non-fossil SOC fraction in the north."

References

Heringa, M. F., DeCarlo, P. F., Chirico, R., Tritscher, T., Dommen, J., Weingartner, E., Richter, R., Wehrle, G., Prevot, A. S. H., and Baltensperger, U.: Investigations of primary and secondary particulate matter of different wood combustion appliances with a high-resolution time-of-flight aerosol mass spectrometer, Atmos. Chem. Phys., 11, 5945-5957, doi: 10.5194/acp-11-5945-2011, 2011.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 15591, 2014.