

General comments:

We thank the reviewer for the careful reading of the manuscript and the valuable remarks and comments. We have done our best to implement all the comments of the reviewer.

We changed the acronym for continental air mass (see below), and explain more clearly the distinction between OCc and OCc,o. However, if possible, we would like to keep the current acronyms. The acronym F¹⁴C was strongly recommended by Reimer et al., (2004) to denote fraction modern and we follow this convention. The most similar acronyms are OCc (all contemporary OC) and OCc,o (other contemporary OC, which denotes all contemporary OC, except OC from primary biomass burning; this is often denoted OCbio, but as one reviewer pointed out in the preliminary review, OCbio is misleading for this fraction). We find it difficult to come up with a very distinct, alternative acronym for this carbon fraction.

Please also not that in response to on comment by reviewer 2, we slightly changed the way quantify EC in the three spring samples presumably affected by pollen events. This changes some numbers in the tables and figures slightly, but does not change any of the discussion or conclusions.

For easier tracking of the changes, we marked all the changes in response to reviewer 1 in yellow and the changes in response to reviewer in grey throughout the revised manuscript.

RC: I lack an analysis regarding the “correctness” of the forecasted HYSPLIT trajectories. How did you assure that these forecasted trajectories were correct? Did you compare the forecasts to the actual trajectories (that “took place”)?

Answer: We will make this more clear in the text: The sampling times were decided based on forecast trajectories, which are not always correct. We then used the actual back-trajectories (based on re-analysis data) to select some of the filters with the most consistent back trajectories for analysis. This is now explained in the paper (page: 13, lines: 22 - 24)

RC: I also lack a clear classification regarding the seasons. Did you classify them by calendar months, days, etc.? Or did you classify them by meteorological means, i.e. temperature? This is of crucial importance when interpreting the results.

Answer: We thank the reviewer for this useful comment. We classify the seasons by month: winter: Dec., Jan., Feb.; spring: March, April, May; summer: June, July, August; fall: Sept., Oct., Nov. This is now stated explicitly in the paper (page: 14, lines: 7-8).

Page 2, line 12. Please replace “it” with what you actually mean, i.e. carbonaceous material.

Done (page: 2, lines: 13)

Page 2, line 10-16. It would be nice if the authors could mention the fraction carbonaceous aerosol in PM₁₀ or PM_{2.5} in Europe. To give the reader an idea of how large this fraction is.

We added the sentence: In Europe this fraction is typically between 30 and 60% of PM_{2.5}

(page: 2, lines: 11)

Page 2, line 32. Can the authors please explain why the ratios are normalized to a $\delta^{13}\text{C}$ value of -25‰.

This is to account for isotopic fractionation during sample pretreatment and measurement. This explanation is now added to the text (page: 2, lines: 33-34)

Page 3, line 13-19. You very nicely explain that the three major sources of carbonaceous aerosol are biogenic, fossil fuel and biomass burning derived. However, you only show references of biomass burning in the later section of the paragraph. I would like to see some references on studies that showed that fossil fuel aerosol mass is rather stable throughout the year, further that the biogenic carbonaceous aerosol is totally dominating in rural areas during summer (Genberg et al. 2011; Yttri et al. 2011)

We added the sentence about biogenic aerosol to the text. (page: 3, lines: 21-22). There is not a very clear reference that shows that fossil fuel aerosol is rather constant throughout the year, because not many long-term studies exists.

Page 3, line 35-37. This information should be given earlier in the introduction if this number is for Europe. If the number is relevant for the Netherlands, please ignore this comment.

This number is relevant for the Netherlands and this is now clarified in the text (page: 4, lines: 1)

Page 4, line 3-5. Perhaps omit this sentence. Also, I find this value rather low, I am not that surprised given the surrounding environment as you mention.

This sentence is now omitted

Page 4, line 15-21. Please state the altitude of the measurement station.

0.7 m below sea level, this is now stated in the text (page: 4, lines: 17)

Page 4, line 15-21. Please state how you differentiated between the different seasons.

We classify the seasons by month: winter: Dec., Jan., Feb.; spring: March, April, May; summer: June, July, August; fall: Sept., Oct., Nov. This is now stated explicitly in the paper (page: 14, lines: 16-17).

Page 4, line 23-28. Please state the flow of the high-volume sampler.

500 l/min, this is now stated in the text (page: 4, lines: 26).

Page 4, line 26-28. How did you assure that the HYSPLIT forecasts were correct, and did you estimate the correctness of the forecasts? For me it is not unlikely that there were cases when the forecast said one thing, but the air masses did in fact arrive from another direction than was forecasted.

The sampling times were decided based on forecast trajectories, but after sampling we calculated actual back trajectories. Only a subset of samples, where the actual back trajectories were satisfactory was selected for analysis. Figure 3 shows the actual back-trajectories (based on re-analysis data) of the filters that were selected for analysis. This is now explained in the paper (page: 13, lines: 21-23)

Page 5, line 4-30. Please clarify for all combustion steps the atmosphere used. Was it pure O_2 in all cases? Also, you did not measure carbon mass in these combustion steps, is that correct?

On page 5, line 9 of the original manuscript it is stated that carbon fractions are

combusted in O₂. In the revised manuscript we added this now for each carbon fraction. (page: 5, lines: 15-20) The mass of carbon (OC; 360°C step and EC; 650 °C) in each sample is determined manometrically. This is added to the text (page: 5, lines: 9-10)

Page 5, line 18-20. How do you differentiate between the EC (in the OC-EC mixture) that is combusted in 450°C and EC combusted in 650°C? Can you estimate the amount of EC evolved in the 450°C step?

With the method as it is set up now, the CO₂ evolved in the 450°C step is discarded and we do not measure the amount of carbon combusted in this step directly. However, because we also measure the amount and F¹⁴C of TC, we can estimate the amount of carbon evolved in this step as the difference between TC – OC(recovered at 360°C) – EC (recovered at 650°C). We can also make a crude estimate of what percentage of EC is recovered by comparing ECr to EC determined by the thermal optical method, or by the calculations outlined in section 2.8. The EC evolved at 450 is consequently EC_{total} – ECr.

Page 5, line 21-23. I don't understand how you derive the mean charring bias of 0.04? Please explain.

This is a very rough calculation, assuming that 5% of recovered EC consists of charred OC and that F¹⁴C of OC is approximately 0.8. Since we cannot measure the charring directly for every sample, such a simple average correction for all the samples is the best we can do at the moment. We now add the explanation of how we arrive at 0.04 (page: 5, lines: 24-25)

Page 5, line 31-36. Did you measure carbon mass on both of these facilities?

The carbon mass was determined at the University of Utrecht on the ACS system, now explained in the manuscript (page: 5, lines: 9-10)

Page 6, line 7. Is it really 500 mg? That's a huge mass. Further, if you mean µg, I still question the number 500, perhaps you mean 50 µg?

Yes, thanks for spotting the error. Unfortunately all the micron symbols were lost, when we applied the ACP template to the manuscript, and even though we did our best to correct this, it seems we overlooked a few instances. Corrected (page: 6, lines: 10)

Page 6, line 17. What do you mean by "Unknown samples"?

"Unknown" was replaced by "The" (page: 6, lines: 20)

Page 6, line 23. Again, I question that you had 10-100 mg/cm² OC on your sample filters. Corrected (page: 6, lines: 26)

Page 7, line 7-15. In this paragraph I lack a motivation to why you should measure sugars in the first place. What types of sugars were your target compounds? I also lack some information that it is the levoglucosan that is of main interest here. Perhaps you can address this in the introduction or here in the method section? Also, what was the measurement uncertainty of the analysis?

We now give a brief motivation at the start of this paragraph: "In addition to F¹⁴C we also measured atmospherically relevant sugars, (e.g., levoglucosan, sucrose, glucose, mannosan). Levoglucosan can serve as an independent tracer of biomass burning and several other sugars can indicate primary biological material, which cannot be traced by ¹⁴C measurements alone." (page: 7, lines: 12-15)

The relative standard deviation of the measurements, determined based on replicate analysis of standards and blanks, was below 10%. (page 7; lines, 24-25)

Page 7, line 17-25. In this paragraph I lack information regarding the He-O₂ mixture, which proportions were used? Further, why did you use the QUARTZ protocol? What are the benefits by using this protocol instead of EUSAAR-2? What was the measurement uncertainty of the analysis?

The mixture contained 10% oxygen (page: 7, line: 31), the analytical uncertainty for OC and EC varied slightly with filter loading from 5% at loadings above 20 ug/cm² to 7% for loadings around 10 ug/cm² (page: 7, line: 34-36).

The Quartz Protocol is used routinely at the University of Vienna, where OC-EC measurements have been ongoing since before the EUSAAR protocol was introduced. Therefore the quartz protocol is still used to ensure comparability with previous measurements and older records. There was no specific scientific advantage for using it in this study.

Page 7, line 17-25. You should here state that you used TOA for comparison to ACS and perhaps the radiocarbon facilities to estimate carbon mass. After reading the whole method section I believed that you estimated the carbon mass by TOA, solely. However, when reaching the result section, I found out that TOA was just a measure of comparison to ACS, is that correct? Either way, the carbon mass measurements need to be clarified. We clarified this by changing the title of section 2.7. to “Estimation of OC and EC by thermal-optical analysis” and of section 2.8 to “Estimation of OC, EC, WIOC concentrations based on carbon fractions recovered by the ACS system” to make clear that we estimate OC and EC in two different ways.

To section 2.7. we added the sentence “EC, OC and TC measured by the thermal optical method are used to evaluate the estimated values of OC and EC based on the data from the ACS system (see section 2.8).” (page: 8, lines 1-3)

Page 7, line 28-29. This sentence should be presented earlier in the ACS method part. In section 2.3 (page 5, line 18 of the original manuscript) we write: “Then the oven temperature is raised to 450 °C for two minutes and in this step a mixture of the most refractory OC and EC is burned off the filter.” We opt to keep this sentence and remove the corresponding sentence in section 2.8 in order not to mention this twice.

Page 9, line 18. You have written EC_{co}, but do you mean EC_f?

Yes, thank you for spotting this typo, we have corrected this (page: 10, line: 2)

Page 9, line 27. You have written EC_{co}, but do you mean EC_f?

Also corrected (page: 10, line: 11)

Page 12, line 9. Levoglucosan should be mentioned earlier, in the introduction or in the proximity of the sugar measurements written in the method section.
done (page: 7, lines 12-14)

Page 12, line 15-16. Here you mention glucose and sucrose. This should be mentioned earlier, in the introduction or in the proximity of the sugar measurements written in the method section.
done (page: 7, lines 12 - 15)

Page 12, line 21-24. Please give a motivation why you chose to replace these values with values obtained from the regression line.

We added the following explanation: This can be seen as a crude correction for the highly refractory part of OC that was apparently incorrectly classified as EC from biomass burning. Without this correction we would overestimate the contribution of biomass burning to the carbonaceous aerosol in spring. (page: 13, lines 4-6)

Page 12, line 30-32. This information should also be mentioned in the method section. This information is present in section 2.3 (page: 5, lines 20-23 in the original manuscript). Therefore we changed the discussion in section 3.1, by referring to section 2.3, and just pointing out that the data shown in Figure 2 are not yet bias corrected. (page: 12, lines 29-30)

Page 12, line 40. In the figure caption of Figure 3 it says 48 h.
Thank you for spotting this mistake, it should say 96 hours in the figure caption.

Page 13, line 10-14. In Figure 3, the blue lines were included into the red lines (modified marine)? Perhaps write this information in the figure caption.
Done

Page 13, line 15-25. It is a bit confusing that you use “co” as an acronym for both “continental” and “contemporary, other”. Consider changing this, it will most likely increase the readability of the paper.
Yes, we changed co to cont for continental conditions in the tables.

Page 13, line 26-28. I think you should add the coverage in days to Table 2.
This is a good suggestion, we added this information.

Page 14, line 32. Here you mention the seasonal pattern of OC_{bb} concentration which is a bit confusing since this parameter is not presented in Table 3.
Thank you for pointing this out. We deleted OC_{bb} here.
Page 15, line 23-24. Here it would be suitable with a reference.
We added a reference (Ohata et al., 2016) (page: 16, lines 4)

Page 16, line 16-28. Again, here it would be nice to know how you classified the seasons. The difference between spring and fall should be small since you can expect these seasons be the intermediate of two extremes (i.e. winter and summer). However, this might not be the case depending on how you classified and defined your seasons. For increasing the interpretation and readability you should mention seasonal classification. We added this information (page: 14, lines 7-8)

Page 16, line 39-40. How do you heat your residents in the Netherlands during winter? Is it non-aerosol producing energy source? Perhaps you can mention this somewhere.
Heating in the Netherlands is done to a large part with gas, which produces very little aerosol. We added a brief discussion of the main sources of EC_f in section 3.2 (page: 15, lines 3-5)

Page 17, line 26. I assume you mean $\mu\text{g}/\text{m}^3$ and not mg/m^3 ?
Thank you. corrected (page: 18, lines 13)

Page 29, Table 1. It would be nice if you could add the references for these numbers in the table.
Here we would prefer to keep the references in text, because not all the references are treated equally, e.g. we cite Szidat and references therein as a basis of our estimate and

adjust the values in this publication, considering several newer publications. Therefore, it would be somewhat misleading to just give the references in the table without all the explanations added in the text.

Page 31, Table 3. In the text you called “contemporary, other” “c,o”, here you just call it “c”. I would like to see consistency between the acronyms in the text and in the table.

As defined in section 2.9 (page 10, line 32 ff), the symbol c stands for all contemporary carbon, i.e., the sum of biomass burning and contemporary other carbon. $OC_c = OC_{bb} + OC_{c,o}$. We will add this to the figure legend to avoid misunderstandings.

Page 31, Table 3. Why did you merge OC_{bb} and $OC_{c,o}$?

Because as can be seen in figure 3, the separation between OC_{bb} and $OC_{c,o}$ is actually more semi-quantitative, and in the table we prefer to give concentrations of carbon fractions that we can determine with greater certainty. This is now mentioned in the text. (page: 14, lines 38-39)

Page 32, Figure 1. I lack an explanation of the y-axis in Figure 1b.

We added a better explanation in the caption of figure 1. We also added the y-axis label “ratio” to indicate that the ratios of ACS EC and Sunset EC are plotted.

Page 33, Figure 2. Is the equation valid only for the blue data points? Please clarify this in the figure caption.

Done

Technical corrections

all the technical corrections have been corrected

Page 12, line 20. Please replace “Weather” with “Whether”.

done (page: 12, line: 39)

Page 15, line 3. Please add “is”. “Therefore we think it is unlikely.....”

done (page: 16, line: 23)

Whole document. Check for discrepancies between “c,o” and “c” acronyms. Including figures and tables.

We checked very carefully and could not find any discrepancies (to our knowledge). Everywhere c refers to all contemporary carbon (i.e. sum of biomass burning and biogenic carbon) and c,o to “other contemporary carbon (i.e. all contemporary OC, except primary OC_{bb}).

References

Genberg et al. 2011. Source apportionment of carbonaceous aerosol in southern Sweden. ACP.

Yttri et al. 2011. Source apportionment of the summer time carbonace