

Interactive comment on "Sources and formation mechanisms of carbonaceous aerosol at a regional background site in the Netherlands: Insights from a year-long radiocarbon study" by U. Dusek et al.

### **General comments**

The authors present a comprehensive study regarding the sources of carbonaceous aerosol fractions in a regional background station in the Netherlands. Large focus is devoted to seasonal patterns in concentration and influence from the regional proximity in contrast to continental Europe.

I regard the scientific value of this work as moderate with the motivation that the used techniques and methods are not very novel. However, the authors prove the applicability of the method and techniques in a satisfactory manner. The presented results are clear and interesting. However, perhaps it is not that surprising that air masses from oceanic areas are cleaner than continental ones. In my understanding, there are limited number of studies that have measured  $\delta^{14}\text{C}$  in the presented carbon fractions, this study adds potentially significant information to this field.

The study is limited to the Netherlands and may be of national interest, however my impression is that there has been very few source apportionment studies of the carbonaceous aerosol conducted in this area. Hence, this study is of potential large interest for aerosol research in the Benelux area.

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")?

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.

Some effort can be spent on reviewing the acronyms used in this paper, there are many and some are very similar. A review and some corrections would increase the readability of the paper.

After some major corrections I believe that this manuscript should be published in ACP.

### **Specific comments**

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

Page 2, line 10-16. It would be nice if the authors could mention the fraction carbonaceous aerosol in PM<sub>10</sub> or PM<sub>2.5</sub> in Europe. To give the reader an idea of how large this fraction is.

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

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)

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.

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.

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

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

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

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.

Page 5, line 4-30. Please clarify for all combustion steps the atmosphere used. Was it pure O<sub>2</sub> in all cases? Also, you did not measure carbon mass in these combustion steps, is that correct?

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?

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

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

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?

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

Page 6, line 23. Again, I question that you had 10-100 **mg**/cm<sup>2</sup> OC on your sample filters.

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?

Page 7, line 17-25. In this paragraph I lack information regarding the He-O<sub>2</sub> 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?

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 needs to be clarified.

Page 7, line 28-29. This sentence should be presented earlier in the ACS method part.

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

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

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.

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.

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

Page 12, line 30-32. This information should also be mentioned in the method section.

Page 12, line 40. In the figure caption of Figure 3 it says 48 h.

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.

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.

Page 13, line 26-28. I think you should add the coverage in days to Table 2.

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.

Page 15, line 23-24. Here it would be suitable with a reference.

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.

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.

Page 17, line 26. I assume you mean  $\mu g/m^3$  and not  $mg/m^3$ ?

Page 29, Table 1. It would be nice if you could add the references for these numbers in the table.

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.

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

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

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

### **Technical corrections**

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

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

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

### **References**

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

Yttri et al. 2011. Source apportionment of the summer time carbonaceous aerosol at Nordic background sites. ACP.