Anonymous Referee #3

We thank the reviewer for their support that our work is sound and deserving of publication in ACP after response to their comments. We respond to each comment individually below. The reviewer's comments are in italics and blue font, our responses are in normal text.

The manuscript by Ots et al. presents a modeling study that explores the uncertainty of the residential and non-industrial combustion emissions sector over the UK and Ireland. The uncertainty estimates, translated into sensitivity experiments in the manuscript, are driven from past studies (Ots et al., 2016) and comparisons with measurements. The model domain covers the whole UK and Ireland, although the analysis is heavily based on comparisons with data in London.
The work presented is a standard modeling approach, where an emissions sector is perturbed and the different model versions are compared against measurements, to evaluate which of the scenarios under study is performing best against some metric, which in this case it is a fraction of organic aerosols (solid fuel OA; SFOA) and black/elemental carbon. The analysis does not have any mistakes, although one can argue that the approach of non-volatility and ageing for SFOA deserves improvement, especially given the temperature-dependent parameterization presented for the emissions during cold days. The results are not surprising either; a low bias in SFOA is improved by increasing its emissions, and rather linearly, as seen in Table 3. For both sites studied, the NMB presented in Table 3 for the combRedist experiment is roughly equal to the arithmetic mean of the Base and Redist simulations, which is what one would expect from the experimental design. In addition, no attempts have been made to link this work with either the study in Belgium mentioned in the manuscript, or with other relevant areas, limiting the scope of the work as presented. The last sentence of the conclusions also supports my concern about the limited scope of the study, again, as presented in the manuscript. Regardless, I believe that the work is sound and de- serves publication in ACP after addressing my comments below.
Specific comments
1. *p. 3, l. 25: The FINN inventory has natural fires only, or all open burning? Some of the open biomass burning is anthropogenic (e.g. deforestation, agricultural fires).*

Yes, thank you. We have now changed "natural fires" to "open burning (including wild fires and agricultural burning)".

2. *Section 2.2: The way I understand the degree-day factors equation is that it does not affect days with temperature higher than 18 C, but it increases emissions for colder days. If this is correct, isn't it going to increase the annual totals? In addition, why not apply the same approach hourly (section 3.2) and get a more natural diurnal variability, instead of the imposed one?*

Yes, the reviewer's understanding is correct, it does not increase the annual total. Regarding the second question (why not apply this on hourly temperatures):
Firstly, the work we present in this manuscript did not create and make methodological changes to how the degree-day factors, we used them as described in Simpson et al. (2012): The EMEP MSC-W chemical transport model - technical description, *Atmos. Chem. Phys.*, 12, 7825–7865, doi:10.5194/acp-12-7825-2012.

Secondly, while we agree that the hourly profiles used in this study (also based on the above reference) need review (in our abstract we write: "The model results also suggest the assumed temporal profiles for residential emissions may require review to place greater emphasis on evening (including 'discretionary') solid-fuel burning.") we do not think hourly variation in temperature has a very direct effect on increased heating. This may be true for thermostat-based central heating systems (gas or electricity `fuelled`), but solid fuel burning means that people need to be home and awake (i.e. even if the temperature drops overnight, people will not wake up and start burning wood and coal). Therefore, the hourly profile of SFOA emissions is unlikely to be driven by changes in temperature on an hourly basis.

3. *p. 5, l. 7: Please start a new paragraph with "The experiments Base, . . ."*

Agreed and done.

4. *Section 3.1: How far apart are the two stations? Are they in adjacent gridboxes, the same one, really far away? How about differences in local influences, if any?*

The two stations are about 4.5 km apart, in separate adjacent model grid cells. Detailed maps of these sites (overlaid with residential and workday population density at 1 km resolution) can be found in Fig. 1 in Ots et al. (2016) "Model simulations of cooking organic aerosol (COA) over the UK using estimates of emissions based on measurements at two sites in London", Atmos. Chem. Phys., 16, 13773-13789, reproduced below:
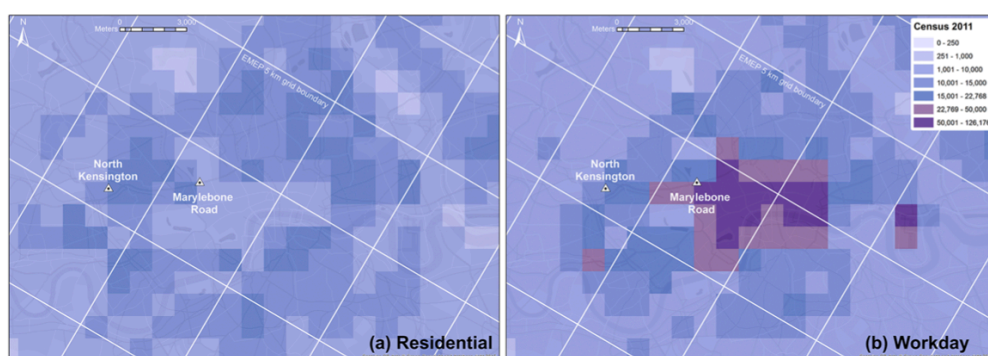

Figure 1. Residential **(a)** and workday **(b)** population density in central London at 1 km × 1 km resolution. The residential population maps are based on Reis et al. (2016). While the same methodology is applied to derive workday population maps, they are not yet published due to delays in the provision of workday population census data for Scotland. Also shown are the measurement sites and the EMEP4UK 5 km × 5 km grid used in this study (white lines). Underlying map contains Ordinance Survey (OS) data © Crown Copyright 2015.

We have now included the following in Section 3.1 (paragraph 2) of this manuscript:
"These two sites are ~4.5 km apart, in adjacent model grid cells, and represent different kinds of urban areas. North Kensington is comparatively residential whereas Marylebone Road is near central London and therefore exhibits very high numbers of people during the workday. More discussion on this, including detailed maps can be found in Ots et al. (2016b)."

5. *p. 11, l. 16-17: Why only correlation and not the other metrics? More generally, this is an important piece of information and should be expanded, even though it is already published.*

Agreed, we have added the following sentences:

"For example, Ots et al. 2016a presented scatter plots of daily averaged concentrations of the different OA components derived from measurements with the two different AMS instruments at the North Kensington site during the winter IOP (the cToF-AMS versus the HR-ToF-AMS). While these comparisons showed good correlations between the two measurements (0.88 to 0.95 for the primary OA components, 0.77 for secondary OA), on some days the absolute measured concentrations of specific components differ, sometimes by more than a factor of two."

6. *Figure 7: Some error bars or other means that present temporal variability can be very informative here.*

Overall, we agree with this comment that it is useful to try to present the variability, and not just the final aggregated (in this case the mean) values. However, since we plot 4 individual lines on each panel here, we think that adding for example the standard deviation around each line would make the information on the panels overlap each other and the plot would become `too busy`. A solution could be to separate pairs of these lines on separate plots (e.g. as we've done in Figs. 5 and 6) but since the focus of this paper is not diurnal profiles we feel that increasing the figure count and the total length of this paper for this is not justified.

7. *The AMS instruments mentioned also measure total PM1 OA. It would have been very informative if the discussion included a comparison with those data as well, either (preferably) alongside the comparisons with SFOA, or (at least) in the same way the BC/EC comparison is presented.*

Comparisons with other components of OA (HOA and ASOA) are presented already in our previous publication: Ots, R., et al.: Simulating secondary organic aerosol from missing diesel-related intermediate-volatility organic compound emissions during the Clean Air for London (ClearfLo) campaign, Atmos. Chem. Phys., 16, 6453–6473, doi:10.5194/acp-16-6453-2016, 2016a. The AMS instruments also assign concentrations to a COA (cooking OA) component which are not routinely modelled because there are no cooking emissions in the emissions inventory for the model. However, in another paper we used the AMS-measured COA concentrations for a model exploration of potential magnitude and spatial distribution of primary COA emissions: Ots et al. "Model simulations of cooking organic aerosol (COA) over the UK using estimates of emissions based on measurements at two sites in London", Atmos. Chem. Phys., 16, 13773-13789, 2016b.

8. *p. 16, l. 18-19 and Figure 13, last row: This site does not add anything to the discussion, I recommend to remove it.*

The paper provides the following explanation for presenting the data shown in Figure 13. "Although the pollutant levels measured close to the traffic source at the Marylebone Road roadside site and the modelled concentrations are not fairly comparable due to the differences in spatial scale (major road vs 5 km model resolution), they are included to illustrate the range of concentrations in a megacity."

We believe that the above remains a relevant reason for presenting the comparisons at Marylebone Road also.

> 9. p. 20, l. 28: I am not entirely convinced that "the combRedist experiments improved the comparisons". Only the negative NMB was really targeted with the experimental design, and it is expected that increased emissions of an inert aerosol tracer will increase aerosol levels at surface, especially close to sources, thus reducing (or even eliminating) the negative bias. Figures 13 and 14, which represent a more regional picture, do not show any significant improvement for that particular simulation either.

We agree that the Base4x experiment only targets the negative model NMB, for the reason the reviewer states: "*it is expected that increased emissions of an inert aerosol tracer will increase aerosol levels at surface*". Hence the aim of the combRedist experiment: namely, to investigate the effect of redistribution of some of the SFOA emissions on both the negative NMB (caused by the fact that the NAEI does not currently assign emissions to smoke control areas), as well as on the other mod-obs metrics presented in this manuscript (r, NMGE, COE), i.e. the redistribution of SFOA sources affects SFOA concentrations in the different air masses arriving at these sites, not just the emissions assigned to the local grid cell. For example, in Detling and Harwell, combRedist had a much lower NMGE and a much better COE than Base4x (Table 4). Furthermore, Figures 13 and 14 show that these tests do not cause completely unnecessary effects in, for example, Northern Ireland, i.e., that we are not completely removing SFOA from lower populated areas and simply ending up with it in the most densely populated areas of London.

> 10. Appendix A1 contains textbook information and it is not necessary, although it consists of a nice collection of references and the discussion is fluid, so I am hesitating to propose to remove it. Appendices A2-A4 should be supplementary material. Appendix A5 should move in section 3.5.

We remain of the opinion that the material in this section contains relevant information to the interpretation of our comparisons of modelled EC with measured BC, the latter of which is measured at many more sites geographically spread across the UK, than with measurements of EC which are made only at 3 sites (2 of which are in London). We believe it is important to challenge the model with the greater amount of measured BC data but also wish to remind readers of issues associated with equating BC and EC. We will take the advice of the ACP editor (or production team) on whether this material is most appropriate as an Appendix or as Supplementary Material, but given its relative brevity we would prefer to keep it as an Appendix.

Technical corrections
> 11. p. 5, l. 4: Is Hjj correct (so please explain) or it should have been Hdd? p. 9, l. 9: Please take this sentence out of the parentheses.

Yes, it should have been Hdd, not Hjj. We have now fixed this, thank you!