

Anonymous Referee #1

We thank the reviewer for their very supportive comments. We respond to each comment individually below. The reviewer's comments are in italics and blue font, our responses are in normal text.

This is a very useful contribution to an ongoing and important debate on the impacts on PM concentrations of wood burning.

- 1. Abstract: It doesn't mention the Redist analysis. Wouldn't it be worth saying that a simple redistribution of emissions according to population is not correct?*

We have included the following in the Abstract:

"A third experiment, *Redist* - all emissions redistributed linearly to population density - is also presented as an indicator of the maximum concentrations an assumption like this could yield."

And the following in the Conclusions:

"A third experiment, *Redist* - all emissions redistributed linearly to population density - is also presented as an indicator of the maximum concentrations an assumption like this could yield. It is recognised however that this is not realistic as the most densely populated areas (of large apartment buildings) are unlikely to have many individual fireplaces."

- 2. Comment on whether the degree day factors actually reflect the use of these wood burners would be useful.*

Yes, people in London do use wood and coal for heating purposes. We have now included the relevant references in "Section 2.2: Model experiments":

"Recent studies in London have shown that local contributions of SFOA coincide with days of low temperature (Fuller et al., 2014; Crilley et al., 2015). Therefore, degree-day factors were included to modulate the daily variation in emissions from the SNAP2 sector according to ambient temperature (i.e. increasing the emissions during colder days)."

- 3. Whilst there is an under prediction at K&C and the Waters paper (3x the emissions) - most experiments are using base emissions of NAEI, why is this? I have read the Waters paper and it not only gives the 3x factor but also the wood use in different UK regions. You did not use these data but could you comment on how the different scenarios you did run compare?*

The Waters' paper is certainly very relevant and should be used to inform a re-evaluation of the National Atmospheric Emissions Inventory (NAEI) assumptions about the extent and spatial distribution of wood and coal burning in urban and rural areas over the UK. However, the paper is a summary of survey information on domestic combustion habits not an emissions database. To derive emissions of SFOA would require combining data and assumptions about amount of wood burned, the appliance types and a range of emission factors, which is well beyond the objectives of our paper. Secondly, the data in the Waters paper were presented for 12 regions (e.g. London, East Midlands, Scotland, Wales, etc.) rather than the much finer grid that could be used in a national atmospheric chemical transport model (e.g. we aggregate NAEI's 1kmx1km emissions to our model grid - 5kmx5km). Again,

the production of new gridded SFOA emissions is well beyond the objectives of our paper, but we note in conclusion of our work that the Waters paper and our modelling study help inform development of updated national emissions inventories for domestic combustion PM.

4. *P13 line 3-4 - What about European assumptions bearing in mind the results in Belgium. Could you comment on how important the long range transport of these emissions are and could be if the results in Belgium and UK are reflected more widely in Europe?*

We have added the following sentences to the top of “3 Results and Discussion” (as that is where we first mentioned European import):

“In our experiments, we did not modify European emissions – we used exactly what has been reported to the CEIP. While there is reason to believe European emissions of SFOA are also underreported, we do not believe this to have a major influence on the surface concentrations of SFOA over the UK as even our Base4x experiment (Fig. 3b) only indicates very modest regional transport of our SFOA to Europe.

5. *It would be good to provide a quick comparison of the Marylebone and Kensington site results. Looking at the map they are close to each other and I guess they have very similar SFOA concentrations. Is this the case? In addition, whilst I realize that you have used what data is available for the UK, could you say something about the limitations in addressing the wood burning emissions inventories UK wide using a small number of sites close to each other in the SE of England.*

This is a good idea and we have now included the annual average measured concentrations in the captions of Figures 5 and 6: “The annual average measured SFOA concentrations at these sites were $0.9 \mu\text{g m}^{-3}$ at Marylebone Road, and $1.0 \mu\text{g m}^{-3}$ at North Kensington.” We agree that only using sites from the same area (Southern England) is not ideal but these datasets are really rather unique – especially in its length as well as the fact that for January 2012 we have 4 sites operating simultaneously, two of each type (urban and rural). However, acknowledging the limitations of using a small number of sites close to each other is exactly why we later present comparisons with the Aethalometer data, including sites from a national network.

6. *Page 11 line 9 - where it says for more discussions see Ots 2016a, why not just add a sentence discussing the measurement uncertainty?*

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 do differ, sometimes by more than a factor of two.”

7. *Page 11 fig 7 - it is clear that the diurnal profile of SFOA is similar at all sites and not reflected in the currently used emissions profile for this source. I have read the work of Fuller in London which showed there to be a strong evening peak in emissions from domestic wood burning, especially at weekends. It would have been good to test an alternative emissions profile, which better reflects the burning of wood and would have been helpful for other model users.*

We agree that the diurnal cycles specified for these emissions need to be modified. We did something very similar in our cooking OA paper (Ots et al. 2016b); cooking oil/meat frying OA is a source that is currently not included in European emissions at all. However, for the work in hand, we felt that including experiments for the spatial distribution and total amount was already quite a lot to present, and that the overall -71% normalised mean bias of modelled vs measured SFOA at London North Kensington should be addressed before the more finer temporal-scale issue of hourly emissions profiles. We acknowledge and call for further work on this in several places in the manuscript, including in the Abstract and the Conclusions.

8. *Fig 8. results - 14th-15th Jan was also a weekend could you comment on the likely weekday to weekend use of wood burners as well as the weekend evening use. What was the evening temperature during these events?*

This is a very good point to make, thank you!

We have included a note about this (plus our subsequent observation that ambient temperatures were somewhat lower during this period) in the Results (italics was already there, **bold font text we have added now**):

*“Nevertheless, southern England did experience a sustained high-pressure weather system during these days, **including noticeably lower temperatures for 14-18 Jan than average (Crilley et al. 2015: Fig.2).** Sustained high pressure usually leads to a very stable atmosphere with descending air masses. Therefore, these high concentrations could have been caused by meteorological build-up, and it is possible the model set-up underestimated the strength of this effect. **Furthermore, 14-15 Jan was a weekend whereby people are more likely to spend time home and therefore potentially use their fireplaces more than on weekdays.**”*

9. *Fig 11 - Comment on Detling daily data. The daily average measurements around the 17th Jan doesn't seem to be reflected in the plot. Is this because of <75% data capture?*

Yes that is the reason. We have now expanded the information about the threshold where Fig. 11 is first introduced:

“Time-series of daily-average concentrations during the winter IOP are shown in Fig. 11. At Detling, the measurements commenced the morning of 16-Jan (Fig. 8a) but since we used a data capture threshold of 75%, 16-Jan and 17-Jan did not include sufficient hourly data points to present measured daily averages for this site.”

10. *Figure 13. I don't really see much point in comparing the results of the model at Marylebone Road, so you should remove this plot.*

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.

11. Figure 14. Could you rescale these plots? You can barely make out the modelled EC in many of them?

We have now added some vertical space to these plots, as well as given the borders of the boxes (previously grey) the same colour as the insides so the very thin ones become more noticeable. The final ACP paper has more space per page than the ACPD format (55 lines vs 35 lines) so the editorial staff will have more space to stretch the figure as well (our image format is PDF so typesetting it on a larger surface will not lose quality). The description of Figure 14 now says “Each box is the interquartile range - IQR,” rather than what is on Figure 13: “Middle line: median, boxes: 25th and 75th quartiles (i.e. the interquartile range - IQR),” as you could not make out the median line on most of the very thin modelled boxes anyway.