

Anonymous Referee #2

We thank the reviewer for their supportive comments on the interest and suitability of the work for publication in ACP. We respond to each comment individually below. The reviewer's comments are in *italics* and blue font, our responses are in normal text.

The paper contains interesting information on important sources of emissions in a major urban area. I have one major concern, but after attention to the points below this paper should be suitable for publication on ACP.

1. *My main worry concerns the assumption that the SFOA (and other POA) emissions are inert. In most VBS modelling studies such emissions are allocated to a number of VBS bins, and allowed to evaporate and react with OH. Further, the results presented for London in Xu et al. (2016) do not show any large SFOA contribution to the low- volatility OM mass, suggesting that the high fractions found in Young et al were of semi- volatile OA. Assuming inert emissions will tend to overestimate the PM concentrations associated with this POA. The authors should re-visit and investigate the implications of their inert assumption.*

The reason this study does not present model simulations with the volatile treatment of SFOA is that for the AMS-PMF data, primary (SF)OA and oxygenated (secondary) OA are separated. Therefore, the direct comparisons with SFOA measurements here do not include the semivolatile components as those would only become condensed after atmospheric ageing but then they would be measured as oxygenated OA, not SFOA. The volatile components and secondary OA precursors are not needed to test the main hypothesis of this paper – that the spatial distribution of wood and coal burning emissions should not be assumed to be zero in smoke control areas. Using primary components to demonstrate this is sufficient.

This is not to say, of course, that SFOA emissions do not include precursors for SOA. The inclusion of semivolatile SOA precursors from SFOA is of course necessary to close the gap between total measured OA and total modelled OA. Indeed, the work by Xu et al. (2016) to which the reviewer refers above (and other work) acknowledge that oxygenated OA likely contains secondary and/or aged SFOA.

However, having reviewed our original text towards the end of Section 2.1 ("In all the experiments presented here, SFOA is assumed to be non-volatile and it does not undergo atmospheric ageing") we acknowledge that we were not clear on the point we make above. We have therefore added another sentence so that this description now reads: "In all the experiments presented here, SFOA is assumed to be non-volatile and does not undergo atmospheric ageing. This is not because it is assumed that there is no aging of SFOA emissions but because the model simulations compare against AMS-PMF determinations of primary SFOA concentrations."

2. *Connected to this, what is the likely status of the emission measurements behind the SFOA inventory for the UK? Do the techniques used to produce the emission factors include condensables? With so much focus on one emission category, and the fact that condensables are a 'hot' topic (Denier van der Gon et al, 2015, Ciarelli et al., 2017), the authors should inform the readers more about such properties.*

This issue of volatility and associated uncertainties seems to be ignored throughout the manuscript.

We have indicated in our response to comment 1 that we have not 'ignored' the issue of volatility because it is not important but because it is not needed in this examination of spatial patterns and magnitudes of primary SFOA PM_{2.5} emissions against primary SFOA measurements. We agree that the issue of 'condensables' in measuring and reporting PM levels is indeed a 'hot' topic. This issue applies to both ambient measurements and to the emissions at source that underpin emissions factors and inventories. It is also an issue that is not just confined to solid-fuel burning. For example, one of our previous papers examined the influence of IVOC emissions associated with diesel vehicles (determined from ambient measurements but not currently included in emissions inventories) on the generation of additional anthropogenic SOA: Ots et al. (2016) "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. The measurement and modelling communities have long been aware of a need to attempt harmonisation in measurement conditions and reporting of PM and associated condensable emissions (e.g. dilution tunnel conditions etc.) across source sectors and across countries. The issue is raised in various fora, e.g. CEIP and TFMM, but it is challenging and slow to agree and implement. In this study we included the best-available county-by-country domestic sector PM_{2.5} emissions as reported to CEIP. This includes instances of countries that do report condensables as part of their PM emissions, e.g. Belgium (who report these as part of the total PM emissions, not separately), and countries that don't. Without considerable more coordination of measurement and reporting it is not possible to make any evidence-based adjustments to the CEIP emissions. In the revised paper we have added a further sentence towards the end of Section 2.1 (after the new sentence inserted in response to comment 1): "It is likely that domestic PM_{2.5} emissions reported to CEIP vary by country according to whether condensables are included in the PM_{2.5} emissions but the specific information is not known."

Other comments

3. *Page 2, L10. The Bergstrom reference is a PhD Thesis. Give the published papers instead.*

Thank you for pointing this out. We now only include the following relevant published reference from the thesis: Denier van der Gon et al. 2015.

4. *Page 2, L5-15. What about emissions from cooking?*

We believe it extremely unlikely that there are significant primary PM emissions associated with use of solid fuels solely for the purpose of domestic cooking in the UK and the rest of Europe. (We assume that this is what the reviewer intended by their question.) We have not encountered any literature to suggest that solid fuel for cooking is a source that needs to be considered. This is not to deny that some domestic solid-fuel heat sources may also be used for cooking. It is certainly the case, however, that there are PM emissions associated with the cooking process, but these have different chemical signature and are categorised separately by AMS-PMF measurements. We have previously undertaken model investigations of this cooking source: 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.

5. Page 2, L16. What is the 'Great London Smog' - give a reference.

We have now included the following reference:

Bell, M. L., Davis, D. L., and Fletcher, T.: A retrospective assessment of mortality from the London smog episode of 1952: the role of influenza and pollution, Environmental Health Perspectives, 112, 6–8, <http://www.ncbi.nlm.nih.gov/pmc/articles/PMC1241789/>, 2004.

6. Page 2, L33. I believe Belgium has also included condensables in their emissions estimates, which brings me back to the point raised above.

Yes, Belgium does include condensables in their total PM emissions estimates from this source, but not separated out. We have responded in detail to the earlier version of this comment above (comment no. 2).

7. Page 3, Sect. 2.1: The text should give some details about the SOA framework used here. What assumptions are made about SVOC, IVOC, and aging? What was done for ASOA and BSOA?

The modelling framework for ASOA and BSOA is as described in the literature cited in the paper, viz:

Ots, R. et al.: Simulating secondary organic aerosol from missing diesel-related intermediate-volatility organic compound emissions during the Clean Air for London (ClearLo) campaign, Atmos. Chem. Phys., 16, 6453-6473, 2016.

Simpson, D. et al.: The EMEP MSC-W chemical transport model - technical description, Atmos. Chem. Phys., 12, 7825-7865, 2012.

As we respond above in response to comment no. 1, we do not undertake any 'aging' treatment for SNAP 2 emissions because the AMS-PMF data provide separate data for primary (SF)OA and oxygenated (secondary) OA. Condensable components of SNAP2 emissions are therefore not needed to test the main hypothesis of this paper – that the spatial distribution of wood and coal burning emissions should not be assumed to be zero in smoke control areas. Using primary components to demonstrate this is sufficient. In order to make this point more directly in our revised paper we have added text to Section 2.1, as we have detailed in our responses above.

8. Page 3. The statistics given for model performance are useful, but they seem only to refer to London. How about elsewhere, since this paper deals with the UK as a whole?

We agree that only using sites from the same area (Southern England) is not ideal but these datasets are really rather unique – especially in their length, time resolution and chemical speciation, 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.

9. *Page 4, Add the ion labels for SO₄, NH₄ and NO₃. (For example NO₃ is a gaseous compounds important for night-time chemistry, whereas I think the authors mean the nitrate ion.)*

Thank you for pointing out this error. The ions are now labelled.

10. *Page 5, L4 claims that Ots et al. (2016a) showed that modelled SFOA were substantially underestimated at North Kensington, but according to Table 3 of that paper the SFOA PMF factors were convolved with the OOA2 factors.*

The data reported in Table 3 was from the following paper, where the measurement dataset and the derivation of the different factors is described in detail: Young, D. E., Allan, J. D., Williams, P. I., Green, D. C., Flynn, M. J., Harrison, R. M., Yin, J., Gallagher, M. W., and Coe, H.: Investigating the annual behaviour of submicron secondary inorganic and organic aerosols in London, *Atmos. Chem. Phys.*, 15, 6351–6366, doi:10.5194/acp-15-6351-2015, 2015.

In the study by Young et al. (2015), a year of measurements were performed using the c-ToF-AMS (compact) and it is the PMF factors from that dataset that contain the two convolved PMF factors. Section 4.4 of the paper describes how these two convolved factors were dealt with. In brief, both factors had a strong and similar diurnal cycle, so the effect of being convolved was reduced by using daily averaged concentrations. In the current work presented here, these daily average concentrations are used for the annual comparisons, thus the issue of the convolved factors should not be significantly influencing the overall observations. In contrast, concentration data from the HR-ToF-AMS (high-resolution) instrument are used for the hourly comparisons presented in the current work. The PMF factors resulting from the HR-AMS data were not found to be convolved, which is likely due to a combination of the fact that the measurements were high resolution and the HR-ToF-AMS was deployed only during the intensive observation periods, rather than the full year like the c-ToF-AMS. Consequently, hourly comparisons presented in this work use the HR-ToF-AMS data.

In summary, whilst we acknowledge the general issue of convolved factors, it is not likely influencing the overall results in the current study since datasets of the appropriate time resolution were used for each of the cases and each of these datasets had been treated so as to reduce the issue.

11. *Page 6, Fig. 2. Units of Mg/km² would be easier for comparison with other studies.*

We have added the following to the caption: “20 Mg per (our) grid square is 0.8 Mg km⁻²”

12. *Page 11, Sect 3.2. Measured profiles of SFOA result from a mixture of emissions profiles, atmospheric dispersion, and PMF interpretation. The model should capture the first two, but I wonder how much PMF contributes. For example, if the diurnal emissions profile is responsible for the concentrations profile, why would SFOA emissions peak around midnight for N. Kensington?*

We anticipate that the peak around this time is due to the product of the impacts of greater evening local emissions and reducing boundary layer height and increasing atmospheric stability during night-time.

13. Page 12, L13 and associated text and Figures. Were these "exceptional" concentrations also seen for other pollutants, for example NO₂. Would model performance for other components help the analysis here? (Also, the word exceptional seems a bit excessive here. Are such concentrations really so infrequent?)

These concentrations (e.g. 12 $\mu\text{g m}^{-3}$ daily average at Harwell) are indeed exceptional as this is just one component. The recommended maximum daily average concentration for total PM_{2.5} is no more than 10 $\mu\text{g m}^{-3}$ and that must accommodate all components: organic and inorganic, not just SFOA. Furthermore, our Figures 5 shows that daily average concentrations throughout the year exceed 4 $\mu\text{g m}^{-3}$ only a few days a year.

Comparisons with other pollutants are presented in Figure 6 in:

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.

14. Page 21. The WRF model is also open source, and details should be included here. I think section 5 and 6 could also be merged, since the code is mentioned in both. Currently it is confusing though, since Sect. 5 says code should be obtained from www.emep.int, but Sect. 6 says code is from the University of Edinburgh.

The ACP submission instructions request these two separate sections. Where we mention "code" in Section 6, we do not mean the core source code but the scripts used to initialise a model simulation (i.e. linking all our input files to the model executable). We have now changed the word "code" to "scripts" in Section 6.

15. Appendices: This type of information is typically provided as Supplementary material.

We will take the advice of the ACP editor (or production team) on this point.