

acp-2017-568: Modelling carbonaceous aerosol from residential solid fuel burning with different assumptions for emissions

Ots, R. et al.

Response to additional review comments

The revised manuscript is much improved, and it is especially good to know the status of the UK's OA emissions inventory, and that condensables are not included.

Response: Thank you!

I just saw one issue with the revised text. In Sect. 2.1, the additional text which starts 'Based on partitioning calculations for a range of OA ...' should be deleted, since the assumptions about a 10 ug/m³ bin are not relevant and this estimation therefore incorrect. The concentrations relevant to a consideration of a 'May et al' type evaporation of POA are those of the dilution chamber, which are far higher than 10 ug/m³.

Response: We have removed this text.

As the authors now state clearly that they are dealing with inert SFOA emissions, I suggest they delete this 10 ug/m³ calculation, and re-arrange the text a little. I would have started with comments about the potential importance of evaporation (citing May et al, Denier van der Gon et al) but then gone on to explain that these complications are not needed since the UK has just inert SFOA in its inventory.

Response: We have removed the calculation and re-arranged the text as suggested (e.g. the UK SFOA sentence now follows the May and van der Gon references).

Descriptions of the modelling framework for ASOA and BSOA are given in Simpson et al. (2012) and Ots et al. (2016a). In all the experiments presented here, SFOA is assumed to be non-volatile and it does not undergo atmospheric ageing.

~~The UK emissions inventory for domestic PM_{2.5} does not include condensables, but this information is not known for the emissions reported to CEIP by other countries.~~

5 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
10 to be zero in smoke control areas. Using primary components to demonstrate this is sufficient. This is not to say 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) acknowledged that oxygenated OA likely contains secondary and/or aged SFOA.

Furthermore, the various sampling methods used to derive emission factors (which are applied by each country reporting emissions to CEIP) vary greatly (Denier van der Gon et al., 2015). The two main types are filter measurements (capturing only
15 solid particles), and dilution tunnel measurements (capturing solid particles and condensable organics). The difference between the two methods can be large - up to 5-fold for woodburning (Denier van der Gon et al., 2015), which is similar to what was shown by May et al. (2013) that up to 80% of the mass of POA from biomass burning may evaporate when diluted from plume to ambient
20 ~~conditions. Based on partitioning calculations for a range of OA (from 0.01 to 10⁵) in Donahue et al. (2006), only the 10 volatility bin exhibits a substantial portions in both gaseous and solid form. The organic components in all other bins are either mostly solid or mostly gaseous. May et al. (2013) also shows that only 10% of biomass burning emissions (Table 2 in May et al. (2013)) is in the 10 bin. Therefore, the potential overestimation arising from not letting these solids in the condition. The UK emissions inventory evaporate is no more than 10% (as the lower volatility materials is almost completely solid, and the higher volatility material is almost completely gaseous) for domestic PM_{2.5} does not include condensables, but
25 this information is not known for the emissions reported to CEIP by other countries.~~

The performance of this version of the EMEP4UK model simulating a standard suite of gas-phase components and secondary inorganic aerosol PM components is reported in Ots et al. (2016a) comparing with a full year of measurements in London in 2012. In brief, Ots et al. (2016a) reported an NMB of -1% and $r = 0.79$ for ozone, an NMB of -32% and $r = 0.78$ for NO_x, an NMB of +6% and $r = 0.73$ for SO₄²⁻, an NMB of -12% and $r = 0.65$ for NH₄⁺, and an NMB of -23% and $r = 0.57$ for
30 NO₃⁻.

2.2 Model experiments

In this study, four different cases were considered. The *Base* case model experiment uses the same emission inventory dataset as Ots et al. (2016a) (i.e. as reported by the NAEI using the splits in Fig. 1), but with a small adjustment in the daily variation in emissions due to temperature, called degree-day factors (Simpson et al., 2012). Recent studies in London have shown