

Dear Editor,

Thank you very much for your prompt decision, as well as for bringing the two very recent and relevant references to our attention. We have included both, the sentences are highlighted below.

For the present study, a COA tracer was added into the model with dry and wet deposition properties similar to other fine (i.e. $\text{PM}_{2.5}$) primary OA components (see Simpson et al. (2012) for aerosol specifications in the EMEP MSC-W model). This COA tracer is non-volatile and does not undergo chemical ageing, but it is included in the total OA budget for the absorptive partitioning of secondary organic aerosol species. Treating POA as non-volatile is a simplification as in reality, some POA may evaporate on atmospheric dilution, go through atmospheric ageing, and condense back into the particulate phase thus becoming secondary OA (SOA; Robinson et al. (2007)). The volatility distribution and ageing rates are, however, still a major source of uncertainty in atmospheric chemistry models (Ots et al. (2016) and references therein). Furthermore, some POA emissions are accompanied by emissions of intermediate volatility organic compounds (IVOCs; e.g. Shrivastava et al. (2008) based on Schauer et al. (1999)), but to our knowledge, there are currently no measurements or estimates of cooking-IVOCs to use as a basis for modelling. [Very recently, Klein et al. \(2016\) presented emission factors of different volatile organic compounds \(VOCs\) from a variety of cooking styles and techniques based on laboratory measurements, but more measurements are needed before these can be included in the chemical schemes of atmospheric chemistry transport models.](#)

2.2 AMS measurements used in this study

The construction of COA emissions estimates were based on measurements made during the ClearLo project (Bohnenstengel et al., 2014) at two sites in London, shown in Fig. 1. Marylebone Road is a ‘kerbside’ site on the edge of a heavily-trafficked urban through-road, whilst the North Kensington site is classified as urban background and is situated in the carpark of a school.

The measurements at Marylebone Road were taken with a Q-AMS (Quadrupole AMS; Jayne et al. (2000)) between 11-Jan-2012 and 1-Feb-2013 and were averaged to hourly values, yielding 5996 data points (Detournay et al. (2015); several gaps in the measured data were caused by problems with the instrument computer). Annual average OA_1 during 2012 at the Marylebone Road site was measured at $8.5 \mu\text{g m}^{-3}$: $0.8 \mu\text{g m}^{-3}$ SFOA (9%), $3.0 \mu\text{g m}^{-3}$ SOA (36%), $2.5 \mu\text{g m}^{-3}$ HOA (29%), and $2.2 \mu\text{g m}^{-3}$ COA (26%).

The measurements at North Kensington were taken with a cToF-AMS (compact Time of Flight AMS; DeCarlo et al. (2006)) between 11-Jan-2012 and 23-Jan-2013, and with a HR-ToF-AMS between 21-Jul-2012 and 19-Aug-2012 (High-Resolution ToF-AMS), hourly averaging yielded 8035 data points (Young et al., 2015). Annual average OA_1 during 2012 at the North Kensington site was measured at $4.2 \mu\text{g m}^{-3}$: $1.0 \mu\text{g m}^{-3}$ SFOA (24%), $1.6 \mu\text{g m}^{-3}$ SOA (38%), $0.8 \mu\text{g m}^{-3}$ HOA (19%), and $0.8 \mu\text{g m}^{-3}$ COA (19%).

Figure S1 shows a satellite image of the Marylebone Road measurement site with food-related commercial establishments (cafes, restaurants, etc.), as known to Google, marked. (The accuracy or comprehensiveness of these establishments marked on Google Maps has not been verified, but are presented to illustrate the number of food outlets in the area.) There is no direct source of cooking emissions close to the Marylebone Road measurement site, so the measured concentrations, although high, are likely to represent an average of the many COA emissions sources in the vicinity.

Positive matrix factorisation (PMF) seeks to reproduce the measured time series of the organic mass spectrum through a linear composition of a (user-selectable) number of factor spectra (representing different OA types or sources) and their mass

contribution, taking into account the precision associated with each measurement. Subjectivity is minimized by comparison of concentration time-series with independent measurements and assessment of the robustness of the solution, e.g. through boot-strapping. COA has been identified as a contributor to urban OA measurements because it exhibits a distinct diurnal cycle and the associated factor spectrum is very similar to that of lab-generated cooking oil aerosol (Allan et al., 2010). Nevertheless, there are some inherent uncertainties involved in deriving COA concentrations with AMS measurements. For example, AMS measurements need to be corrected for the fraction of aerosol that is not effectively vaporised due to bounce from the hot surface involved in the AMS's detection mechanism. Whilst this is well characterised for typical, internally-mixed ambient aerosols (e.g. Middlebrook et al. (2012)), it is possible that the COA measured by the AMS is not well mixed with other aerosol components and could therefore be detected at a higher efficiency. If this were the case, AMS measurements may overestimate COA concentrations by up to a factor of 2.

Furthermore, Hayes et al. (2013) observed that the correlation between HOA+COA and CO is stronger than the correlation between just HOA and CO (0.71 vs 0.59). They speculated this could mean the COA component identified may also include some particulate mass from non-cooking sources such as traffic.

Indeed, a study comparing AMS-PMF derived concentrations of PM components with those estimated based on measurements and a chemical mass balance (CMB) model at the North Kensington site during a 2-week period in the same campaign used in this study concluded that AMS derived COA was on average 1.6 times higher than the CMB derived values, but good correlation was seen (a linear fit of $AMS_{COA} = 2.24 \times CMB_{COA} - 0.33$, with $r = 0.89$), Yin et al. (2015)), which is consistent with the AMS collection efficiency (CE) being higher than the usual 0.5. ~~There are also additional sources of uncertainty with PMF, in particular rotational ambiguity, which can cause both over- and underestimates (Allan et al., 2010; Paatero et al., 2002).~~ However, the CMB approach is also not without its uncertainties, in particular that the COA marker(s) used in the CMB may not be fully representative, and because of the need to scale marker concentration to COA concentration. There are also additional sources of uncertainty with PMF, in particular rotational ambiguity, which can cause both over- and underestimates (Allan et al., 2010; Paatero et al., 2002). Using an optimized constrained statistical approach on their PMF analysis of AMS measurements in China, Elser et al. (2016) observed the contribution of COA to total OA to reduce by a factor of 2.8 (from 19.9% to 7.7%) compared with the unconstrained PMF approach. While source apportionment analysis in the UK may be more straightforward compared to China because of additional important sources in China (especially coal), uncertainties in PMF apportionment in general remain and are not only due to collection efficiency.

In summary, the full quantification of COA by AMS (and any other approach) requires further research but it is currently more likely that the AMS overestimates the COA than underestimates it.

2.3 Spatial distribution of COA emissions

Figure 1a shows the residential population density data in the central London area at $1 \text{ km} \times 1 \text{ km}$ resolution, overlaid by the EMEP4UK grid cells ($5 \text{ km} \times 5 \text{ km}$), and Fig. 1b the equivalent workday population¹ density. These datasets were compiled

¹The *workday population* is a redistribution of the usually resident population to their place of work, while residents who are not in work remain at their area of residence. The workday population of an area is defined as “all usual residents aged 16 and above who are in employment and whose workplace

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