

Interactive comment on “Anthropogenic Secondary Organic Aerosols Contribute Substantially to Air Pollution Mortality” by Benjamin A. Nault et al.

Anonymous Referee #3

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The study represents an attempt to estimate the premature mortality linked to Anthropogenic Secondary Organic Aerosols. Using 11 urban areas on three continents and specific volatile organic compounds emission ratios were estimated and a budget for ASOA is attempted. With the studied dataset the SIMPLE parameterization for ASOA in the GEOS-Chem model is updated to reproduce observed ASOA. Finally an attribution of ASOA PM_{2.5} premature deaths is attempted.

General comment:

My greatest concern for the specific study is the overall omission of solid fuel combustion in all calculations, both for ASOA production (emissions and subsequent pro-

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cessing/oxidation/ageing) as well as its contribution to premature mortality. Not only biomass burning for heating purposes but also forest fires, burning of crops etc. This leads to unaccounted emissions from urban areas such as Europe/US during winter from household heating but also from forested areas such as the Amazon, Canada, Siberia, Southeast Asia.

Specific comments:

1) Line 110-114: Isn't solid fuel combustion/biomass burning aged SOA considered as ASOA? According to Kodros et al. 2020 in active fire regions bbOOA increases by more than 50-60% from fast oxidation processes even in the dark. Significant contribution of primary BBOA oxidation to the oxygenated OA have also been identified in large urban centers such as Paris (Petit et al., 2014) and Athens (Stavroulas et al., 2019).

2) Line 119-132: Isn't the current study also under-predicting ASOA by ignoring bbOOA? Furthermore, there is also the additive effect of the different pollutants when considering premature mortality. For example, Kodros et al. 2017 estimate joint exposure from household solid fuel use and ambient PM_{2.5} pollution and find 18% more deaths than by separating household and ambient mortality calculations. Which shows that solid fuel combustions is important for mortality as well, not only for ASOA calculations.

3) Fig.5a and Line 174-180, Fig. 6 and line 423-428: Authors only mention the uncertainties in x- and y-axis values. Does really by removing just one point increases the slope that much? The y-axis has an upper value of 140 compared to x-axis of 6! Why only ~25% of the observed ASOA can be associated with BTEX? What about the rest? Isn't this a solid proof that solid fuel combustion (BBOA) should definitely be taken into account?

4) Section 2.5.2 Once more, by not including solid fuel combustion in ASOA all the respective chemistry and oxidation is missing, losing 50-60% of SOA from fast oxidation of BBOA, even in the dark (NO₃ radicals) (Kodros et al., 2020).

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5) Line 578-579: How is the “model constrained to atmospheric observations for a more accurate contribution of ASOA” when an important source of ASOA such as solid fuel combustion is omitted?

Technical corrections:

Fig. 7 & 8: USOA? Should it be ASOA?

References

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