Response to reviewers' comments on the paper "Anthropogenic Secondary Organic Aerosols Contribute Substantially to Air Pollution Mortality"

We would like to thank the reviewers for their time and for their useful comments that have helped to improve and clarify our paper. For ease, comments from reviewers are in black, responses in blue, and new text added to paper in **bold blue**.

Reviewer #1

1.0 This manuscript leverages an impressive breadth and diversity of data to shed light on a critical public health and environmental policy question: how many premature deaths can be avoided annually with reductions in emissions of organic compounds that humans have direct control over? The methods and approaches that are developed are generally sound, although far from perfect, for this kind of high-level endeavor. My main issue with the paper in its current form are the confusing organization and presentation of ideas, some slight misplaced focus on particular organic aerosol model updates, and the decision to ignore solid fuel burning in the model formulation. These and other issues below should be addressed before publication.

We thank the reviewer for the overall positive review and the detailed input. We have replied to all the specific points below.

General Comments:

1.1 I think the main ideas in the paper are quite compelling: a) reconstruct measured SOA from in situ campaigns using correlations with likely predictors, b) incrementally improve a streamlined parameterization for SOA prediction (SIMPLE), c) integrate the SIMPLE predictions into a full-science CTM prediction of PM2.5 and use satellite data to further refine the predictions, d) feed those predictions to a premature death parameterization to quantify human health impact, and e) investigate key sensitivities. I would reorganize the entire paper so that the methods, results and discussion each flow in that order. Currently, the introduction gives little clue about how the pieces will fit together or the goals of the paper, beyond showing that ASOA is important. Much of (b) above is discussed inappropriately in the current results section 4. Manuscript sections 3, 4, and 5 contain quite a lot of methods discussion that should be moved out to section 2. For example, equations rarely belong in a results section. I could even suggest that most of section 2.2 and 2.4 be moved to SI. The details of the chemical mechanism used in 2.4 are a bit irrelevant once the SOA/R_BTEX enhancement ratio is confirmed for use in SIMPLE. If the mechanism were more sophisticated (e.g. HOM formation, carbon-conserving fragmentation to lower MW products, oligomerization, etc) then I think there would be more cause for focusing on it, but the schemes used here are relatively close to the SIMPLE approach in terms of one-way generation of SOA.

Perhaps some confusion arises from the fact that the paper has two sets of major results. The explanation of the parameters controlling the variability of ASOA at major locations is an important result by itself. The application of state-of-the-art methods (that apply the improved quantification of ASOA) to provide the first realistic estimate of the mortality associated specifically with ASOA is a second important result. Perhaps they could have been reported in two separate papers, but we decided to report them together. Therefore the structure of the paper does make sense, because (a) and (d) are both key results, and (b) and (c) are needed to connect those results.

We have nevertheless made an effort to streamline the structure of the paper to reduce possible confusion for some readers. We have added more discussion in the introduction to better frame the uncertainty in ASOA production impacting both models and the ability to apportion emissions to reduce premature mortality. We have both expanded the methods section (e.g., moving some sections, such as 4.1, into methods) and moved some methods into the SI (those that seemed less important for understanding the entirety of the study, e.g., Error Analysis, Box Model, and GEOS-Chem Description). Further, we have expanded the discussion in Sect. 4 concerning the discussion of implementation and improvement of the SIMPLE model compared to what is currently used in GEOS-Chem.

The text added is:

In introduction:

"The main method to estimate premature mortality with $PM_{2.5}$ is to use measured $PM_{2.5}$ from ground observations along with derived $PM_{2.5}$ from satellites to fill in missing ground-based observations (van Donkelaar et al., 2015, 2016). To go from total $PM_{2.5}$ to species-dependent and even sector-dependent associated premature mortality from $PM_{2.5}$, chemical transport models (CTMs) are used to predict the fractional contribution of species and/or sector (e.g., Lelieveld et al., 2015; van Donkelaar et al., 2015, 2016; Silva et al., 2016). However, though CTMs may get total $PM_{2.5}$ or even total species, e.g., organic aerosol (OA), correct, the model may be getting the values right for the wrong reason (e.g., de Gouw and Jimenez, 2009; Woody et al., 2016; Murphy et al., 2017; Baker et al., 2018; Hodzic et al., 2020). This is especially important for OA in urban areas, where models have a longstanding issue under predicting secondary OA (SOA) with some instances of over predicting primary OA (POA) (de Gouw and Jimenez, 2009; Dzepina et al., 2009; Hodzic et al., 2010; Woody et al., 2016; Zhao et al., 2016a; Janssen et al., 2017; Jathar et al., 2017). Further, this bias has even been observed for highly aged aerosols in remote regions (Hodzic et al., 2020). As has been found in prior studies for urban areas (e.g., Zhang et al., 2007; Kondo et al., 2008; Jimenez et al., 2009; DeCarlo et al., 2010; Hayes et al., 2013; Freney et al., 2014; Hu et al., 2016; Nault et al., 2018; Schroder et al., 2018) and highlighted here (Fig. 1), a substantial fraction of the observed submicron PM is OA, and a substantial fraction of the OA is composed of SOA (approximately a factor of 2 to 3 higher than POA).

Thus, to better understand the sources and apportionment of $PM_{2.5}$ that contributes to premature mortality, CTMs must improve their prediction of SOA versus POA, as the sources of SOA precursors and POA can be different.

However, understanding the gas-phase precursors of photochemically-produced anthropogenic SOA (ASOA, defined as the photochemically-produced SOA formed from the photooxidation of anthropogenic volatile organic compounds (AVOC) (de Gouw et al., 2005; DeCarlo et al., 2010)) quantitatively is challenging (Hallquist et al., 2009). Note, for the rest of the paper, unless explicitly stated otherwise, ASOA refers to SOA produced from the photooxidation of AVOCs, as there are potentially other relevant paths for the production of SOA in urban environments (e.g., Petit et al., 2014; Kodros et al., 2018, 2020; Stavroulas et al., 2019). Though the enhancement of ASOA is largest in large cities, these precursors and production of ASOA should be important in any location impacted by anthropogenic emissions (e.g., Fig. 1). ASOA comprises a wide range of condensable products generated by numerous chemical reactions involving AVOC precursors (Hallquist et al., 2009; Hayes et al., 2015; Shrivastava et al., 2017). The number of AVOC precursors, as well as the role of "non-traditional" AVOC precursors, along with the condensable products and chemical reactions, compound to lead to differences in the observed versus predicted ASOA for various urban environments (e.g., de Gouw and Jimenez, 2009; Dzepina et al., 2009; Hodzic et al., 2010; Woody et al., 2016; Janssen et al., 2017; Jathar et al., 2017; McDonald et al., 2018). One solution to improve the prediction in CTMs is to use a simplified model, where lumped ASOA precursors react, non-reversibly, at a given rate constant, to produce ASOA (Hodzic and Jimenez, 2011; Hayes et al., 2015; Pai et al., 2020). This simplified model has been found to reproduce the observed ASOA from some urban areas (Hodzic and Jimenez, 2011; Hayes et al., 2015) but has issues in other urban areas (Pai et al., 2020). This may stem from the simplified model being parameterized to two urban areas (Hodzic and Jimenez, 2011; Hayes et al., 2015). These inconsistencies impact the model predicted fractional contribution of ASOA to total $PM_{2.5}$ and thus the ability to understand the source attribution to $PM_{2.5}$ and premature deaths."

Other updates for the introduction can be found in other specific comments (e.g., R1.6, R1.7, R1.8, and R1.9).

For Sect. 3.1, added information can be found in R1.3, and for Sect. 3.2, added information can be found in R1.2.

In Sect. 4, about the SIMPLE model improvements, the following has been added:

"The "improved" SIMPLE shows higher ASOA compared to the default VBS GEOS-Chem (Fig. 6a,b). In areas strongly impacted by urban emissions (e.g., Europe, East Asia, India, east and west coast US, and regions impacted by Santiago, Chile, Buenos Aires, Argentina, Sao Paulo, Brazil, Durban and Cape Town, South Africa, and Melbourne and Sydney, Australia), the "improved" SIMPLE model predicts up to 14 μg m^{-3} more ASOA, or ~30 to 60 times more ASOA than the default scheme (Fig. 6c,d). As shown in Fig. 1, during intensive measurements, the ASOA composed 17-39% of PM_1, with an average

contribution of ~25%. The default ASOA scheme in GEOS-Chem greatly underestimates the fractional contribution of ASOA to total $PM_{2.5}$ (<2%; Fig. 6e). The "improved" SIMPLE model greatly improves the predicted fractional contribution, showing that ASOA in the urban regions ranges from 15-30%, with an average of ~15% for the grid cells corresponding to the urban areas investigated here (Fig. 6f). Thus, the "improved" SIMPLE predicts the fractional contribution of ASOA to total $PM_{2.5}$ far more realistically, compared to observations. As discussed in Sect. 2.3 and Eq. 11, having the model accurately predict the fractional contribution of ASOA to the total PM is very important, as the total $PM_{2.5}$ is derived from satellite-based estimates (van Donkelaar et al., 2015), and the model fractions are then applied to those total $PM_{2.5}$ estimates. The ability for the "improved" SIMPLE model to better represent the ASOA composition provides confidence attributing the ASOA contribution to premature mortality."

The figures that have been added to the paper to accompany the text above are:

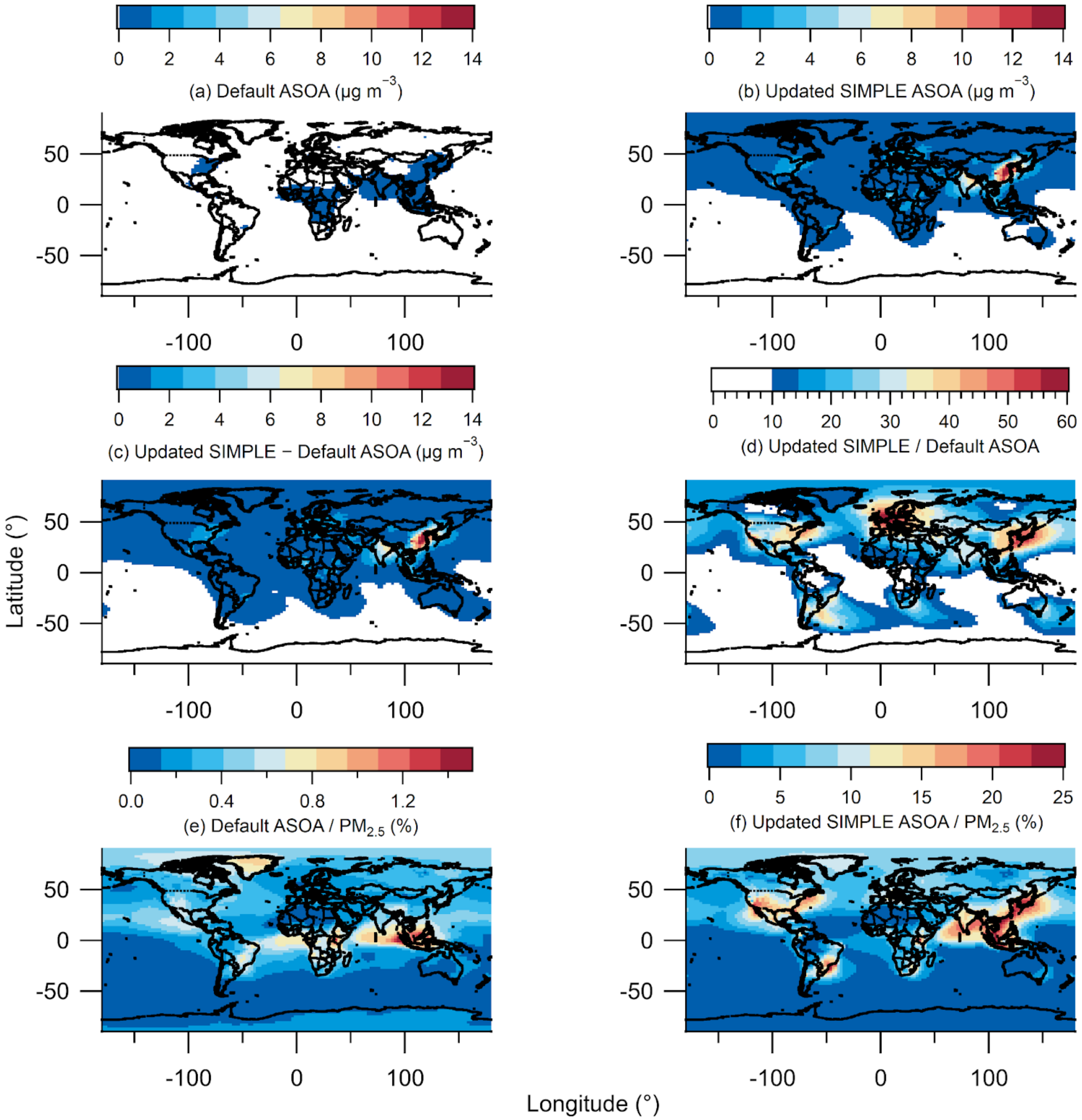


Figure 6. (a) Annual average modeled ASOA using the default VBS. (b) Annual average modeled ASOA using the updated SIMPLE model. (c) Difference between annual average modeled updated SIMPLE and default VBS. (d) Ratio between annual average modeled updated SIMPLE and default VBS. (e) Percent contribution of annual average modeled ASOA using default VBS to total modelled $PM_{2.5}$. (f) Percent contribution of annual average modeled ASOA using updated SIMPLE to total modelled $PM_{2.5}$.

Finally, added text for Sect. 5 can be found in R1.3.

1.2 The authors repeatedly compare their updated approach including semivolatile POA to previous efforts to assess ASOA impacts on human health using nonvolatile POA assumptions.

The implication here is that treating POA as semivolatile might be as important to getting ASOA correct as the dramatic increase in ASOA precursors. But one look at Fig. 6 shows that in most cases it's the way BTEX, IVOCs and Other Aromatics are treated (emissions, SOA yields, aging) that is really driving the ASOA formation. So while I think the update to semivolatile POA is a good one and it gives room for greater ASOA production, I think the authors focus on it too much in this study (see specific comment #3 below and lines 574-581 for examples that should be addressed). Instead there should be much more focus in the budget section on how the parameters have chosen for their VCP emissions and SOA chemistry scheme are driving larger IVOC and VOC contributions and how confident they are in those parameters. For the SIMPLE and GEOS-Chem sections, the key sensitivity is the SOA/R_BTEX ratio (see specific comment #7) and there should be more discussion on its impact. Also, the SIMPLE rate constant k is parameterized from the CalNex data, correct? Why was that not revisited and optimized for performance among all the measurement campaigns?

Perhaps there is some misunderstanding of Fig. 6 from the ACPD submission. So first, we would like to clarify Fig. 6. In Fig. 6, the two panels are not two different ways the VOCs are treated, but two different ways to apportion the same VOCs: on the left by type of species, on the right by the source of the species.

We have added the following text at the beginning of Sect. 3.2 to clarify this point:

"To investigate the correlation between ASOA and R_{BTEX}, a box model using the emission ratios from BTEX (Table S5), other aromatics (Table S8), IVOCs (Sect. S1), and SVOCs (Sect. S1) was run for five urban areas: New York City, 2002, Los Angeles, Beijing, London, and New York City, 2015 (see Sect. S1 and S3 for more information). The differences in the results shown in Fig. 4 are due to differences in the emissions for each city."

We believe the point made in lines 573 - 575 is very important, as many models may get the total OA approximately correct while getting SOA vs POA incorrect (e.g., Hodzic et al., 2020). This in turn can mean that focus of emission controls may be misplaced on reducing POA while neglecting the emissions that lead to the observed ASOA concentrations (e.g., IVOCs from traditional and non-traditional sources). Though POA and IVOC emissions may sometimes originate from similar sources, e.g., diesel (Zhao et al., 2014), the IVOCs will also be emitted from sources that do not include POA, e.g., VCPs (McDonald et al., 2018).

We agree that more emphasis by the community on VCP and IVOC emissions and their SOA production is important. The present paper can also be viewed as a follow-up study to McDonald et al. (2018) that shows the applicability of VCP emissions outside the United States.

Further, we have added the following text to further address these points:

First, about SVOCs:

“Note, the emissions investigated here ignore any oxygenated VOC emissions not associated with IVOCs and SVOCs due to the challenge in estimating the emission ratios for these compounds (de Gouw et al., 2018). Further, SVOC emission ratios are estimated from the average POA observed by the AMS during the specific campaign and scaled by profiles in literature for a given average temperature and average OA (Robinson et al., 2007; Worton et al., 2014; Lu et al., 2018). As most of the campaigns had an average OA between 1 and 10 µg m^{-3} and temperature of ~298 K, this led to the majority of the estimated emitted SVOC gases in the highest SVOC bin. However, this does not lead to SVOCs dominating the predicted ASOA due to taking into account the fragmentation and overall yield from the photooxidation of SVOC to ASOA.”

We agree the key parameter is SOA/R_{BTEX}, and the purpose of Sect. 3.2 is to explore this ratio. As shown in Fig. 6 of the original manuscript, assuming a constant ratio for SOA/R_{BTEX} (the slope from Fig. 5), we are able to explain most of the observed ASOA with the box model and emission inventories. We have added the following text to clarify this point:

“This investigation shows that the bottom-up calculated ASOA agrees with observed top-down ASOA within 15%. As highlighted above, this ratio is explained by the co-emissions of IVOCs with BTEX from traditional sources (diesel, gasoline, and other fossil fuel emissions) and VCPs (Fig. 5) along with similar rate constants for these ASOA precursors (Table S12). Thus, the ASOA/R_{BTEX} ratio obtained from Fig. 2 results in accurate predictions of ASOA for the urban areas evaluated here, and this value can be used to better estimate ASOA with chemical transport models (Sect. 4).”

The rate constant of the SIMPLE model, as stated in Line 513 - 520 in the original manuscript, was originally parameterized to the observations from both Mexico City and Los Angeles. It is also generally consistent with observations of ASOA formation with a time scale of 1 day in other studies (e.g., de Gouw et al., 2005; DeCarlo et al., 2010; Nault et al., 2018; Schroder et al., 2018).

The following has been added to the text to also reflect this point:

“This rate constant is also consistent with observed ASOA formation time scale of ~1 day that has been observed across numerous studies (e.g., de Gouw et al., 2005; DeCarlo et al., 2010; Hayes et al., 2013; Nault et al., 2018; Schroder et al., 2018).”

1.3 The authors spend some time in the discussion addressing the fact that solid fuel combustion emissions are missing from this study. I'm still very concerned that much of the global results they show are corrupted by this emission, not just in southern Asia and Africa. Large regions of Northern/Western Europe and North America will also be affected by residential wood fuel burning, especially in the winter. The authors should at least justify their omission of solid fuels for the measurement campaigns citing tracer analyses, for example. To address global comparisons, can you add a reference to SOA/CO ratios for wood combustion and comment on their similarity or difference from what has gone into SIMPLE for this study?

This is not quite correct. Unfortunately, we did not emphasize enough that two studies used to constrain the ΔSOA/ΔCO vs R_{BTEX}/ΔCO slope shown in Fig. 5a, and thus constrains the updated SIMPLE model, are from campaigns that include large contributions from solid fuel combustion. These include a wintertime campaign in the Northeast US (Schroder et al., 2018) and a late winter, early spring campaign in China (Hu et al., 2013). Both of these studies were strongly impacted by solid fuel combustion, as highlighted in Table S9 in the "Other POA" category (for NYC as we do not have reliable emissions inventory for the observations from Hu et al. (2013)).

Importantly, as we discuss with the updated analysis on the influence of any one point for the slope shown in Fig. 5a (see response to R1.12), the data from these two studies are very close to the slope and do not influence the results. Thus, within the limitations of the available datasets, solid fuels are included and do not result in deviations for the parameterization derived in this study. Clearly it is useful to investigate this point further using data from future campaigns, as we are not aware of any other past campaigns with complete enough data to perform these analyses.

We have added the following text to the revised paper to explain this point in more detail:

"An important aspect of this study is that most of these observations occurred during spring and summer, when solid fuel emissions are expected to be lower (e.g., Chafe et al., 2015; Lam et al., 2017; Hu et al., 2020). Further, the most important observations used here are during the afternoon, investigating specifically the photochemically produced ASOA. These results here might partially miss any ASOA produced through nighttime aqueous chemistry or oxidation by nitrate radical (Kodros et al., 2020). However, two of the studies included in our analysis, Chinese Outflow (CAPTAIN, 2011) and New York City (WINTER, 2015), occurred in late winter/early spring, when solid fuel emissions were important (Hu et al., 2013; Schroder et al., 2018). We find that these observations lie within the uncertainty in the slope between ASOA and R_{BTEX} (Fig. 2a). Their photochemically produced ASOA observed under strong impact from solid fuel emissions shows similar behavior as the ASOA observed during spring and summer time. Thus, given the limited datasets currently available, photochemically produced ASOA is expected to follow the relationship shown in Fig. 2a and is expected to also follow this relationship for regions

impacted by solid fuel burning. Future comprehensive studies in regions strongly impacted by solid fuel burning are needed to further investigate photochemical ASOA production under those conditions."

In addition, we have also added the following text to section 5 to address the potential uncertainties:

"Solid fuels are used for residential heating and cooking, which impact the outdoor air quality as well (Hu et al., 2013, 2016; Lacey et al., 2017; Stewart et al., 2020), and which also lead to SOA (Heringa et al., 2011). As discussed in Sect. 3.1, though the majority of the studies evaluated here occurred in spring to summer time, when solid fuel emissions are decreased, two studies occurred during the winter/early spring time, where solid fuel emissions were important (Hu et al., 2013; Schroder et al., 2018). These studies still follow the same relationship between ASOA and R_{BTEX} as the studies that focused on spring/summer time photochemistry. Thus, the limited datasets available indicate that photochemically produced ASOA from solid fuels follow a similar relationship to that from other ASOA sources.

Also, solid fuel sources are included in the inventories used in our modeling. For the HTaP emission inventory used here (Janssens-Maenhout et al., 2015), small-scale combustion, which includes heating and cooking (e.g., solid-fuel use), is included in the residential emission sector. Both CO and BTEX are included in this source, and can account for a large fraction of the total emissions where solid-fuel use may be important (Fig. S15). Thus, as CO and BTEX are used in the updated SIMPLE model, and campaigns that observed solid-fuel emissions fall within the trend for all urban areas, the solid-fuel contribution to photochemically-produced ASOA is accounted for (as accurately as allowed by current datasets) in the estimation of ASOA for the attribution to premature mortality.

Note that recent work has observed potential nighttime aqueous chemistry and/or oxidation by nitrate radical from solid fuel emissions to produce ASOA (Kodros et al., 2020). Thus, missing this source of ASOA may lead to an underestimation of total ASOA versus the photochemically-produced ASOA we discuss here, leading to a potential underestimation in the attribution of ASOA to premature mortality. From the studies that investigated "night-time aging" of solid-fuel emissions to form SOA, we predict that the total ASOA may be underestimated by 1 to 3 µg m^{-3} (Kodros et al., 2020). This potential underestimation, though, is less than the current underestimation in ASOA in GEOS-Chem (default versus "Updated" SIMPLE)."

Have also added the following figure in SI to go with the text above:

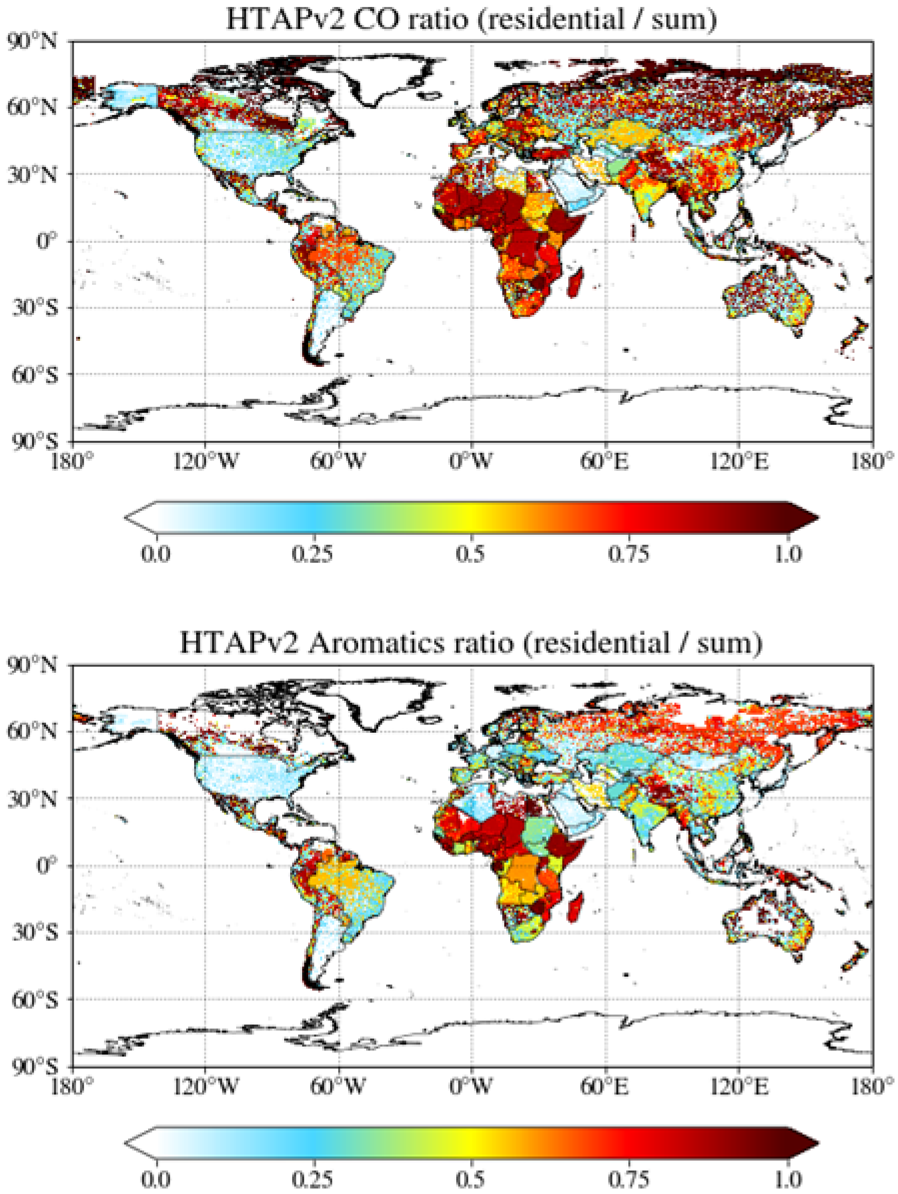


Figure S15. (top) Fractional contribution of CO emissions from residential sources to total emission sources from HTAP. (bottom) Fractional contribution of BTEX emissions from residential sources to total emission sources from HTAP. Residential sources include small-scale combustion, such as heating and cooking, which may include solid-fuel emissions.

1.4. Why does SIMPLE model now rely on BTEX alone? It seems to be doing better overall than when it just relied on CO, but why not use more than one variable with BTEX to develop a multilinear fit for the SOAP emissions? For example, Seoul is likely a problematic point in the SOA vs. BTEX regression (see specific comment #7). It's driving down the ratio and thus probably leading to under-representing impacts in the Northeast US and LA. Taken together, Figs. 3 and 4 suggest that in Seoul there are SOA sources associated with CO emissions that are not as highly associated with BTEX. Using too many independent variables would surely end up overfitting, but why not add 1 or 2 key variables (like CO and POA) since you have a good idea that the relative contributions of sources (e.g. vehicles, VCPs, and solid fuel use) vary from city to city?

As we discuss in response to 1.12, Seoul is not driving the relationship and thus is not a problematic point.

Also, there is perhaps some confusion. The updated version of SIMPLE does not rely on BTEX alone, but rather on both BTEX and CO emissions (e.g., eq. 7 in the ACPD version) as well as OH concentrations within the model. This is an improvement from the original SIMPLE model, in which the parameterization only depended on CO and the model OH fields.

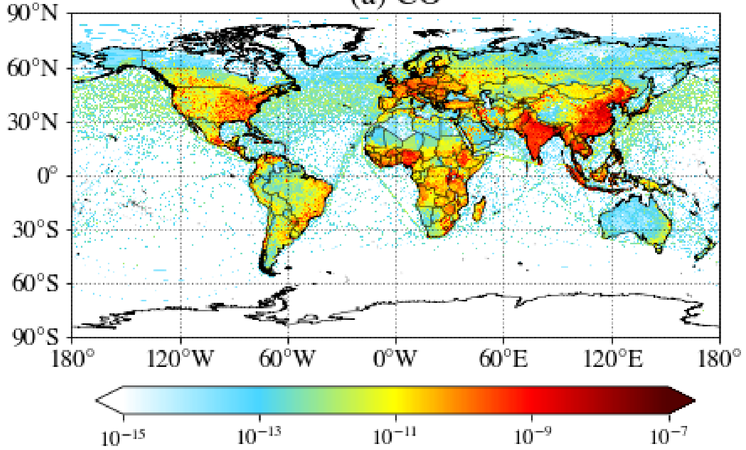
We do not see a reason for a more complex parameterization, since the available data are well-fit with the updated parameterization proposed in the paper. Of course more complex parameterizations could be devised, but they would be underconstrained by the observations. Indeed, Fig. 2 in the ACPD version shows that BTEX is co-emitted in both "traditional" and "non-traditional" sources (fossil fuel versus VCP), and both these sources account for the majority of the predicted ASOA (Fig. 6 of ACPD version). Finally, most emission inventories have BTEX, providing a more straightforward method to implement this parameterization into chemical transport models.

We have added the following text to address this point:

"The $R_{aromatics}/\Delta CO$ allows a dynamic calculation of the E(VOC)/E(CO) = SOA/ΔCO. Hodzic and Jimenez (2011) and Hayes et al. (2015) used a constant value of 0.069 g g^{-1}, which worked well for the two cities investigated, but does not for the expanded dataset studied here. Thus, both the aromatic emissions and CO emissions are used in this study to better represent the variable emissions of ASOA precursors (Fig. S5)."

The following figure has been added to address the comment as well:

(a) CO



(b) BTX (weighted by OH reactivity)

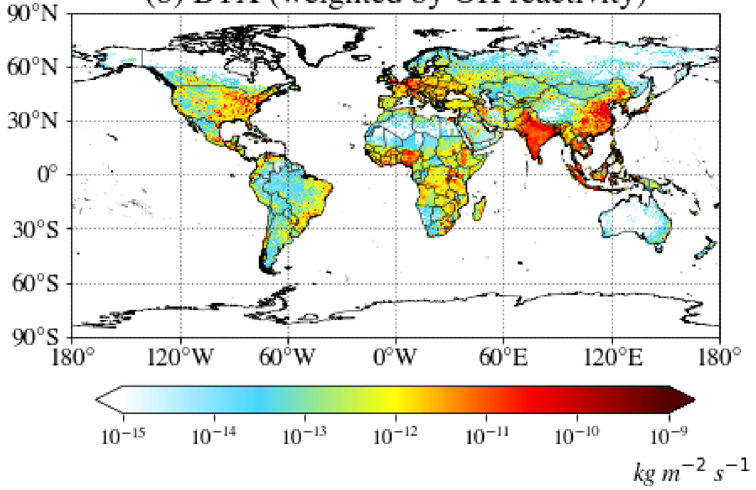


Figure S5. (a) Annually average CO emissions from HTAP. (b) Annually average benzene, toluene, and xylenes (BTX) emissions, weighted by their OH reaction rate

$$(E_{weight} = N\frac{\sum_i E_i k_{OH,i}}{\sum_i k_{OH,i}},\ i = B, T, X\ ;\ \mathbf{N=3}).$$

1.5. Line 455-457: The observation about SVOCs is difficult to believe based on existing NMOG and POA profiles in literature. I have yet to see a volatility profile for any source where the SVOC accounted for half of the total ASOA precursor, let alone 88%. Is there something unexpected going on with the CO normalization of POA vs. VOCs here? I confess this one catches me completely by surprise, and likewise the large influence of SVOCs in Fig. 3 looks strange as well. The authors have made an emphatic case for the dominant and growing role of VCPs. Wouldn't these be overwhelmingly VOCs and IVOCs? Cooking emissions are used to explain this to some degree, but if the Robinson et al. (2007) profile is used for cooking emissions, I would expect a lot more 10^3 and 10^4 C* compounds. Regardless, SVOC should probably be included on Fig. 2 as a separate series like BTEX and IVOC. And I recommend adding more description about how SVOC smission ratios are derived.

There are multiple prior publications, where ASOA formed from SVOCs were accounted for by several tens of percent of the total SOA (e.g., Dzepina et al., 2009; Ma et al., 2017). As stated in that section "Beijing has the highest contribution of SVOCs to ASOA precursors due to the use of solid fuels and cooking emissions (Hu et al., 2016)." Thus these results are not so surprising. They are correct and are based on the detailed inventories reported here.

We have moved Fig. 3 from the main paper to the SI, as it is not a finding but more a tool that was used to estimate ASOA. Part of the reason is that for the emissions with C* greater than 10^6 µg m^{-3} only include the VOCs reported in the SI, as the campaigns used here either had missing oxygenated VOCs and/or the challenge of estimating oxygenated VOC emission ratios (e.g., de Gouw et al., 2018; McDonald et al., 2018). Not including OVOCs may lead to an underestimation of the emission ratios at high volatility.

We have added the following text to the SI to describe how the SVOC emission profile was determined:

"To estimate the SVOC mass concentration in equilibrium with the POA (Table S9) in each bin, the POA mass concentration is first multiplied by the fraction of POA measured in each bin from literature. This yields the concentration of POA for that specific volatility bin. Then the total POA + SVOC concentration for that bin is obtained divided by the amount of material found in the particle phase for that bin for the average temperature (~298 K) and OA mass concentration (~10 µg m^{-3}). Then, the gas-phase SVOC concentration is calculated by multiplying the total concentration by the gas-phase fraction. Thus, e.g., SVOC in the C* = 100 µg m^{-3} bin, ~91% of the SVOC mass will be found in the gas-phase."

Specific Comments:

1.6 Line 95-98: These generalities about IVOCs and SVOCs are perhaps useful for an introduction for those who may be unfamiliar but based on more current understanding of emissions sampling and speciation, they may be more confusing than helpful. Lu et al. (2018) show in their Fig. 1 that most of the IVOC would have missed the filters for the vehicles they studied, but much of the SVOC is expected to be captured by the filters. Even more SVOC would presumably be captured for stationary sources at conditions relevant for "condensable particulate matter" measurements (i.e. low dilution, cooled temperatures); see Morino et al. (2018). As for IVOCs, VBS profiles for biomass burning sources like those in May et al. (2013)

show that IVOCs are probably included in many if not most PM emission factors measured for those sources if the emissions are not diluted enough. The authors here are not focused on wildfires, but certainly cooking/residential wood-burning PM emission factors may include these IVOCs. Admittedly, the problem is even more complicated by the fact that many countries report wood-burning PM emission factors at high temperature conditions, so they may not actually be capturing the IVOCs. Still, it's highly uncertain to what extent they are already measured. I urge the authors to update their discussion of these classes of compounds to better reflect some of the nuances we now understand better.

We have removed that line and have expanded the discussion of S/IVOCs in the introduction to read:

"Many of these prior studies generally investigated AVOC with high volatility, where volatility here is defined as the saturation concentration, C*, in µg m^{-3} (de Gouw et al., 2005; Volkamer et al., 2006; Dzepina et al., 2009; Freney et al., 2014; Woody et al., 2016). More recent studies have identified lower volatility compounds in transportation-related emissions (e.g., Zhao et al., 2014, 2016b; Lu et al., 2018). These compounds have been broadly identified as intermediate-volatile organic compounds (IVOCs) and semi-volatile organic compounds (SVOCs). IVOCs have a C* generally of 10^{3} to 10^{6} µg m^{-3} while SVOCs have a C* generally of 1 to 10^{2} µg m^{-3}. Due to their lower volatility and functional groups, these classes of compounds generally form ASOA more efficiently than traditional, higher volatile AVOCs; however, S/IVOCs have also been more difficult to measure (e.g., Zhao et al., 2014; Pagonis et al., 2017; Deming et al., 2018). IVOCs generally have been the more difficult of the two classes to measure and identify as these compounds cannot be collected onto filters to be sampled off-line (Lu et al., 2018) and generally show up as unresolved complex mixture for in-situ measurements using gas-chromatography (GC) (Zhao et al., 2014). SVOCs, on the other hand, can be more readily collected onto filters and sampled off-line due to their lower volatility (Lu et al., 2018). Another potential issue has been an under-estimation of the S/IVOC aerosol production, as well as an under-estimation in the contribution of photochemically produced S/IVOC from photooxidized "traditional" VOCs, due to partitioning of these low volatile compounds to chamber walls and tubing (Krechmer et al., 2016; Ye et al., 2016; Liu et al., 2019). Accounting for this under-estimation increases the predicted ASOA (Ma et al., 2017). The inclusion of these classes of compounds have led to improvement in some urban SOA budget closure; however, many studies still have indicated a general short-fall in ASOA budget even when including these compounds from transportation-related emissions. (Dzepina et al., 2009; Tsimpidi et al., 2010; Hayes et al., 2015; Cappa et al., 2016; Ma et al., 2017; McDonald et al., 2018)."

1.7 Line 99-118: I encourage the authors to add residential wood burning/cookstoves to their list, and possibly also the recent work on asphalt emissions (Khare et al., 2020).

We have added the following:

“. . . as well as cooking emissions (Hayes et al., 2015), asphalt emissions (Khare et al., 2020), and solid fuel emissions from residential wood burning and/or cookstoves (e.g., Hu et al., 2013, 2020; Schroder et al., 2018). . .”

1.8 Line 119-132: I think the authors get somewhat stuck on the SVOC portion of the ASOA problem in this paragraph and would do well to keep the broad focus on both IVOC-SOA and SVOC-SOA they have been introducing so far. For one thing, I’m not sure how important revising the (terrible) assumption of POA nonvolatility is for connecting urban PM to health impacts in the context of annual mean guidelines. Of course it’s important to know how much of the PM started as an SVOC vapor for the purposes of control. But meanwhile, if we think that a portion of the SOA mass was emitted in the particle phase and then evaporated, oxidized and recondensed after dilution, then how does updating our conceptual picture to consider that portion volatile necessarily help us control it better - we could still control it with particulate filters. To me, the important reasons to update the conceptual model form nonvolatile POA to semivolatile are to 1) better track composition of the OA because maybe it has different toxicity or efficiencies for losses as it is oxidized, 2) sensitivity to temperature and concentration swings might have an impact on urban scale versus suburban or rural exposure or diurnal timing of concentration peaks and thus impacts on human exposure. Adding in the SVOC and IVOC vapors helps us achieve a total mass balance on the amount of carbon with potential to make SOA and this is really a separate point. In short, the authors could make it more clear in this paragraph, at least qualitatively, which sources of uncertainty they are most concerned about in previous estimations of PM mortality. Is it a) poor traditional POA models, b) undersampled SVOC and IVOC emissions from known sources, c) underestimated yields (i.e. vapor wall-losses, etc), d) missing or unacknowledged sources of vapor precursors or e) something else. Right now, it seems like (a) is their chief concern.

We have addressed some of these concerns ((b) and (c)) in response to 1.6. We have softened this section to be less focused on SVOC, and instead the under estimation of SOA most likely due to IVOC and “non-traditional” sources. We have changed it to clarify:

“Due to the uncertainty on the emissions of ASOA precursors and on the amount of ASOA formed from them, the number of premature deaths associated with urban organic emissions is largely unknown. Since numerous studies have shown the importance of VCPs and other non-traditional VOC emission sources, efforts have been made to try to improve the representation and emissions of VCPs (Seltzer et al., 2021), which can reduce the

uncertainty in ASOA precursors and the associated premature deaths estimations. Currently, most studies have not included ASOA realistically (e.g., Lelieveld et al., 2015; Silva et al., 2016; Ridley et al., 2018) in source apportionment of the premature deaths associated with long-term exposure of $PM_{2.5}$. These models represented total OA as non-volatile POA and "traditional" ASOA precursors (transportation-based VOCs), which largely under-predict ASOA (Ensberg et al., 2014; Hayes et al., 2015; Nault et al., 2018; Schroder et al., 2018) while over-predicting POA (e.g., Hodzic et al., 2010; Zhao et al., 2016a; Jathar et al., 2017). This does not reflect the current understanding that POA is volatile and contributes to ASOA mass concentration (e.g., Grieshop et al., 2009; Lu et al., 2018). Though the models are estimating total OA correctly (Ridley et al., 2018; Hodzic et al., 2020; Pai et al., 2020), the attribution of premature deaths to POA instead of SOA formed from "traditional" and "non-traditional" sources, including IVOCs from both sources, could lead to regulations that may not target the emissions that would reduce OA in urban areas. As PM_1 and SOA mass are highest in urban areas (Fig. 1), also shown in Jimenez et al. (2009), it is necessary to quantify the amount and identify the sources of ASOA to target future emission standards that will optimally improve air quality and the associated health impacts. As these emissions are from human activities, they will contribute to SOA mass outside urban regions and to potential health impacts outside urban regions as well."'

1.9 Line 141: A complete introduction or general description of the modeling approach is needed to begin the methods section. Before the authors get into the extreme details (e.g. how data were averaged), we reader would do well to learn what the basic idea of the study is going to be (i.e. parameterize ASOA in cities using campaign data, replace ASOA in GEOS-Chem with these results, plug new PM2.5 into relative risk and premature death parameterizations, assess the impact, and explore some key sensitivities). For example, on line 142, I'm not sure what 'values' are being discussed, how they are measured, or how they will be used.

We have added the following to introduce everything discussed in Sect. 2:

"Here, we introduce the ambient observations from various campaigns used to constrain ASOA production (Sect. 2.1), description of the simplified model used in CTMs to better predict ASOA (Sect. 2.2), and description of how premature mortality was estimated for this study (Sect. 2.3). In the SI, the following can be found: description of the emissions used to calculate the ASOA budget for five different locations (Sect. S1), description of how the ASOA budget was calculated for the five different locations (Sect. S2), description of the CTM (GEOS-Chem) used in this study (Sect. S3 - S4), and error analysis for the observations (Sect. S5)."

1.10 Figures are introduced out of order in the methods section.

We have removed reference to figures in Sect. 3 to instead be references to the sections themselves.

1.11 It looks to me like the emission ratio in Tables S5-S8 that were calculated with Eq. 3 are in most (though not all) cases well outside the range of measured emission ratios from other campaigns. For example, o-xylene in Table S5 is all as high or higher than the maximum observations, propene in Table S7 as well. The values for London in Table S8 are either below the minimum observed or above the maximum, depending on the species. Are these predictions expected by the authors? Can they be explained by variations among cities? I recommend calculating and reporting the performance of the Eq. 3 model in reproducing the observed values in Table S5-S8. Also, what values of t were used to calculate the emission ratios in Eq. 3? I assume many values used and then all averaged together? Or were the values for each daily averaged and then a campaign average derived from that? What is the spread in the intermediate emission ratio values? I think this paragraph (lines 150-161) could be written more clearly to better describe the multiple levels of averaging and error analysis taking place here.

Prior studies have shown very large variability across different cities for the same compound, e.g., Bon et al. (2011) showed an order of magnitude difference in ethane emission ratios across three different studies and a factor of ~20 difference in propane across three different studies. Further, as shown in Bon et al. (2011) and Apel et al. (2010) and highlighted in the table made below, there can be large differences, especially for the alkanes, for the same location, depending on how the emission ratio was determined. Thus, there can be large variability across cities as well as potential uncertainty, which is most prominent for the longer lived compounds that minimally contribute to ASOA production.

We have added the following text in the SI to discuss and clarify this point:

"A further potential source of uncertainty in this analysis is the calculated VOC emission ratios for the studies that did not have ratios published previously (Houston 2000, London, Houston 2013, and Seoul). To investigate how well Eq. 3 does in estimating the VOC emission ratios, a comparison of the estimated VOC emission ratios versus previously published ratios for two different cities, Mexico City (Apel et al., 2010; Bon et al., 2011) and Los Angeles (de Gouw et al., 2017) was made (Table S10). Also, for Mexico City, two locations, an urban and a suburban site, were compared both against each other (Apel et al., 2010; Bon et al., 2011) and the calculated values from Eq. 3.

First, as shown in Table S10, even for the same location (suburban Mexico City), different values in the emission ratio, especially for the alkanes, can be observed, by as much as a factor of 7. This can be partially explained by differences in how the emission ratios were

determined. For both Apel et al. (2010) and Bon et al. (2011), the authors took the slope of VOCs versus CO and used different regression techniques and different time periods. Comparing their technique with ours, we generally estimate VOC emission ratios within 50% of the reported values, and the estimation improves for shorter lived compounds (e.g., aromatics). However, de Gouw et al. (2017) more carefully took chemistry into consideration for any potential losses of the VOCs prior to observation to determine emission ratios, similar to this study. We believe the comparison with de Gouw et al. (2017) provides a more useful comparison in the method presented here. We find, at most, a 30% difference in the emission ratios, with an average difference of 4±15% for all compounds. Thus, from this analysis, we conclude that (1) there is large variability in VOC emission ratios across urban areas around the world, which has been highlighted in other studies (Warneke et al., 2007), and (2) the method that considers losses of VOCs is the more accurate procedure to estimate VOC emissions and leads to the best reproducibility across studies and lowest uncertainty (< 30%, ~4% on average).”

The following table has been added to the SI:

Table S10. Comparison of estimated VOC emission ratios from two studies from Mexico City (Apel et al., 2010; Bon et al., 2011), one study from Los Angeles (de Gouw et al., 2017), and this study.

VOC Ratio	Apel et al. (2010) Downtown MC	This Study	Apel et al. (2010) Suburbs MC	Bon et al. (2011) Outskirt MC	This Study	de Gouw et al. (2017) LA	This Study
Ethane	7.4	8.2	3.0	21.5	8.2	16.5	18.9
Propane	41.5	36.9	49.3	61.7	38.4	13.4	14.0
n-Butane	15.1	14.9	15.3	21.7	14.1	5.0	5.7
i-Butane	4.8	4.8	5.3	7.2	4.9	3.2	3.5
n-Pentane	2.1	2.9	2.1	2.5	2.1	3.4	3.4
i-Pentane	2.7	3.6	3.2	3.3	3.1	8.7	7.8
n-Hexane	1.5	1.9	1.3	1.5	1.2	1.4	1.7
Ethene	8.4	6.1	7.9	7.0	7.1	11.2	9.6
Propene	2.6	1.3	2.9	3.0	1.6	4.1	3.9
Benzene	0.9	1.0	1.2	1.2	1.3	1.3	1.4
Toluene	7.5	9.2	5.2	4.2	4.1	3.4	3.0

Ethylbenzene	**0.9**	**0.8**	**0.4**	**4.3***	**0.4**	**0.6**	**0.6**
m+p-Xylene	**1.1**	**0.7**	**0.5**	**No Data**	**0.4**	**2.1**	**1.9**
o-Xylene	**0.4**	**0.2**	**0.2**	**No Data**	**0.2**	**0.8**	**0.7**
Trimethylbenzenes	**No Data**	**No Data**	**No Data**	**No Data**	**No Data**	**1.6**	**1.1**
Ethyltoluenes	**No Data**	**No Data**	**No Data**	**No Data**	**No Data**	**0.6**	**0.4**
Propylbenzene	**No Data**	**No Data**	**No Data**	**No Data**	**No Data**	**0.1**	**0.1**

*In Bon et al. (2011), they reported the sum of C8 aromatics, which is the sum of ethylbenzene and xylenes

1.12 Lines 181-188: I appreciate the spirit of the leave-one-out sensitivity study and the results presented in Table S10. However, I do not think it accomplishes what the authors intended, which is to justify the regressed slope of 24.8. The reference to 95% confidence intervals seems misleading, perhaps because a clear null hypothesis is not stated. I'm not sure I've seen confidence intervals used to prove two slopes are statistically similar before, but I'd be interested to learn if the authors can show their work. A conventional leave-one-out would calculate the error in predicting the removed point and then average the errors across all trials. I'm not sure how knowing this error statistic would be helpful either though, except to perhaps compare among similar leave-one-out analyses for the other slopes in Fig. 5. In my opinion, a better analysis would involve an assessment of the degree to which the Seoul data point is influencing the slope parameter. For example, the Cook's distance is commonly used in regression approaches to flag highly influential data points. If the point is determined to be influential, then the authors need to discuss what impact the change in slope from 24.8 to 34.0 has on the conclusion of the paper.

The equation we had used to investigate statistical difference in slopes was:

$$t = \frac{b_1 - b_2}{\sqrt{s_{b_1}^2 + s_{b_2}^2}}$$

Where b_i is the slope and s_i is the standard deviation about the slope.

In addition, we have also conducted the Cook's distance test, of which we were not aware. We appreciate the reviewer bringing this statistical tool to our knowledge. We have found that the T-test, Cook's distance test, and the difference in fits test all show that the one point from Seoul is not an outlier. We have added the following table and text to the paper:

Table S11. Statistical analysis of the data used in Fig. 2 to determine if any point is influencing the slope, using the T-test, Cook's Distance test, and Difference in Fits test. For

the T-test, the point is influential if the t value is < 0.05 while for the Cook's Distance and Difference in Fits test, the point is influential if the value is > 1.

Campaign	**T-test**	**Cook's Distance**	**Difference in Fits**
NE US Ship	0.63	0.06	-0.29
NE US Aircraft	0.12	0.27	0.73
Mexico City	0.39	0.06	0.33
Los Angeles	0.32	0.08	0.38
Changdao Island, China	0.41	0.09	-0.38
Beijing	0.42	0.06	-0.32
London	0.31	0.13	-0.48
NYC	0.90	0.00	-0.05
Seoul	0.99	0.00	0.01

We have updated the text to say:

"Statistical analysis for the influence of the data from Seoul on the figure was conducted, including a T-test, Cook's Distance test, and Difference in Fits test (Table S11). All three statistical tests show that the data from Seoul (and all the data in general) is not overly influencing the reported slope."

1.13 The SIMPLE model relies on having an accurate BTEX field for input. So how consistent were the HTAPv2 emission inputs with each of the measurement campaigns, allowing the expected deviations for year to year trends?

We have added the following text in the SI:

"Analysis of the HTAP emissions, compared to other emission inventories, generally showed the highest correlation with observations (R^2 = 0.54), versus the other inventories (CEDS R^2 = 0.26, MACCity R^2 = 0.00, and RETROv2 R^2 = 0.04), leading to the selection of this emission inventory."

1.14 Why not add a supplemental figure showing the average spatial distribution of CO and R_btex emissions so readers can get a sense for which is driving the SIMPLE predictions in the various countries? I recommend at least plotting this as country average, if not both country averages and grid cells.

Please see response to comment 1.4.

1.15 Consider adding to the conclusions the ASOA-associated premature death estimates you are most confident in.

We respectfully disagree on this point, as prior studies have discussed $PM_{2.5}$-associated premature death estimates with less investigation into how well the models predicted the composition of the total $PM_{2.5}$ while still attributing premature deaths to different sources (e.g., Lelieveld et al., 2015; Silva et al., 2016; Ridley et al., 2018).

Minor comments:

1.16 Line 60: Rewrite "anthropogenic reactivity of specific organic compounds" to "reactivity of specific anthropogenic volatile organic compounds"?

Completed.

1.17 Line 66: "results in up to . . ."

Completed.

1.18 Line 67: "extrapolation" of what data specifically? Is it more informative to say "extrapolation from regions where detailed emission inventory data are available to other regions where uncertainties in emissions are larger."?

We have updated the line to say:

"A limitation of this study is the extrapolation from cities with detailed studies and regions where detailed emission inventories are available to other regions where uncertainties in emissions are larger."

1.19 Line 68: I agree that comprehensive air quality campaigns are certainly helpful and possibly necessary, but it seems that robust national-scale institutions (government, academic, or private) are absolutely necessary to accurately catalogue emission factors and activity data to the level required to reduce the uncertainties discussed in this manuscript. Perhaps this sentence could be broadened to something like: "In addition to further development of institutional air quality management infrastructure, comprehensive air quality campaigns . . ."

We have updated the line to say:

“In addition to further development of institutional air quality management infrastructure, . . .”

1.20 Lines 104 - 106: Suggest rewriting: “Biogenic SOA (BSOA) in urban areas typically results from advection of regional background concentrations rather than processing of locally emitted biogenic VOCs.”

Updated to this.

1.21 Lines 116 - 118: Seltzer et al. (2021) is currently finalizing discussion in ACPD and presents a detailed VCP emission inventory for the U.S. Based on this the authors may want to update this sentence to include that step forward, but it’s their choice.

We have added the following to address this:

“Since numerous studies have shown the importance of VCPs and other non-typical VOC emission sources, efforts have been made to try to improve the representation and emissions of VCPs (Seltzer et al., 2021), which can reduce the uncertainty in ASOA precursors and the associated premature deaths estimation.”

1.22 Line 119: “uncertainty on the (burden of -or- emissions of) ASOA precursors. . .”

Updated the text to say:

“. . . uncertainty on the emissions of ASOA precursors . . .”

1.23 Eq. 3: Recommend adding an exp subscript to [OH] here to make it clear that it is calculated from Eq. 2.

Updated.

1.24 Table S10. Please indicate which slope is being shown here (delta_SOA/R_BTEX)

We have removed this Table due to response 1.12 and have replaced it with the table shown there.

1.25 Line 201: Many of the BTEX values are modeled with equation 3 right? Please make this clear.

For the IVOCs used in this analysis, only 1 city had BTEX was calculated with Eq. 3 (London) while the rest of the BTEX are used from studies (NE USA, LA, Beijing, and NYC).

We have added the following to clarify:

"The IVOC:BTEX emission ratio from inventories are multiplied with the observed BTEX, either the reported value from studies (NE US aircraft (Warneke et al., 2007), Los Angeles (de Gouw et al., 2017), Beijing (Wang et al., 2014), and New York City (Warneke et al., 2007)) or estimated from Eq. 3 (London), . . ."

1.26 Line 213: C* range is not consistent with how IVOCs are usually defined.

We have updated the values to $10^3 < C^* < 10^6$ μg m^{-3}.

1.27 Line 209 - 210: Based on the reference, it appears the authors are specifically referring to underestimation of IVOCs in the ambient. Please make that more clear in the sentence.

We have rephrased the sentences to clarify this point:

"Additionally, we rely on inventories for estimating atmospheric abundances of IVOCs because it has been challenging to measure the full range of IVOC precursors that are emitted into urban air due to many of the IVOCs from VCPs being oxygenated VOCs. These compounds are challenging to measure using traditional instrumentation (e.g., gas chromatography-mass spectrometry), leading to potential underestimation of the IVOC emission ratios (Zhao et al., 2014, 2017; Lu et al., 2018)."

1.28 Line 214 - 216: It's unclear to me how the IVOCs and unspeciated SOA precursors relate to each other here. Are the authors saying they used SOA yields from Jather et al. (2014) to define the IVOC SOA yields uniformly for all C* bins? Please clarify. For example, a clearer way of making that point might be, "SOA yields from IVOC oxidation were parameterized with data from n-tridecane for gasoline engines and n-pentadecane for diesel engines (Jathar et al., 2014)."

We have updated to the text to say:

"The ASOA yields and rate constants for IVOC oxidation were parameterized with data from n-tridecane and n-pentadecane for gasoline and diesel emissions, respectively (Jathar

et al., 2014), and for VCPs, the yields and rate constants for IVOC oxidation were parameterized with data from n-tetradecane (McDonald et al., 2018).”

1.29 Lines 216 - 218: Should VOCs be IVOCs here? Again, aren’t all the IVOCs in this study unspeciated? If so, why make the distinction?

We have added the I before the VOCs here. Also, we made the distinction for unspeciated specifically for VCPs as the IVOCs are unspeciated; however, BTEX, which can be in VCPs (Fig. 2), is speciated.

1.30 Line 224: Why was the Huffman et al. (2009) distribution not used for the cooking VBS distribution?

To be consistent with Ma et al. (2017), we used the same profiles as those authors used in their analysis.

We have added the following to clarify:

“These profiles were selected to be consistent with Ma et al. (2017).”

1.31 Table S9: What is the HOA and Other POA mass normalized to? Shouldn’t these also be normalized to CO, or is POA a separate variable in the inventories? POA is never mentioned in the SI in the discussion of the inventory development.

We have updated the table to include the CO term; thus, the units are µg sm^{-3} $ppmv^{-1}$. As described in line 217 - 218, the SVOC emission ratios were estimated relative to the POA mass concentrations.

1.32 SI Line 70: The emission ratios are small, or the range is small?

The range is small and has been updated.

1.33 Lines 250 - 273: Is the TSI parameterization with the Ma parameterization or are they different cases that are explored? It seems like Ma et al. (2017) is used for IVOC SOA yields instead of Jather et al. (2014). There are a lot of parameterizations, precursors classes and products in this model approach. I strongly recommend adding a table(s) explicitly specifying all of the SOA yields and the corresponding precursors used in this study.

No, the TSI parameterization was not used, but the “WOR + ROB + MA” case from Ma et al. (2017). We have added the following table, from Ma et al. (2017), as the compounds used and their rate constants were already included:

Table 13. Parameters for VOC, IVOC, and SVOC aerosol yields. The yields are taken from Ma et al. (2017).

Compound	Stoichiometric SOA yield High-NOx, 298 K (µg m^{-3})				
	0.1	1	10	100	1000
Benzene	N/A	0.276	0.002	0.431	0.202
Toluene					
Ethyltoluene					
Propylbenzenes					
Xylenes	N/A	0.310	0.000	0.420	0.209
Trimethylbenzenes					
IVOC C* = 6	0.007	0.090	0.206	0.350	0.00
IVOC C* = 5	0.0498	0.0814	0.456	0.278	0.00
IVOC C* = 4	0.053	0.103	0.464	0.266	0.00
IVOC C* = 3	0.064	0.0914	0.562	0.209	0.00
HOA C* = 2	N/A	N/A	0.28	N/A	N/A
HOA C* = 1	N/A	0.18	N/A	N/A	N/A
HOA C* = 0	0.12	N/A	N/A	N/A	N/A
COA C* = 2	N/A	N/A	0.1881	N/A	N/A
COA C* = 1	N/A	0.1188	N/A	N/A	N/A
COA C* = 0	0.0594	N/A	N/A	N/A	N/A

We have also updated the rate constant table to include the rate constants for IVOCs and SVOCs.

1.34 Line 273: Recommend rephrasing “increase in mass of 0.99” to “change in mass of 0.99” or “decrease in mass of 1%”.

We have updated the text to say:

“. . . a decrease in mass of 1%. . .”

1.35 Line 276 - 281: This opening sentence is overly dense and meandering. What is the point of the appositive, “for ASOA apportionment (Fig. 1)”? It seems redundant. Should the second “apportionment” be “attribution”? The last portion of the sentence, after the GEOS-Chem reference should be broken off into its own sentence.

We have changed this text to say:

“The model used in this study is GEOS-Chem v12.0.0 (Bey et al., 2001; The International GEOS-Chem User Community, 2018). This model is used for the following calculations: (1) ASOA apportionment (Fig. 1), (2) apportionment of ASOA to total PM2.5 for premature mortality calculations (Sect. 5), and (3) sensitivity analysis for ASOA production and emissions on premature mortality calculations. GEOS-Chem is operated at 2°×2.5° horizontal resolution.”

1.36 Line 335: Recommend presenting Eq. 4 as the summation of premature deaths among all considered causes.

We disagree, as this is how the equation is typically presented in epidemiology papers (e.g., Burnett et al., 2018), and we stated in line 333 that the equation varies according to both the particular disease category and geographic region. The combination of these two dependencies would make writing the summation harder to understand.

Reviewer #2

2.0 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 PM2.5 premature deaths is attempted.

General comment:

2.1 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 processing/oxidation/ageing) as well as its contribution to premature mortality. Not only biomass burning for heating purposes but also forest forest, 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.

The purpose of this paper was to investigate the role of photochemically produced anthropogenic SOA. We provide further discussion and clarification of this point in response to 1.3.

We have also added the following line in the introduction to clarify this point:

"Note, for the rest of the paper, unless explicitly stated otherwise, ASOA refers to SOA produced from the photooxidation of AVOCs, as there are potentially other relevant paths for the production of SOA in urban environments (e.g., Petit et al., 2014; Kodros et al., 2018, 2020; Stavroulas et al., 2019)."

Specific Comments:

2.2 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 (2014) and Athens (2019).

We have added the following:

". . . and solid fuel emissions from residential wood burning and/or cookstoves (e.g., Hu et al., 2013, 2020; Schroder et al., 2018), . . ."

Further, as discussed in R1.3, we further emphasize two studies that did have important impacts from solid fuel emissions. The results for these studies fall within the trend of the photochemically-produced ASOA.

However, we do not have the ability to potentially constrain or include "dark-aging" of bbOA into bbOOA. Thus, as we noted in R1.3, we suggest that the ASOA concentrations in this study may be an underestimate for this reason.

2.3 Line 119 - 132: Isn't the current study also under-predicting ASO by ignoring bbOOA? Furthermore, there is also the additive effect of the different pollutants when considering premature mortality. For example, Kodros et al. (2018) estimate joint exposure from household solid fuel use and ambient PM2.5 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.

The Kodros et al. (2018) study investigated indoor and outdoor exposure; whereas, the purpose of this study is to only investigate the role of exposure to outdoor $PM_{2.5}$. We agree that this source of indoor pollution (among others, e.g., HOMEChem and other references) could be important additional sources of $PM_{2.5}$ exposures, and thus contributors to premature mortality, Kodros et al.

(2018)

acknowledged a large source of uncertainty associated with the indoor estimation of solid-fuel use and thus associated premature mortality.

To clarify this point, we have added the following:

“Though there are potentially other important exposure pathways to PM that may increase premature mortality, such as exposure to solid-fuel emissions indoors (e.g., Kodros et al., 2018), the focus of this paper is on exposure to outdoor ASOA and its associated impacts to premature mortality.”

2.4 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 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?

See response to comment 1.12 concerning a more robust statistical analysis to determine if any one point could be driving the slope or not.

We have updated the text (see R1.3) to reflect that we have two studies (Chinese Outflow 2011 and New York City 2015) that had major impacts of solid fuel emissions (coal combustion for Chinese Outflow and biomass burning from New York City). As discussed in response to 1.12, these two points are not outliers and do not individually overly influence the slope; thus, the update we propose here for the SIMPLE model that is used in GEOS-Chem appears to capture and not underestimate the *photochemical* ASOA production from those sources as well. See response to 1.3 for updated text and clarification on this point.

In regards to the 25% of observed ASOA being explained by BTEX, as stated on pg 18, lines 410 - 413 in the original manuscript:

“However, BTEX alone cannot account for much of the ASOA formation (see budget closure discussion below), and instead, BTEX may be better thought of as both partial contributors and also as indicators for the co-emissions of other (unmeasured) organic precursors that are also efficient at forming ASOA.”

BTEX only explaining 25±6% of the observed ASOA is not shocking, as has been highlighted in prior studies (e.g., Dzepina et al., 2009; Ensberg et al., 2014; Hayes et al., 2015; Ma et al., 2017; Nault et al., 2018), which led to the McDonald et al. (2018) study and the importance for the finding of VCPs potentially explaining a large fraction of the missing *photochemically* produced ASOA. To further clarify this point, we have added the following:

"BTEX only explaining 25% of the observed ASOA is similar to prior studies that have done budget analysis of precursor gases and observed SOA (e.g., Dzepina et al., 2009; Ensberg et al., 2014; Hayes et al., 2015; Ma et al., 2017; Nault et al., 2018)."

As explained throughout Section 3.2 and with Fig. 6, the remaining budget of the observed *photochemically* produced ASOA is explained by other aromatic compounds, IVOCs, and SVOCs. This is the important finding, as we have expanded the work from McDonald et al. (2018) to show the importance of IVOCs and VCPs in the production of *photochemically* produced ASOA. Specifically, we find (pg 21, line 464 of original submission) 85±12% of the observed ASOA for the five different cities to be explained by BTEX, aromatic compounds, IVOCs, and SVOCs.

2.5 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).

As explained in response to comments 1.3 and 2.4, solid fuel combustion is included in both the experimental studies and the model. We do not have a way to include the dark oxidation of BBOA, and we have acknowledged that this may lead to an underestimate of the concentrations and thus the health effects of ASOA (see response to R1.3).

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

As summarized in response comments 1.3, 2.4, and 2.5, this is a misunderstanding. Solid fuel combustion is included. We have updated this text to say:

"Using a model constrained to day-time atmospheric observations (Fig. 2 and Fig. 4, see Sect. 4) leads to more accurate estimation of the contribution of photochemically-produced ASOA to $PM_{2.5}$ associated premature mortality that has not been possible in prior studies."

Technical corrections:

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

Updated:

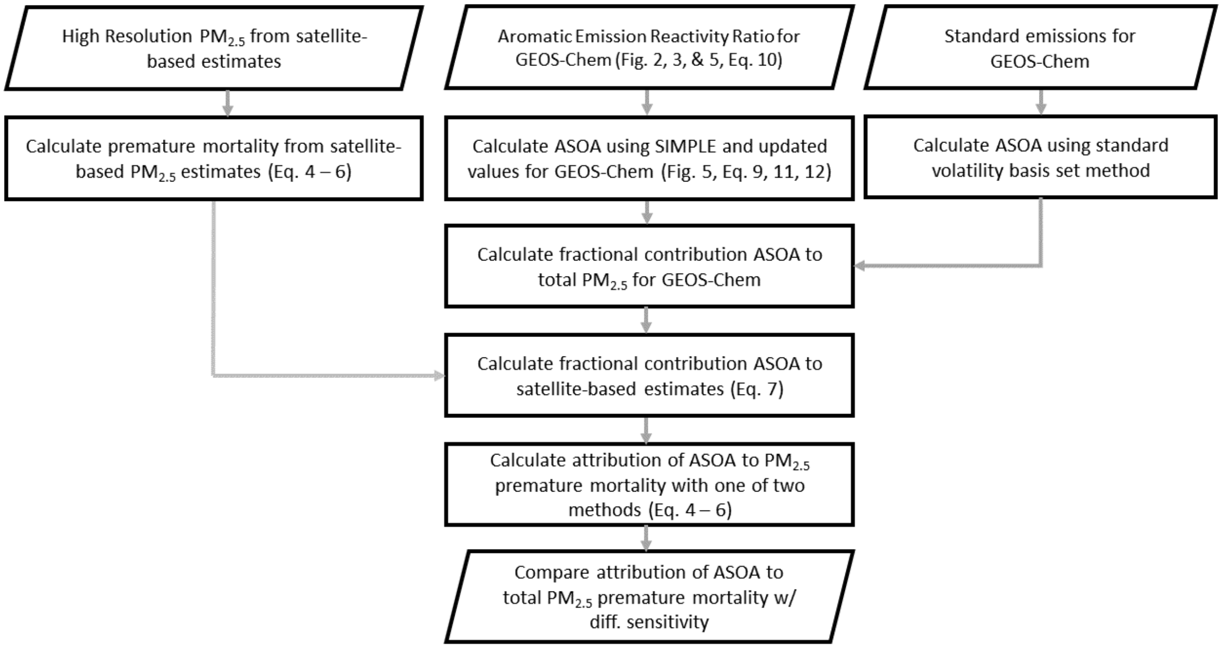


Figure 7. Flowchart describing how observed ASOA production was used to calculate ASOA in GEOS-Chem, and how the satellite-based $PM_{2.5}$ estimates and GEOS-Chem $PM_{2.5}$ speciation was used to estimate the premature mortality and attribution of premature mortality by ASOA. See Sect. 2 for further information about the details in the figure. SIMPLE is described in Eq. 9 and by Hodzic and Jimenez (2011) and Hayes et al. (2015). The one of two methods mentioned include either the Integrated Exposure-Response (IER) (Burnett et al., 2014) with Global Burden of Disease (GBD) dataset (IHME, 2016) or the new Global Exposure Mortality Model (GEMM) (Burnett et al., 2018) methods. For both IER and GEMM, the marginal method (Silva et al., 2016) or attributable fraction method (Anenberg et al., 2019) are used.

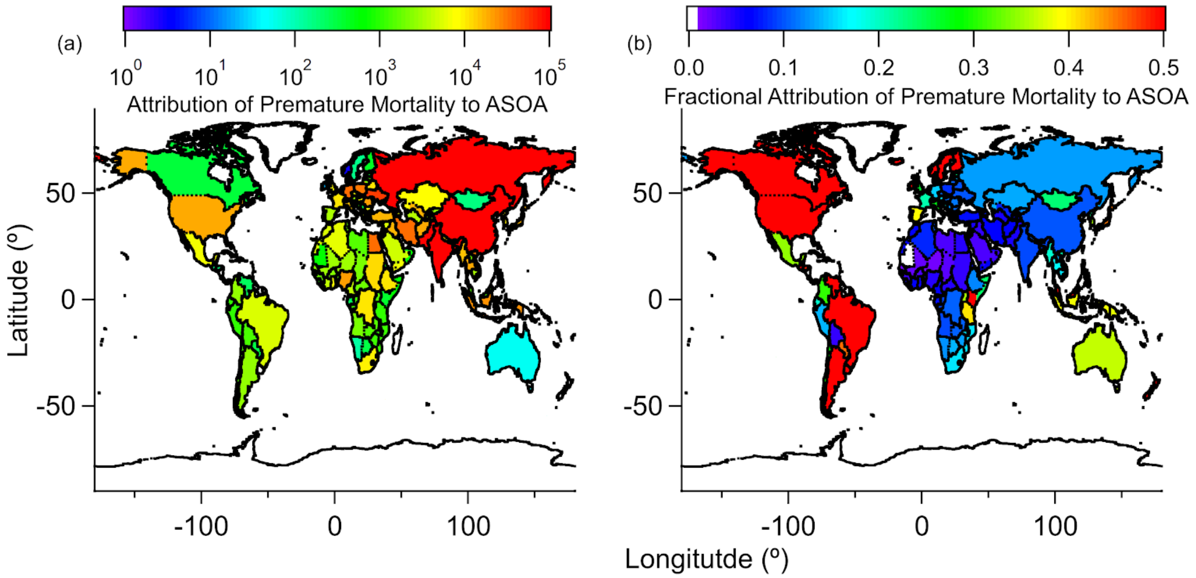


Figure 8. Five-year average (a) estimated reduction in $PM_{2.5}$-associated premature deaths, by country, upon removing ASOA from total $PM_{2.5}$, and (b) fractional reduction (reduction $PM_{2.5}$ premature deaths / total $PM_{2.5}$ premature deaths) in $PM_{2.5}$-associated premature deaths, by country, upon removing ASOA from GEOS-Chem. The IER methods are used here. See Fig. S9 and Fig. S12 for results using GEMM. See Fig. S10 for 10×10 km^2 area results in comparison with country-level results.

We also noticed an error in the labels in Fig. 6 and have updated and include the update below:

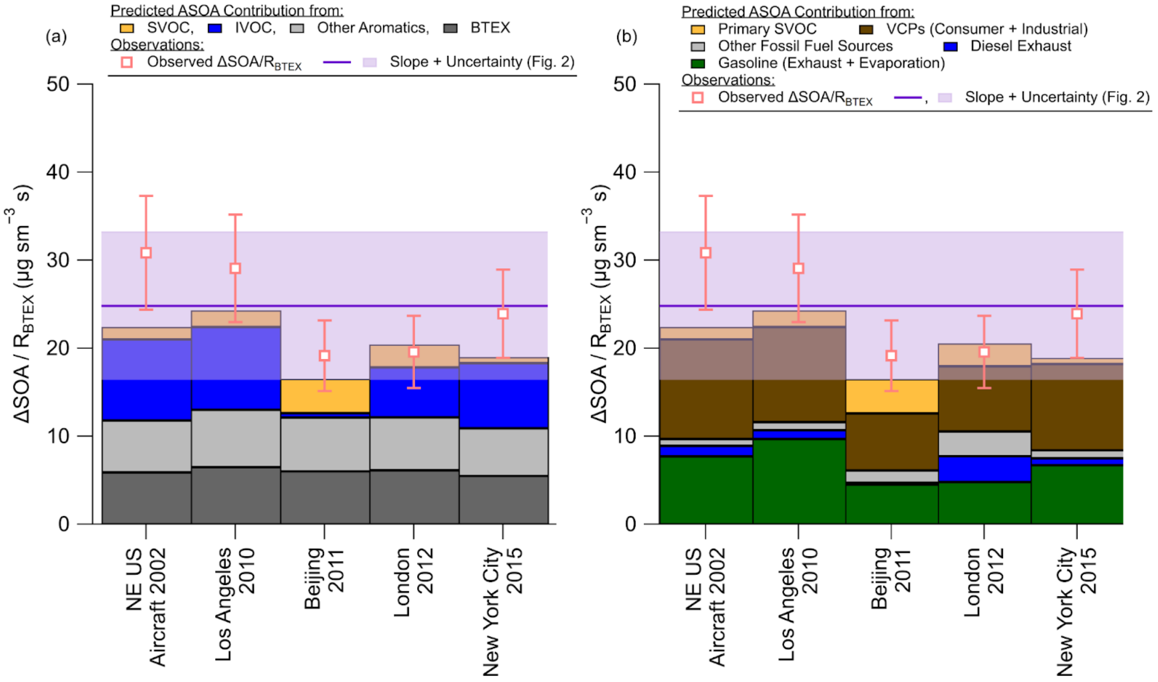


Figure 4. (a) Budget analysis for the contribution of the observed ΔSOA/R_{BTEX} (Fig. 25) for cities with known emissions inventories for different volatility classes (see SI and Fig. 2 and Fig. S6). (b) Same as (a), but for sources of emissions. For (a) and (b), SVOC is the contribution from both vehicle and other (cooking, etc.) sources. See Sect. 2 and SI for information about the emissions, ASOA precursor contribution, error analysis, and discussion about sensitivity of emission inventory IVOC/BTEX ratios for different cities and years in the US.

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