Authors' Response to Anonymous Referee #2

We thank the referee for their thoughtful comments. Our responses are outlined below. We hope that the manuscript is judged to be sufficiently improved to allow its publication.

In this manuscript, the authors studied secondary organic aerosol (SOA) production in urban and forest outflows using the explicit gas-phase chemical mechanism generator GECKO-A. Their simulations show several-fold increases in SOA mass continuing for several days in the urban outflow while the SOA mass increase in the forest outflow is more modest (_50 %) and of shorter duration (1–2 days). The GECKO-A model treats millions of reactions and species. While the present work relies on box model simulations with many assumptions and approximations, the detailed simulations provide useful insights about the SOA production from multi-day multi-generational reactions. For example, it was shown that the production in the urban outflow can be attributed to multi-generational reaction products of both aromatics and alkanes. This work emphasizes the importance of considering these anthropogenic-origin SOA in regional and global aerosol models. The content of this manuscript is suitable for ACP and the manuscript is well written. I can recommend the publication of this manuscript in ACP after the following comments are properly addressed.

1. The explicit GECKO-A model is computationally very expensive. Based on the detailed information from GECKO-A simulations, can the authors provide some simplified schemes that can be used in regional or global models? In Section 4, the authors briefly mentioned that only a few species classes contribute a large proportion of the predicted mass production. It will be very helpful if specific and practical suggestions can be provided on how to represent such processes in 3-D models.

- We agree that it would be highly desirable to produce simplified schemes. However that is a large task, made more complex by the probability that simplified schemes would need to vary based on the specific precursor mixes at different locations. Also, the precise chemical details of our predicted aerosol are likely influenced by several assumptions about the precursor mix and the ensuing chemistry. We have suggested specific molecules as targets for observational studies. Detection of these molecules would increase confidence in the chemical details of our results, and thus in any reduced scheme we might propose in future work.

2. The production of SOA in urban plumes has been studied in LT11. The additional formation of SOA during the urban plume dilution, which has already been pointed out in LT11, is a main focus of present study. In page 18004, the authors mentioned the modifications made to LT11. What are major new findings of the present study (compared to LT11)?

- Major new findings are: 1) multiday aerosol production is not an artifact of the M&Y vapor pressure scheme used in LT-11, as it is also seen under the less aggressive Nannoolal parameterization; 2) multiday production is robust for this urban precursor mixture as the production is shown to occur under a variety of environmental conditions; and 3) by contrast, aerosol mass production is predicted to cease after only 1-2 days for a gas-phase-only chemical mechanism operating on biogenic precursors. We already stated findings 2) and 3) in the Abstract, and have now amended the Abstract as follows to make finding 1) clearer:

"The results show that the predicted production is a robust feature of our model even under changing atmospheric conditions and different vapor pressure schemes…"

3. How were the photochemistry and diurnal variations of key oxidants represented in the box model simulations? How sensitive are the results to these? It will be useful it the authors can provide a figure showing the values and variations of key oxidants.

- We have added a new section (new Section 3.1, and re-numbering subsequent sections) discussing the oxidant concentrations, and included a Supplementary Figure showing oxidant concentrations:

"**3.1 Photochemical Environment**

The concentrations of key oxidants simulated within our urban scenario source region have similar profiles to those shown in Figure 3 of LT-11 for Mexico City. (Oxidants are plotted in Figure S.I.1) Peak urban source region concentrations are: [OH] = 3.2×10^6 molec cm^{-3}, [O_3] = 116 ppbv, and [NOx] = 260 ppbv. These values represent highly-polluted urban conditions, where [OH] is suppressed by high [NOx], and are within the range of observations (Dusanter et al, 2009). In the outflow, [OH] increases until stabilizing on day 5 at ~8.5×10^{6}molec cm^{-3}. Meanwhile, [NOx] drops rapidly to <0.8 ppbv, and O_3 also declines in response to dilution, to ~60ppbv. The forest case shows oxidant concentrations towards the high end of remote observations (e.g. Wolfe et al, 2014): In the forest outflow [OH] is fairly constant at ~ 8×10^{6}molec cm^{-3}, [O3] decreases from 62 to 50 ppbv and NOx falls to consistently low values (~0.2ppbv).

The reduced-dilution sensitivity runs demonstrate that net O_3 production continues in the outflow, even as its base case concentrations decrease. In case NODIL, O_3 concentrations increase, weakly in the forest scenario to ~70 ppbv on day 4, and strongly in the urban scenario to ~175 ppbv on day 5. NODIL NOx is roughly double base case values in both scenarios, which raises forest [OH] levels slightly (to 9×10^{6}molec cm^{-3}) but suppresses urban-outflow [OH] to daily maxima of only ~1.3×10^{6}molec cm^{-3}. Case SLOWDIL produces [O_3] and [NOx] levels intermediate between the base and NODIL values, and [OH] similar to NODIL in the urban scenario and similar to base values in the forest scenario. In sensitivity case HV+, urban scenario [OH] is doubled, [O_3] is 50% higher, and afternoon [NOx] 50% lower relative to the base case, while the forest scenario has 30% higher [OH], but largely unaffected [O_3] and [NOx]. The urban scenario enhancements continue to high, but not unprecedented (Rohrer et al, 2014) peak values of ~17 $\times10^{6}$molec cm^{-3}, indicating that case HV+ provides a good test of the effects on particle mass formation of accelerated gas-phase photochemistry. Sensitivity studies T+10K and SEED/2 have little or no effect on oxidant outflow concentrations."

4. It would be helpful if the authors could provide time series (either in tables or figures) of dilution corrected gas and particle phase carbon mass as well as net loss for species of different carbon numbers or ranges of carbon numbers (4-9, 10-15, 16-22, 23-30, etc.).

- We have added a Table (new Table 2) containing the daily breakdown of carbon mass reservoirs and made reference to it in the text: "Figure 6 and Table 2 illustrate the temporal development of gas-particle partitioning for the urban case."

"Table 2. Carbon partitioning budget timeseries for the urban outflow simulation. Values are assessed at midnight on the days indicated. Losses are assessed relative to 4pm on day 1. Values < 100 are rounded to either two significant figures or two decimal places."

day	Particle phase carbon (mgC initial m^{-3})				Gas phase carbon (mgC initial m^{-3})				Net carbon loss (mgC initial m^{-3})			
	C4-C9	C10-C15	C16-C21	C22-C30	C4-C9	C10-C15	C16-C21	C22-C30	C4-C9	C10-C15	C16-C21	C22-C30
1	0.88	0.61	219	1.8	171	7.3	2.1	0.29	4.0	0.12	0.04	0.01
2	1.9	1.6	2.7	2.0	146	5.5	1.0	0.14	27	1.0	0.25	0.01
3	2.4	2.5	3.1	2.0	122	3.5	0.48	0.07	52	2.1	0.47	0.02
4	3.3	3.2	3.2	2.1	95	1.4	0.17	0.03	80	3.4	0.67	0.03
5	4.0	3.5	3.1	2.1	65	0.44	0.07	0.02	107	4.2	0.81	0.04
6	4.3	3.4	3.1	2.1	47	0.19	0.05	0.01	125	4.5	0.90	0.05
7	4.5	3.3	3.0	2.1	36	0.11	0.04	0.01	136	4.6	0.96	0.06

5. Page 18005, lines 1-5. These simplifications may be far from what happen in the real atmosphere. The authors should discuss the potential impacts of these approximations to the conclusions.

- We already mention most of these points either in the Discussion or in LT11, which we refer to in numerous places. The kinetic limitation point is most important in nucleation situations, which do not apply here. We intend in future studies to collaborate with complementary models thus allowing investigation of more realistic situations.

6. Page 18008, line 3. Why do not use a more realistic diurnally variable temperature?

- Our study in LT-11 already used a diurnally-varying temperature. The current study was intended to explore the sensitivity of the simulated OA production rate to several factors including temperature. Simulations with fixed temperature were chosen to avoid ambiguity about the relative influences of temperature and solar irradiance (which is always diurnally varying in these simulations).

7. Page 18011, line 1. It appears from Fig 2b “20%” should be ~ “30%”.

- We have re-evaluated the differences and find that they remain <25% relative to the reference case. We have amended the text accordingly.