Atmos. Chem. Phys. Discuss., 15, C8244–C8247, 2015
www.atmos-chem-phys-discuss.net/15/C8244/2015/

Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Impact of chamber wall loss of gaseous organic compounds on secondary organic aerosol formation: explicit modeling of SOA formation from alkane and alkene oxidation" *by* Y. S. La et al.

Anonymous Referee #2

Received and published: 20 October 2015

General:

This manuscript describes the effect of vapor phase wall loss on alkane and alkene SOA yields obtained during chamber experiments. Hydrocarbon oxidation, vapor-wall interactions, and SOA formation are modeled using the GECKO-A modeling tool, and the generated SOA yields are compared with previously published data. Generally, vapor-phase wall loss is required for the simulated results to match those observed. For the SOA precursors studied in the manuscript, SOA yields were underestimated

Full Screen / Esc
Printer-friendly Version
Interactive Discussion
Discussion Paper

up to 0.35 fractional yield units if vapor-phase wall loss was not taken into account. This manuscript presents new and interesting results that are within the scope of ACP. I recommend the manuscript for publication after my comments have been addressed.

Major comments:

The Introduction section contains a thorough discussion of gas-particle and vapor-wall partitioning as equilibrium processes following Raoult's law. Figure 2 shows that the distribution of species at 1 h of photooxidation differs greatly from that at equilibrium (photooxidation duration not specified, but presumably longer than 1 h). It is also noted that at 1 h of photooxidation, partitioning is under kinetic, not thermodynamic control. The model is compared with photooxidation experiments of 1-h in duration, but equilibrium behavior is not discussed beyond the Introduction section. Discussing equilibrium behavior in the Introduction section is irrelevant if this behavior is not addressed later in the manuscript. If the authors choose to leave discussion of equilibrium behavior in the manuscript, then how would the presence of semisolid SOA affect equilibrium partitioning of modeled species? Also, what photooxidation duration is required to reach equilibrium, and how does this duration compare with the amount of time ambient particles remain in the atmosphere?

Simulation behavior is tested against experiment results using ΔHC values which only represent data taken at the beginning and end of experiments. This neglects many processes occurring during photooxidation. How does time-dependent modeled hydrocarbon decay match with experimental results? Are NO_x and O_3 experimental data available, and if so, how do they match with model predictions? NO_x and O_3 data would be especially important when addressing the fate of DHF.

Use of the GECKO-A modeling framework allows the authors to study wall uptake by species category and oxidation generation. The discussion of wall uptake by species uptake in Section 4.2 is very nice. It would also be informative to discuss the effect on SOA yield of wall uptake for different generation photooxidation products by comparing the simulated wall and no wall cases. From Figure 8, it appears that during dodecane photooxidation there is substantial wall loss of first-generation photooxidation products; whereas, for hexadecane, first-generation photooxidation products are less affected by wall loss. For which compound is there a greater difference between modeled wall and no wall cases?

Additional comments:

p. 23897, line 9. Are values for $C_w/(M_w\gamma_w)$ chamber-dependent? How do the values of τ_{gw} used in this manuscript compare with those calculated using the wall deposition rates proposed by Zhang et al. (ACP, 2015)?

p. 23900, line 11. How did the mass of DOS seed aerosol compare with the effective chamber wall mass?

p. 23903, line 2. Why was a SOA density of 1 used?

p. 23904, line 26. In the experiments studied, only a fraction of the initial hydrocarbon was reacted during the experiments. In this case, does the fact that the parent alkanes/alkenes partition to the walls before the start of experiments affect the SOA yields (i.e., is this hydrocarbon coming off the walls during the experiment leading to an incorrect ΔHC value)?

p. 2390, Section 4.1. Do experimental and modeled yields take into account particle wall loss, and if so, how is gas-particle partitioning to deposited particles treated?

p. 23922, Figure 2. The time evolution component of this figure is unclear from the figure and caption. The plots should be labeled with the simulation duration, or this should be stated in the figure caption.

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper