Influence of vapor wall-loss in laboratory chambers on yields of secondary organic aerosol

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Secondary organic aerosol (SOA) constitutes a major fraction of sub-micrometer atmospheric particulate matter. Quantitative simulation of SOA within air quality and climate models—and its resulting impacts—depends on the translation of SOA formation observed in laboratory chambers into robust parameterizations. Worldwide data have been accumulating indicating that model predictions of SOA are substantially lower than ambient observations. While possible explanations for this mismatch have been advanced, none has addressed the laboratory chamber data themselves. Losses of particles to the walls of chambers are routinely accounted for, but there has been little evaluation of the effects on SOA formation of losses of semi-volatile vapors to chamber walls. Here, we demonstrate through comparison between observations of SOA formation from toluene photooxidation experiments conducted at different seed particle surface area concentrations and a tunable model of SOA formation, the Statistical Oxidation Model (SOM) that such vapor losses can lead to substantially underestimated SOA formation, by factors as much as 4. Further, we find that SOA growth can only be explained in these 18 hour experiments if the mass accomodation coefficient for uptake of vapors is much smaller than unity, $O(10^{-3})$. Vapor wall-loss is also shown to be an important factor in understanding SOA formation from a broad suite of historical experiments, although the exact influence is found to depend on the particular system under consideration (e.g. VOC identity, NO_x level). Accounting for such vapor wall-losses has the clear potential to bring model predictions and observations of organic aerosol levels into much closer agreement.