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Title: Evaluation of New and Proposed Organic Aerosol Sources and Mechanisms using the Aerosol Modeling Testbed: MILAGRO, CARES, CalNex, BEACHON, and GVAX

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Abstract: This work investigated the formation and evolution of organic aerosols (OA) arising from anthropogenic and biogenic sources in a framework that combined state-of-the-science process and regional modeling, and their evaluation against advanced and emerging field measurements. Although OA are the dominant constituents of submicron particles, our understanding of their atmospheric lifecycle is limited, and current models fail to describe the observed amounts and properties of chemically formed secondary organic aerosols (SOA), leaving large uncertainties on the effects of SOA on climate. Our work has provided novel modeling constraints on sources, formation, aging and removal of SOA by investigating in particular (i) the contribution of trash burning emissions to OA levels in a megacity, (ii) the contribution of glyoxal to SOA formation in aqueous particles in California during CARES/CalNex and over the continental U.S., (iii) SOA formation and regional growth over a pine forest in Colorado and its sensitivity to anthropogenic NOx levels during BEACHON, and the sensitivity of SOA to (iv) the sunlight exposure during its atmospheric lifetime, and to (v) changes in solubility and removal of organic vapors in the urban plume (MILAGRO, Mexico City), and over the continental U.S.. We have also developed a parameterization of water solubility for condensable organic gases produced from major anthropogenic and biogenic precursors based on explicit chemical modeling, and made it available to the wider community. This work used for the first time constraints from the explicit model GECKO-A to improve SOA representation in 3D regional models such as WRF-Chem.

Summary of the results:

Our DOE project focused on five science topics:

(i) Contribution of trash burning emissions to OA levels in a megacity (Mexico City),

(ii) Contribution of glyoxal to SOA formation in aqueous particles in California during CARES/CalNex and over the continental U.S.,

(iii) SOA formation and regional growth over a pine forest in Colorado and its sensitivity to anthropogenic NOx levels during BEACHON,

(iv) Sensitivity of SOA to the sunlight exposure during its lifetime in the troposphere,

(v) Sensitivity of SOA to changes in solubility and removal of organic vapors both in the urban plume (MILAGRO, Mexico City), and over the continental U.S..

Our work also contributed technical improvements to the WRF-Chem model as several formation pathways, and a solubility parameterization were included and evaluated within this model. Christoph Knote worked on this project as a post-doc fellow for two years on regional modeling with WRF-Chem, and Julia Lee-Taylor was employed on this project for 3 months to help the PI with GECKO-A.

These activities have led so far to six publications led by the PI / postdoc and one collaborative publication acknowledging DOE, and one paper is currently in preparation:

1. Hodzic A., Wiedinmyer C., Salcedo D., Jimenez J.L., Impact of Trash Burning on Air Quality in Mexico City, Environ. Sci. Technol., 46, 4950-4957, 2012.

2. Knote, C., Hodzic, A., Jimenez, J. L., Volkamer, R., Orlando, J. J., Baidar, S., Brioude, J., Fast, J., Gentner, D. R., Goldstein, A. H., Hayes, P. L., Knighton, W. B., Oetjen, H., Setyan, A., Stark, H., Thalman, R., Tyndall, G., Washenfelder, R., Waxman, E., and Zhang, Q., Simulation of semi-explicit mechanisms of SOA formation from glyoxal in aerosol in a 3-D model, Atmos. Chem. Phys., 14, 6213-6239, doi:10.5194/acp-14-6213-2014, 2014.

3. Hodzic A., Madronich S., Aumont B., Lee-Taylor J., Karl T., Camredon M. and Mouchel-Vallon C., Limited influence of dry deposition of semi-volatile organic vapors on secondary organic aerosol formation in the urban plume, Geophys. Res. Lett., 40, 3302–3307, doi:10.1002/grl.50611, 2013.

4. Hodzic A., Aumont B., Knote C., et al., Volatility Dependence of Henry's Law Constants Of Condensable Organics: Application to Estimate Depositional Loss of Secondary Organic Aerosols, Geophys. Res. Lett., 41, 13, 4795-4804, doi: 10.1002/2014GL060649, 2014.

5. Knote, C., Hodzic, A., and Jimenez, J. L., The effect of dry and wet deposition of condensable vapors on secondary organic aerosols concentrations over the continental U.S., Atmos. Chem. Phys., 15, 1-18, 2015.

6. Lee-Taylor, J., Hodzic, A., Madronich, S., Aumont, B., Camredon, M., and Valorso, R., Multiday production of condensing organic aerosol mass in urban and forest outflow, Atmos. Chem. Phys., 15, 595-615, 2015.

7. Hodzic, A., Madronich, S., Kasibhatla, P.S., Tyndall, G., Aumont, B., Jimenez, J.L., Lee-Taylor, J., and J. Orlando, Organic photolysis reactions in tropospheric aerosols: effect on secondary organic aerosol formation and lifetime, Atmos. Chem. Phys. Discuss., 15, 8113-8149, 2015.

We also contributed 3 highlights to the DOE ASR website:

- http://www.arm.gov/science/highlights/RNTM2/view
- http://www.arm.gov/science/highlights/R00534/pdf
- http://www.arm.gov/science/highlights/RNTYx/view

We summarize below the key results of each study:

(i) Quantifying the contribution of the trash burning emissions to OA levels in a megacity (Hodzic et al., ES&T, 2012):

In addition to large uncertainties in organic aerosol processes, unaccounted emission sources contribute to OA underestimation in current models. Trash burning is a common disposal method in poor areas that is uncontrolled in many emerging countries, and its contribution to air pollution and climate forcing is unclear due to large uncertainties in its emissions. As part of this project we have built a trash burning emission inventory for Mexico City based on inverse socioeconomic levels and recently measured emission factors, and applied a chemistry-transport model to analyze the effects of these emissions on pollutant concentrations. Trash burning was estimated to emit a similar amount of primary OA as the fossil fuel emissions in Mexico City, and lead to an increase in average OA concentrations up to 2 $\mu g/m^3$ in highly populated suburbs of Mexico City. The study suggests that the mitigation of trash burning could reduce the levels of organic aerosols by 2-40% and those of $PM_{2.5}$ by 1-15% over the metropolitan area (see Figure 1). The impact on other gaseous and secondary inorganic aerosol pollutants was more limited (changes less than 3%). The findings of this study are of great importance for developing future emission control strategies in developing countries where this type of emissions is abundant and uncontrolled.

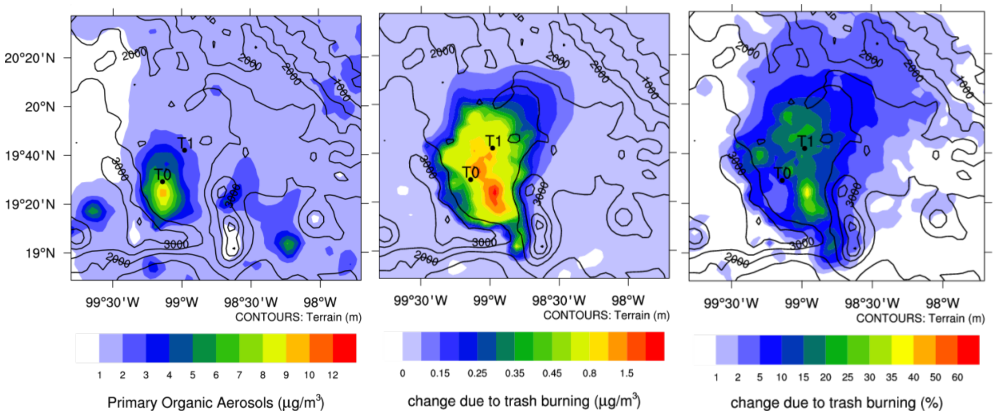

Figure 1: Surface primary organic aerosol (POA) concentrations predicted by WRF-Chem in the vicinity of Mexico City between 20 and 29 March 2006 (left), and increase in POA due to trash burning emissions (middle). Percentage increase in total organic mass due to trash burning emissions is also shown (right). Adapted from Hodzic et al., 2012.

(ii) Simulation of semi-explicit mechanisms of SOA formation from glyoxal in aqueous particles in WRF-Chem (Knote et al., ACP, 2014) over California and U.S.:

Glyoxal was postulated recently as one of the important SOA precursors. It has both anthropogenic and biogenic sources, and readily partitions into the aqueous-phase of cloud droplets and also deliquesce aerosols. In this work we have updated WRF-chem to include detailed glyoxal gas-phase chemistry as well as a module describing its partitioning and reactions in the aqueous-phase of aerosols. A comparison of several proposed mechanisms allowed quantifying the relative importance of different formation pathways and their regional variability. The CARES/CalNex campaigns over California in summer 2010 were used as case study to evaluate the model against observations. In all simulations the LA basin was found to be the hotspot for SOA formation from glyoxal, which contributes between 0.15% and 15% of the modeled SOA depending on the mechanism used. Our results indicated that a mechanism based on a simple uptake coefficient, as it has been previously employed in global modeling studies, leads to higher SOA contributions from glyoxal compared to a more detailed description that considers aerosol phase state and chemical composition. We find that this is not due to restricting glyoxal SOA formation to deliquesce aerosols, but rather due to a kinetic limitation in the current formulation of the reversible pathways. If this limitation is removed, volume pathways contribute >70% of glyoxal SOA mass, and the total mass formed is comparable to the simple uptake coefficient formulation without consideration of aerosol phase state and composition. The improved WRF-Chem model now allows identifying hotspots in other regions (see Figure 2), and therefore could provide guidance in selecting promising locations for future field campaigns, and help analyze future field campaigns (e.g. TCAP, GoAMAZON, SOAS). This updated code was also made available to the community within the latest release of the WRF-Chem model V3.6.1 coupled with the MOSAIC aerosol scheme.

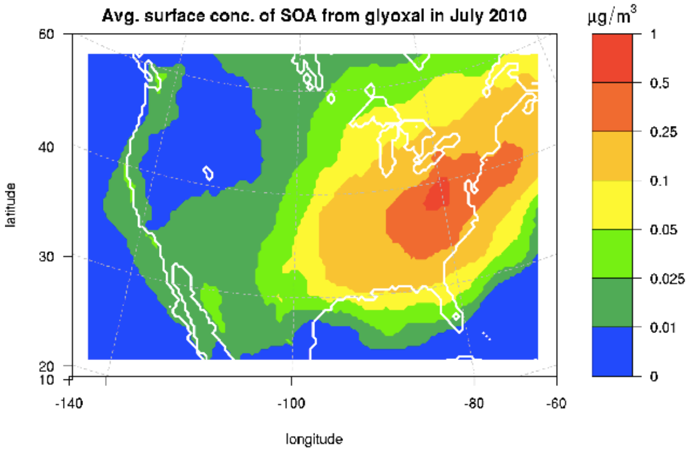

Figure 2: Average surface level concentrations of glyoxal SOA in July 2010. From Knote et al., 2014.

(iii) SOA formation and regional growth over a pine forest in Colorado and its sensitivity to anthropogenic NOx levels during BEACHON

The multi-day production of SOA cannot be well characterized from short laboratory experiments and derived yields. In this project we have applied the explicit chemical model GECKO-A to investigate how SOA is produced over a terpene-dominated forest as well as downwind of an urban area, and gain insights into SOA formation and ageing downwind of these sources.

- ***SOA production downwind of a forest (Lee-Taylor, Hodzic et al., 2015; Hodzic et al., in prep):*** We have been investigating how SOA grows downwind of a forest site using GECKO-A, and are finding that the gas-phase oxidative chemistry is responsible for ~50% increase of the initial SOA mass after the plume has been aged for 3 days. We have tested the sensitivity of our results to different vapor pressure schemes, to the amount of aerosols, temperature and dilution of the plume, and found that this growth is still happening in the model. When the plume gets diluted organic vapors have the tendency to evaporate, however due to the further oxidation in the gas-phase they become progressively less volatile and partition back into the particle-phase. This increase in biogenic environment is occurring within the first day, and is much more moderate (~1.5 times) than downwind of a megacity (5 times). There are currently no available measurements that can allow verifying if this multiday growth predicted by GECKO-A is realistic. We have been comparing GECKO-A results with the PAM (Potential Aerosol Mass, see Figure 3) flow tube and currently analyzing the differences between the model predicted growth and the PAM simulations that are done at 100-1000 times larger OH levels than observed in the atmosphere.

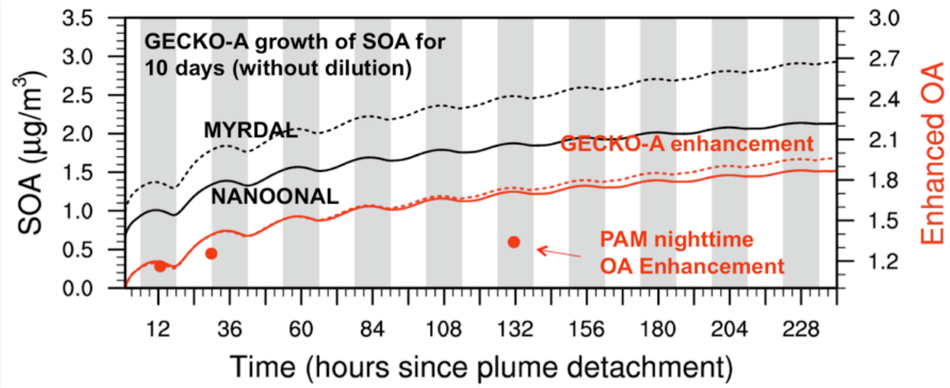

Figure 3: GECKO-A simulated SOA concentrations (black curves) and OA mass enhancements (red curves) as a function of estimated equivalent atmospheric age. PAM measurements are shown by red dots. The airmass introduced into the PAM instrument or model simulator for ageing corresponds to ambient OA levels during the BEACHON-RoMBAS field study. Results of two vapor pressure schemes are compared, Myrdal and Yalkowsky (1997) (dashed curves) and Nanoonal et al. (2008) (solid curves).

The PI has contributed our simulations for biogenic mixtures to the collaborative study targeting the SOA growth (Lee-Taylor, Hodzic et al., ACP 2015). We are also working on preparing another publication to present GECKO-A results and their sensitivity to NOx levels for the BEACHON site (see below).

- ***Anthropogenic-biogenic interactions: Influence of anthropogenic NOx on SOA formation (Hodzic et al., in prep):*** Understanding the effect of NOx on SOA formation is key to understanding perturbations due to anthropogenic activities to SOA. We have run both WRF-Chem and GECKO-A during the BEACHON-RoMBAS-2011 field study to investigate the effect of 50% reductions in anthropogenic NOx on SOA levels measured and predicted at the BEACHON site as well as predicted regionally over Colorado. The effect of NOx on SOA formation is accounted through NOx-dependent yields in WRF-Chem, whereas it is explicitly calculated in GECKO-A through the formation of more or less volatile organic oxidation products and changes in OH, O_3, and NO_3 levels. The results suggest a small reduction in SOA levels up to 10% mainly due to lower levels of NO_3 and less SOA formation through NO_3 nighttime chemistry. WRF-chem showed greater sensitivity to NOx changes than the box model. The results are being currently analyzed and prepared for publication.

- ***SOA production downwind of Mexico City (Hodzic et al., GRL, 2013):*** We also studied how SOA forms downwind of an urban area and what precursor species are responsible for that growth. We find that in Mexico City ~90% of SOA may be explained by long-chain (C>12) alkanes, while ~10% is from the light hydrocarbons (C<13) typically used in 3D models. However, as shown in Figure 4 the contribution of light hydrocarbons to the SOA budget approaches that of the long-chain alkanes at the regional scale after 2-5 days of chemical processing, and is responsible for the SOA regional growth. This result suggests that considering only first generation products from anthropogenic precursors, as often done in 3D models, is insufficient to predict the anthropogenic contribution to regional and global SOA (Hodzic et al., 2013 GRL). We are currently comparing the growth as predicted by the volatility basis set (VBS) typically used in 3D models with GECKO-A predicted SOA growth.

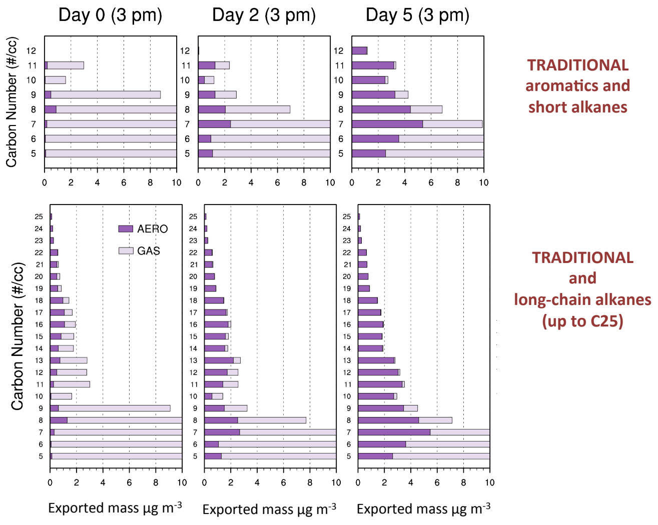

Figure 4: 5-day evolution of gas-particle partitioning of the organic mass as a function of carbon number as simulated for traditional precursors (aromatics and short alkanes, 1st row) and for a more complex mixture including long-chain alkanes (up to C25). From Hodzic et al., GRL, 2013.

(iv) Sensitivity of SOA to sunlight exposure (Hodzic et al., 2015, ACPD):

There is increasing evidence from laboratory experiments that UV exposure of organic aerosols leads to a substantial decrease in their mass or to their smaller production yields. Processes responsible for this decrease are however still unknown, and in particular it is currently unclear how much of this decrease is due to gas-phase fragmentation versus fragmentation occurring inside the particle.

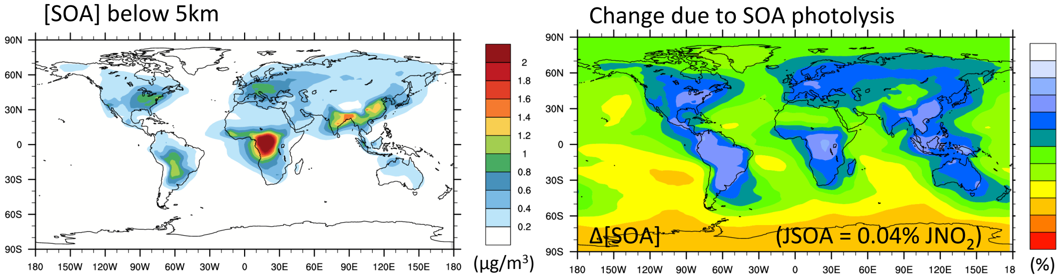

Figure 5: GEOS-Chem simulation for 2009 of SOA concentrations within the lower troposphere, and the relative (%) reductions in SOA concentrations due to photolytic loss reactions. The photolysis rate of JSOA = 0.04% JNO2 is considered.

In this project, we performed the first modeling estimates of the potential effect of gas- and particle-phase organic photolysis reactions on the formation and lifetime of SOA. Typically only photolysis of smaller organic molecules (formaldehyde) for which explicit data exist is included in 3D models. In our study, we specifically examine the photolysis of larger molecules that actively partition between the gas and particle phases. The chemical mechanism generator GECKO-A was used to explicitly model SOA formation from α-pinene, toluene, and C12 and C16 n-alkane reactions with OH at low- and high-NOx. Simulations were conducted for typical mid-latitude conditions and a solar zenith angle of 45°. Our results show that after four days of chemical aging under those conditions (equivalent to eight days in the summer mid-latitudes), gas-phase photolysis leads to a moderate decrease in SOA yields i.e ~15% (low-NOx) to ~45% (high-NOx) for α-pinene, ~15% for toluene, ~25% for C12-alkane, and ~10% for C16-alkane. The small effect on low volatility n-alkanes such as C16-alkane is due to the rapid partitioning of early-generation products to the particle-phase where they are assumed to be protected from gas-phase photolysis. Minor changes are found in the volatility distribution of organic products and in oxygen to carbon ratios. The decrease in SOA mass seems increasingly more important after a day of chemical processing, suggesting that most laboratory experiments are likely too short to quantify the effect of gas-phase photolysis on SOA yields. Our results also suggest that many molecules containing chromophores are preferentially partitioned into the particle phase before they can be photolyzed in the gas-phase. Given the growing experimental evidence that these molecules can undergo in-particle photolysis, we performed sensitivity simulations using an estimated SOA photolysis rate of $J_{SOA}=4\times10^{-4}J_{NO2}$. Modeling results indicate that this photolytic loss rate would decrease SOA mass by 40-60% for most species after ten days of atmospheric aging at mid-latitudes. It should be noted that in our simulations we do not consider in-particle or aqueous-phase reactions which could modify the chemical composition of the particle, and thus the amount of photolabile species. The atmospheric implications of our results are significant for both the SOA global distribution and lifetime. Applications in a global model suggest that particle-phase photolytic reactions could be an important loss process for SOA in the atmosphere, removing aerosols from the troposphere on timescales of less than 7 days that are comparable to wet deposition (see Figure 5).

(v) Sensitivity of SOA to changes in solubility and removal of organic vapors (Hodzic et al. GRL, 2013, 2014; Knote et al. ACP, 2015):

The dry deposition removal of organic compounds from the atmosphere and its impact on SOA mass is currently highly uncertain in chemistry-climate models. The main reason for this uncertainty is that current models use simplified SOA mechanisms that lump precursors and their products into volatility bins (VBS algorithm), therefore losing information on other properties of individual molecules (or groups) that are needed to calculate dry deposition. In this project, we used an explicit model to first calculate the solubility for all organic oxidation intermediates, and performed idealized simulations to quantify the sensitivity of SOA to their removal. Then we built a simplified parameterization of the solubility that can be used in 3D models with any VBS SOA formation scheme, and we applied this parameterization within WRF-Chem to estimate regional effects of this process. Details of these 3 studies are described below:

- ***Quantifying the importance of dry deposition of organic vapors in an urban plume in a box model (Hodzic et al., GRL, 2013):*** First, we have applied the explicit chemistry model GECKO-A to simulate SOA formation and estimate the influence of dry deposition of SVOCs on SOA concentrations downwind of Mexico City. SOA precursors considered include short- and long-chain alkanes (C3-25), alkenes, and light aromatics. We show that dry deposition of oxidized organic gases is not a substantial sink of anthropogenic SOA, as it removes <5% of SOA within the city's boundary layer and ~15% downwind in this idealized case study. We investigated reasons for this limited influence, and investigated separately the impacts on short and long-chain species. We showed that the dry deposition is competing with the uptake of gases to the aerosol phase, and because dry deposition of submicron aerosols is slow, condensation onto particles protects organic gases from deposition and therefore increases their atmospheric burden and lifetime. In the absence of this condensation, ~50% of the regionally produced mass would have been dry-deposited. This study was the first step towards understanding how these processes can be simplified and parameterized into 3D models.
- ***Building parameterization of water solubility of organic compounds (Hodzic et al., GRL, 2014):*** The water solubility of oxidation intermediates of volatile organic compounds that can condense to form SOA is unconstrained in current chemistry and climate models ranging from 0 to 10^5 M/atm. We used the GECKO-A model to calculate Henry's law constants for these intermediate species. We have shown that water solubility is strongly correlated with saturation vapor pressures of organics, and investigated how this relationship depends on precursor species, extent of photochemical processing, and NOx levels. We have made available the Henry's law constants as a function of volatility over a wide range of vapor pressures for use in 3D models. This study was also the first study to parameterize GECKO-A for use in 3D models. We have established the methodology and gained great experience in running GECKO-A for a large number of individual precursors and mixtures at various NOx levels, and initial conditions. We have been configured GECKO-A to run on the NCAR super computer, allowing us to perform several hundreds of simulations in one day. This feature is key for enabling the use of explicit models such as GECKO-A for building parameterizations for 3D chemistry-climate models.
- ***Application within WRF-Chem to estimate the effect of the water solubility on the SOA lifetime (Hodzic et al. GRL 2014; Knote et al. ACP 2015):*** Finally,

we implemented the water solubility representation of organic vapors into WRF-Chem, and evaluated its regional impacts on SOA concentrations. We showed that dry and wet deposition of condensable organic vapors leads to major reductions in SOA over the continental U.S., decreasing surface concentrations by ~50% (10%) for biogenic and ~40% (6%) for short-chain anthropogenic precursors under the considered volatility conditions. The effect in the 3D model was larger than the predicted effect within the GECKO-A box model in Hodzic et al., 2013, because we found that biogenic SVOCs were much more soluble than anthropogenic ones, and because the WRF-Chem SOA (based on currently used VBS, Lane et al., 2008) produces more volatile organic vapors than GECKO-A. Sensitivity of our results to volatility distribution of the SOA formation mechanism, and to the dry deposition scheme were also investigated in the study, and our conclusions were shown to be robust i.e. dry deposition of organic vapors modulates the burden and lifetime SOA.

Other papers acknowledging this grant:

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