Secondary Organic Aerosol Formation Regulates Cloud Condensation Nuclei in the Global Remote Troposphere

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Abstract
Formation of secondary organic aerosols (SOA) through the atmospheric oxidation of organic vapors has potential to enable particle growth to cloud condensation nuclei (CCN)-relevant sizes. In this work, we constrain a global aerosol model by using aircraft measurements to reveal the global importance of SOA formation in CCN production. Our improved model, with explicit size-resolved aerosol microphysics and parametrizations of semivolatile organic oxidation products, presents a state-of-the-art performance in simulating both particle number concentrations and organic aerosol concentrations dominated (80–95%) by SOA in the remote atmosphere, which have been challenges in previous modeling studies. The SOA formation in concert with aerosol nucleation contributes to more than 50% of CCN concentrations in those pristine environments featuring low background aerosol concentrations. We estimate that the SOA-derived CCN alters the magnitude of cloud radiative forcing by ∼0.1 W m⁻². Our findings underscore the necessity for aerosol-climate models to represent controls on CCN concentrations by SOA production.

Plain Language Summary
Atmospheric aerosols with diameters of larger than 60 nm or so can serve as cloud condensation nuclei (CCN) that affect cloud properties and Earth’s radiative balance. The determinants of CCN remain one of the largest uncertainties in the assessment of aerosol-radiation-cloud interactions. The secondary organic aerosols (SOA) formed by the oxidation and gas-aerosol partitioning of precursor organic vapors is an important, yet unsettled, source of CCN. The extent to which SOA formation impacts the global distribution of CCN has rarely been reported, in part because previous global aerosol models had a poor ability in simulating both SOA and particle number concentrations. Here, by constraining an aerosol-climate model using in situ aircraft measurements for number and mass concentrations of aerosols, we find that SOAs, formed by the oxidation of primary organic vapors from anthropogenic and natural emissions, dominate the growth of small particles to CCN in the global remote troposphere. The formation of SOAs exerts larger percentage contributions to CCN in preindustrial atmosphere than in the present day, appreciably altering the magnitude of cloud radiative forcing.

1. Introduction
The aerosol-cloud interaction, which involves the activation of aerosol into cloud droplets by serving as cloud condensation nuclei (CCN) and the resulting changes to cloud albedo and lifetime, is a leading cause of the large uncertainties in assessing aerosol effects on climate by global chemistry-climate models (Bellouin et al., 2020; Sockol & Small Griswold, 2017). Reducing those uncertainties in models rely on our understanding toward the ability of particle population to act as CCN and how available CCN controls cloud properties, in particular related to particle number size distributions, chemical constituents, and their hygroscopicity (Farmer et al., 2015; McFiggans et al., 2006; Rosenfeld et al., 2019).

Liquid clouds in the marine remote atmosphere span enormous regions of Earth’s surface and strongly affect the global energy balance (Wood, 2012). It is of great importance to constrain the particle number size distributions in the marine atmosphere in climate models with in situ measurements. The NASA Atmospheric Tomography Mission (ATom) offers a comprehensive global-scale measurement data set of particle number concentrations over the Pacific and Atlantic Oceans (Wofsy, 2018). By employing this latest observational database, recent studies found a poor performance of state-of-the-art models in simulating particle number concentrations in both nucleation-and accumulation-modes (Ranjithkumar et al., 2021; Williamson et al., 2019). The representation of atmospheric aerosol processes (e.g., new particle formation and further growth by condensation of inorganic...
and organic vapors) in models has a controlling influence on the simulated vertical profiles of particle number concentrations (Merikanto et al., 2009).

The growth of small particles to form CCN requires vapor condensation and coagulation processes to reach the critical diameter at a specific supersaturation (Merikanto et al., 2009). While sulfuric acid is commonly known as a vital component in the nucleation of clusters and their condensational growth, condensable organics have been recently suggested to be an important or even dominant contributor to the growth of particles in the remote marine troposphere based on limited in situ measurements (Kupc et al., 2020; Zheng et al., 2020). Some modeling studies evaluated simulations of organic aerosol (OA) concentration and its effects on particle number size distributions using only continental near-surface observations (D’Andrea et al., 2013; Sengupta et al., 2021). The ATom measurements show that organic components are abundant from the surface to the upper troposphere over remote oceans and constitute a major fraction of sub-micron particle mass (Hodzic et al., 2020). In particular, secondary organic aerosols (SOA) account for more than 90% of the total OA mass during the campaigns and have large potential to contribute to CCN.

It remains ambiguous how condensable organic vapors contribute to the growth of particles to CCN in the global troposphere, especially in remote regions. Herein, we explore the extent to which secondary organics contribute to CCN budgets by using a global aerosol microphysical model that treats both aerosol size evolution due to aerosol nucleation and subsequent growth and process-based organic aerosol formation. We leverage the ATom data set to constrain the modeled particle number concentrations in nucleation and accumulation modes and bulk mass concentrations of particulate organics, and then quantify the impacts of secondary organics on particle condensational growth and their modifications to CCN budgets and radiative forcings.

2. Results

2.1. Constraints on OA Mass and Particle Number Concentrations

We use the volatility basis set (VBS) scheme (Donahue et al., 2006) for the simulation of SOA formation in our global aerosol model (CAM-ATRAS; see Text S1 in Supporting Information S1 and the references therein for the details) (Matsui, 2017; Matsui & Mahowald, 2017). CAM-ATRAS simulations have been validated in our recent studies for the mass and number concentrations of various aerosol components and their optical properties using a wide range of measurements (Gliß et al., 2021; Kawai et al., 2021; Liu & Matsui, 2021a, 2021b; Matsui & Liu, 2021; Matsui & Moteki, 2020; Matsui et al., 2018, 2022). In the VBS approach, we define several surrogate volatility species to represent the oxidation and aging of semi- and intermediate volatile organic compounds (S/I VOCs), which have been widely included to improve simulated OA mass concentrations over both land and ocean areas (Matsui et al., 2014; Shrivastava et al., 2011; Tsipidi et al., 2016). Given the limited understanding on ocean-produced organic aerosol and its minor role in the global OA budget (Brooks & Thornton, 2018; Burrows et al., 2014; Tsigris et al., 2013), only organic matters produced from continental sources (i.e., anthropogenic, biomass burning, and biogenic activities) are considered in this work. The simulated OA species are grouped into POA (primary organic aerosol), OPOA (OA from oxygenated S/I VOCs), ASOA (OA from oxygenated anthropogenic and biomass burning VOCs), and BSOA (OA from oxygenated biogenic VOCs) based on their precursors. The baseline simulation (referred to as the base case) is performed for the years from 2015 to 2018 to cover all ATom measurement campaigns, and the last three years of model outputs are used for analysis.

First, we validate the simulated OA mass profiles against the ATom data at four latitudinal bands across the Pacific and Atlantic oceans (Figures 1a–1d; also see Figure S1 in Supporting Information S1 for the geographical ranges of the four latitudes). Our model reproduces mostly within a factor two of the observed OA profiles throughout the troposphere in the Northern Hemisphere, especially in the midlatitudes. The model measurement comparisons in this study resemble the multiple-model ensemble mean results by Hodzic et al. (2020). The correlation coefficients (0.53–0.60) between the simulation and the measurements during ATom-1 and ATom-2 (Table S1 in Supporting Information S1) are close to the results in Hodzic et al. (2020) (0.48–0.66).

Importantly, the temporal mean fractions of SOA in total OA mass in our simulations range between 80% and 95% over the Pacific and Atlantic troposphere, which agree in general with the measurement-derived estimates (~90%) by Hodzic et al. (2020). The atmospheric oxidation and aging of VOCs and S/I VOCs are taken into account in our VBS scheme and the resulting SOA (OPOA + ASOA + BSOA) production dominates OA mass budgets from the surface to the upper troposphere (Figures 1e–1h). The simulated mass fractions of OA in the
submicron particles (diameters less than 1.0 μm, i.e., PM$_{1}$) are averaged at 56% in the free troposphere along the ATom-1 flight track and 46% along the ATom-2, in accord with the observational results of PM$_{1}$ (56% and 43%). These validations suggest that our global simulation with the treatment of SOA formation reproduces the observed OA mass concentrations and their secondary fractions reasonably well in the remote troposphere.

The OA simulations are also compared to observations in polluted regions, including East Asia and North America (Figure S2 in Supporting Information S1). The simulated OA mass concentrations agree well with aircraft in situ measurements over South Korea, while showing larger biases across different campaigns over North America, possibly related to inaccurate anthropogenic emission inventory and physical processes (Pai et al., 2020). Overall, our simulated global mean mass burdens of OA are 3.3 Tg, comprised of OPOA (1.6 Tg), ASOA (1.1 Tg), BSOA (0.4 Tg), and POA (0.2 Tg), similar to the simulation result (3.0 Tg yr$^{-1}$) by Tsimpidi et al. (2014) but higher than the value (2.4 Tg yr$^{-1}$) in Pai et al. (2020) due to inconsistent estimates of ASOA.

We then evaluate the simulations of particle number concentrations linked to CCN availability using the ATom measurements. Figure 2 depicts the comparisons of modeled particle number concentrations that are greater than 3 nm ($N_{3}$; cm$^{-3}$) and 60 nm ($N_{60}$) with the corresponding ATom measurements (expressed as mean values and standard deviations) over the four latitude bands. The Base simulation mostly reproduces the magnitude and vertical variability of observed $N_{3}$ values, with some overestimation in the upper troposphere of the southern mid-latitudes (Figures 2a–2d). Specifically, the high number concentrations of small particles (e.g., $N_{3–7}$; particle with diameters of between 3 and 7 nm) exceeding 1,000 cm$^{-3}$ in the tropical upper troposphere are captured well by our model (Figures 2e–2h), which are associated with frequent deep convection there favoring abundant nucleation events there (Weigel et al., 2011; Williamson et al., 2019). Note that $N_{3–7}$ tends to be overestimated in the upper troposphere of midlatitudes. The number concentrations of nucleation-mode particles are not only related to aerosol nucleation rates, but also affected by available vapors to promote the growth of small particles and the abundances of larger preexisting particles (i.e., accumulation-and coarse-modes) (Pierce & Adams, 2007).

Figure 1. Constraints on organic aerosols (OA) mass concentrations using Atmospheric Tomography Mission measurements at four latitudinal regions. (a–d) Comparison of simulated OA mass concentrations (red lines) with the corresponding measurements (black lines) in the atmosphere (0–12 km); (e–h) simulated mass fractions of OA components in the Base case. Shaded areas denote one standard deviations from the observation means. The model results with three hourly resolution are matched to observations in location (latitude, longitude, and altitude) and time.

![Image of Figure 1](image-url)
N₆₀ can serve as an indicator for CCN numbers at less than 1% supersaturation. The modeled N₆₀ profiles match remarkably well with the ATom measurements over the four ocean basins, with their discrepancies overwhelmingly within a factor of two (Figures 2i–2l). To our knowledge, few studies have presented such a good performance for CCN-sized particle number concentrations over the global remote regions in climate models.
As a comparison, Williamson et al. (2019) revealed underestimates of around a factor of five in \( N_{60} \) by multiple models during the ATom campaigns. Furthermore, the variabilities of condensation sink (CS), reflecting the probability of condensation and coagulation and governed by the population of large particles, are captured well by our model (Figures 2m–2p). Both the simulations and observations demonstrate that the \( N_i \) are dominated by nucleated particles (e.g., \( N_{n} \)), in the upper troposphere, where more nucleated particles can survive under the relatively clean environment with lower CS values. Overall, the Base simulation results are well constrained by the ATom measurements and reproduce the particle number concentrations realistically in the global remote troposphere, especially for those in CCN-sized bins.

2.2. Global Impacts of SOA Formation on Particle Growth

To examine the contribution of condensable secondary organics to the growth of particles, we switch off the formation of SOA under the Base case (referred to as the SOAoff case) such that all OA in the atmosphere are from the POA emissions without further oxidation and gas-aerosol partitioning processes. On the other hand, we explore the importance of aerosol nucleation mediated by sulfuric acid in particle number distributions by closing the nucleation mechanisms under the Base case (Nucloff; see Text S1 in Supporting Information S1).

As illustrated in Figure 2 for the particle number profiles derived from the SOAoff case (blue lines), notable decreases of \( N_{60} \) are found across four latitude bands compared to the Base simulation results (red lines), which in turn suggest the significant contributions of secondary organics to particle growth. In the tropics and midlatitudes, the gaps of simulated \( N_{60} \) profiles between the Base and SOAoff cases (i.e., differences between red and blue lines) are progressively enlarged from the near-surface to the higher altitudes. The difference between the Base and SOAoff cases suggest that the oxidation of organic vapors and associated condensation on small particles contribute to 45–85% of \( N_{60} \) in the upper troposphere across different regions (200–400 hPa). The increased importance of secondary organics with altitude is associated with active deep convection, whereby primary particles are removed efficiently during air parcel updrafts and simultaneously quantities of organic vapors can be lifted into upper troposphere and contribute remarkably to the growth of nucleated particles there (Liu & Matsui, 2021b; Yu et al., 2019). Over the Arctic and surrounding areas (55–80°N), the contributions of secondary organics to \( N_{60} \) are marked throughout the troposphere due to low background aerosol concentrations (Figure 2i).

Note that the nucleation-mode particle number concentrations \( (N_{3,7}) \) are elevated slightly by removing SOA formation, because of the reductions in particle surface areas and the coagulation sinks (lower condensation sink). The \( N_i \) simulations in the SOAoff case maintain a good consistency with the ATom measurements, despite the substantial changes of \( N_{60} \). Hence, only the evaluation on total particle number concentrations cannot guarantee the good performance of cloud condensation nuclei (CCN)-sized particles, like \( N_{60} \). Our results highlight that it is necessary for models to treat SOA formation and organic vapor condensation to reproduce the observed particle number concentration profiles at CCN-sized bins.

When turning off aerosol nucleation, the modeled particle number concentrations are directly from primary aerosols and no nucleation-mode particle exist in this simulation. Both \( N_i \) and \( N_{60} \) are lowered by orders of magnitude in the Nucloff case (green lines in Figure 2), despite the condensation of inorganic and organic vapors on primary particles. This finding in turn suggests that the contributions of SOA formation on CCN-sized particles is facilitated by the condensational growth of nucleation mode particles. The nucleation and SOA formation processes jointly regulate the CCN budget in the global remote atmosphere. The sensitivities of CCN to different aerosol nucleation schemes need to be further investigated.

Taking the tropics and northern midlatitudes as examples, we calculate the pressure-dependent particle size distributions throughout the troposphere (Figure 3). The model results in the Base case agree well with the corresponding ATom observations, with nucleation-mode (<10 nm) particles predominant in the upper troposphere and accumulation-mode (60–500 nm) particles more important in the lower troposphere (Figures 3a–3d). Further, by comparing the differences in the results between the Base and the SOAoff cases, we find a shift in the particle number distributions toward larger sizes by considering SOA formation. Secondary organics contribute to 72% and 59% of the particle number concentrations at diameters within 15–80 nm in the upper troposphere for the tropics and the midlatitudes (Figures 3e and 3f), respectively, suggesting a major role of secondary organic vapors in condensational growth of particles. Representing the formation and condensation of secondary organic vapors thus greatly enhances the potential of particle growth to CCN.
2.3. SOA Contributions to CCN in the Global Remote Troposphere

We next quantify the contributions of secondary organics to CCN at 0.2% supersaturation (CCN0.2) and 1.0% supersaturation (CCN1.0), respectively. Figure 4 presents the pressure-latitude zonal mean distributions of CCN0.2 and CCN1.0 on a global scale and the fractional contributions by SOA in the present-day atmosphere. The simulated CCN concentrations for the Base case are highest in the lower troposphere of the midlatitudes and tropics, with CCN0.2 and CCN1.0 values reaching above 300 cm$^{-3}$, reflecting the substantial contributions of anthropogenic emissions and biomass burning (Merikanto et al., 2009; Yu & Luo, 2009). The SOA contributions to CCN significantly increase from below 5% near the surface to more than 50% at high altitudes. SOA plays a more dominant role in CCN1.0 than CCN0.2 because SOA formation facilitates the growth of nucleated particles to CCN sizes at high supersaturation of 1.0%. Our simulations reveal that SOA formation contributes to 50% (28%) of the mean CCN1.0 (CCN0.2) budget in the global atmosphere at pressures between 400 and 200 hPa, which is in accord with the changes in $N_{60}$ profiles between the Base and SOAoff cases (Figure 2). In the lower stratosphere of the extratropical regions (e.g., <200 hPa), the SOA contributions to CCN1.0 are higher than 50% in our simulations (Figure 4d), because organic compounds constitute a major fraction of submicron particles there (Figure S3 in Supporting Information S1). Comparably, Murphy et al. (2021) show that smaller-mode submicron particles in the lowermost stratosphere are rich in organics that are likely transported from the troposphere.

The annual-mean SOA contributions to column-averaged CCN throughout the troposphere are estimated for both present-day conditions (PD) using the 2010 emissions and the preindustrial conditions (PI) using the 1750 emissions to provide implications on cloud radiative forcings (Figure S4 in Supporting Information S1). Our simulations reveal that in the PD atmosphere, secondary organics contribute to 10–20% of the column-averaged CCN0.2 and CCN1.0 over most of the Pacific and Atlantic Oceans, and as high as 40% in the Arctic. The average percent contributions to CCN0.2 (CCN1.0) over the marine troposphere are estimated to be 17% (18%) globally and 28% (21%) in the Arctic. In the PI atmosphere, however, anthropogenic emissions and the associated CCN are negligible, and organic vapors from natural sources, including wildfire and biogenic emissions, could play a
more important role in the growth of particles to serve as CCN than the PD conditions. Under the cleaner PI conditions, secondary organic vapors contribute to 24% (21%) and 54% (37%) of the CCN0.2 (CCN1.0) over the global marine troposphere and in the Arctic, respectively. The contribution of SOA was calculated as the difference in total CCN between the Base and SOAoff simulations.

We estimate that the effect of semivolatile SOA formation on CCN reduces the magnitude of the global mean cloud radiative forcing by 0.08 [0.02–0.13] W m$^{-2}$ (see Methods for the estimation), which means that the cloud forcing becomes less negative. This reduction of cloud radiative forcing is attributable to the combined effect of higher cloud albedo sensitivity to CCN changes in the PI era than in the PD, as suggested by Carslaw et al. (2013), and the concurrent larger SOA contributions (in percentage) to CCN in the cleaner PI atmosphere found in this study. Such result indicates that the modifications on cloud radiative forcing by natural SOA formation is dependent not only on organic nucleation (Gordon et al., 2016; Scott et al., 2014), but also on particle growth by condensing secondary organics. In addition, we find that including SOA formation gives rise to a more negative OA instantaneous radiative forcing due to aerosol-radiation interactions, from $-0.04$ W m$^{-2}$ to $-0.22$ W m$^{-2}$. The results imply a strong dependence of aerosol radiative forcings on the treatment of SOA formation.

3. Summary and Implication

This work presents an assessment of the global contributions of semivolatile organic aerosol formation to CCN abundances in the remote troposphere by constraining a global size-resolved aerosol model (CAM-ATRAS) with
the ATom aircraft measurements. Our model simulation successfully captures both particle number size distribution and OA mass concentrations across the global remote atmosphere by considering such SOA formation from anthropogenic and natural S/I VOCs aging processes, and thus provides a realistic picture of CCN-relevant particle distributions. The results further suggest a vital role of SOA formation in the growth of nanosized particles to CCN sizes in the remote upper troposphere and pristine environment. The aerosol nucleation and SOA formation processes therefore jointly regulate the CCN budget in the global remote atmosphere. The inclusion of SOA formation contributes more in percentage to global CCN in the preindustrial atmosphere than in the present day, which can modify the magnitude of the cloud radiative forcing by the order of 0.1 W m$^{-2}$. Our results highlight the importance of SOA treatment in shaping vertical profiles of particle number concentrations and the resulting climate effects via aerosol-cloud interactions over remote oceans.

Our findings will shed light on the modeling of global CCN concentrations in future studies and provide several implications and further research needs for better understanding aerosol-cloud interactions. First, the simulations of CCN availability in the remote atmosphere should be constrained by observations for both particle number concentrations in CCN-sized bins and SOA mass fractions, as CCN abundance strongly depends on the contributions of secondary components, especially those from semivolatile and condensable organic vapors. Second, the condensational growth of newly formed particles by secondary organics could be another important aerosol process altering the estimates of PI-to-PD radiative forcings, which in conjunction with biogenic organic nucleation complicates the effects of organic aerosols on global radiative balances. The source appointment of SOA-derived CCN can aid in determining the baseline state of CCN in the PI atmosphere, especially for those from natural emissions like wildfires, biogenic VOCs, and marine organics. Moreover, this study does not consider the role of extremely low volatile organic vapors in new particle formation, as suggested by Tröstl et al. (2016) and Gordon et al. (2017), which could be important for CCN estimates over continental regions. The model results of aerosol compositions and their size distributions in the lower stratosphere are somewhat uncertain and need to be further evaluated because current understanding on the source and sink of organic aerosols in the stratosphere is limited (Martinsson et al., 2019). We call for more effort to reduce the model uncertainties in organic aerosol behaviors, CCN distributions attributable to organics, and their influences on cloud properties.

**Conflict of Interest**

The authors declare no conflicts of interest relevant to this study.

**Data Availability Statement**

The NASA Atmospheric Tomography Mission data can be accessed from the links: https://daac.ornl.gov/ATOM/campaign/data.html or https://doi.org/10.3334/ORNLDAAC/1581. Simulation results used to reproduce the figures are publicly available at the link: https://zenodo.org/record/6117612.

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