



# Global budgets of atmospheric primary and secondary organic aerosols constrained by full-volatility-range organic emissions

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**Abstract.** Organic aerosol (OA) constitutes a major fraction of tropospheric submicron particulate matter, with primary (POA) and secondary (SOA) components exhibiting different physicochemical properties and health impacts. The POA and SOA budgets are however highly uncertain, and the results from different model studies are confusing because of more or less consideration of the volatility distributions of organic precursor emissions and inconsistent attributions of model tracers  
30 in model-observation comparisons. Here we develop an OA simulation framework in GEOS-Chem that resolves the full volatility spectrum of organic precursor emissions from anthropogenic sources and open biomass burning. The model reasonably reproduces the observed OC, POA, and SOA concentrations from comprehensive surface, shipborne, and airborne datasets, providing a consistent global validation. The model simulations suggest greater POA (0.5 Tg) and SOA burdens (2.0 Tg) and potentially stronger and more widespread impacts of the OA components on air quality, health, and  
35 radiation than previous estimates, led by both of the emission updates and the revised OA scheme. The simulated global SOA production is about 106 Tg in 2018, 46% of which is contributed by open biomass burning. The results demonstrate distinct regional variations in the dominant source types and population exposure distributions of POA and SOA,



40 highlighting the needs for SOA mitigation, multi-sector control measures, and clean energy replacements for long-term health-oriented air quality improvements globally. The model results are sensitive to the emissions and wet-deposition parameterization, calling for more measurement constraints on local emission factors and the deposition fluxes of OA and its components.

## 1 Introduction

Organic aerosol (OA) constitutes a major fraction of tropospheric submicron particulate matter, with important implications for air pollution, climate, and human health (Jimenez et al., 2009; Heald et al., 2014; Shiraiwa et al., 2017; Shrivastava et al., 45 2017). OA comprises primary organic aerosol (POA), emitted directly or formed immediately by gas-particle partitioning of semi-volatile organic gases, and secondary organic aerosol (SOA), formed through atmospheric multiphase chemical oxidation of organic precursors. These components differ fundamentally in physicochemical properties and climate and human health impacts. SOA is generally more hygroscopic and thus more effective at forming cloud condensation nuclei (Thalman et al., 2017). Differences in chromophore composition lead to distinct optical properties of POA and SOA (Lin et al., 2014; Li et al., 2025). Moreover, both POA and SOA contribute to aerosol oxidative potential (Verma et al., 2015; Daellenbach et al., 2020), yet epidemiological evidence in the United States suggests a stronger association of long-term SOA exposure with cardiopulmonary mortality than POA (Pye et al., 2021; Pond et al., 2022). These contrasts underscore the need for quantitative constraints on POA and SOA at the global scale.

Despite decades of development, chemical transport models (CTMs) still struggle to represent OA consistently (Hallquist et al., 2009; Tsigaridis et al., 2014; Hodzic et al., 2016; Riipinen et al., 2026). A central challenge lies in the wide volatility spectrum of organic precursors, spanning semivolatile and low-volatility organic compounds (S/LVOCs;  $C^* < 300 \mu\text{g}/\text{m}^3$ ), intermediate-volatility organic compounds (IVOCs;  $300 \mu\text{g}/\text{m}^3 < C^* < 3 \times 10^6 \mu\text{g}/\text{m}^3$ ), and volatile organic compounds (VOCs) ( $C^* > 3 \times 10^6 \mu\text{g}/\text{m}^3$ ) (Donahue et al., 2012). While VOC emissions are relatively well constrained, emissions of S/LVOCs and IVOCs remain highly uncertain. Most models infer S/LVOCs from bulk organic carbon (OC) emissions without resolving volatility distributions, leading to systematic biases (Pye and Seinfeld, 2010; Jathar et al., 2011; Shrivastava et al., 2015; Hodzic et al., 2016). Assuming nonvolatile POA in CTM can overestimate the POA concentrations by over 100% in outflow and remote regions (Jo et al., 2013; Murphy et al., 2017; Brewer et al., 2023). By contrast, CTM could reproduce the large fractional contributions of oxidized OA observed in the atmosphere only if some of the S/LVOCs in the model were allowed to evaporate and subsequently oxidize in the atmosphere (Shrivastava et al., 2008). Overly volatile assumptions 65 however underestimate urban POA by over 50% (Woody et al., 2016; Jiang et al., 2019; Miao et al., 2021). Uncertainties are even larger for IVOCs, whose emissions are typically derived from proxies such as POA, non-methane VOCs (NMVOCs), and speciated IVOCs (i.e., naphthalene) using empirical scaling (Shrivastava et al., 2008; Hodzic et al., 2016; Pye and Seinfeld, 2010). Global emission estimates of IVOCs span more than an order of magnitude ( $16.2 \text{ Tg yr}^{-1}$  to  $234 \text{ Tg yr}^{-1}$ ) (Jathar et al., 2011; Hodzic et al., 2016; Pye and Seinfeld, 2010; Miao et al., 2021), with substantial discrepancies in source



70 attribution, seasonality, and spatial distributions. For example, POA-based estimates show greater emissions from residential  
combustion and stronger seasonal variation than NMVOC-based estimates (Miao et al., 2021; Yang et al., 2025). These  
uncertainties propagate directly into SOA formation, where yields vary strongly with volatility and molecular structure,  
ranging from  $<0.1$  to  $>0.4$  for IVOCs (Lu et al., 2020).

Recent advances now enable a more explicit treatment of this complexity. New bottom-up inventories quantify emissions  
75 across the full volatility range—from VOCs to S/LVOCs—using measurements from advanced mass spectrometry (Chang  
et al., 2022; Pye et al., 2023; Huang et al., 2023; Xu et al., 2026). The newly-developed MEIC-global-FVOC inventory  
incorporates sector and region-specific emission factors and suggests 28-41% higher global anthropogenic emissions of full-  
volatility range organic compounds (FVOCs) than previous estimates (Xu et al., 2026). These volatility-resolved emissions  
enable more mechanistic representations of SOA formation. For example, model studies incorporating volatility-binned  
80 IVOCs with bin-specific SOA yields report substantially reduced urban-rural differences with enhanced SOA concentrations  
(Lu et al., 2020; Miao et al., 2021; Manavi and Pandis, 2024; Scholz et al., 2025), suggesting greater and more widespread  
health impacts of OA than previously recognized. However, evaluations of such frameworks remain limited, particularly at  
the global scale, and constraints on POA–SOA partitioning are still sparse.

Here we develop a global OA simulation framework in the GEOS-Chem chemical transport model that resolves emissions  
85 across the full volatility spectrum. We evaluate model performance against a comprehensive observation dataset that  
includes surface, shipborne, and airborne measurements, and further assess POA and SOA against aerosol mass spectrometer  
source apportionment. This framework enables a quantitative reassessment of the global distribution and budget of POA and  
SOA, providing new constraints on their sources and variability.

## 2 Methods

### 90 2.1 Model configurations

We use the GEOS-Chem (version 14.3.0, DOI: 10.5281/zenodo.10640536) to simulate OA. Global simulations are run at  $2^\circ$   
 $\times 2.5^\circ$  horizontal resolution with a vertical resolution of 47 layers from surface to 0.01 hPa. Nested-grid simulations are  
conducted over North America, Europe, Asia, and the Atlantic Ocean at a horizontal resolution of  $0.5^\circ \times 0.625^\circ$ , with  
boundary conditions from the global simulations. All simulations are driven by meteorological data from the MERRA2  
95 reanalysis with a one-month spin-up. The base year for our simulations is 2018.

The model includes ozone-NO<sub>x</sub>-hydrocarbon-aerosol-halogen chemistry for the troposphere (Bey et al., 2001). The  
ISORROPIA-II thermodynamic module is applied to simulate sulfate-nitrate-ammonium aerosols (Fountoukis and Nenes,  
2007; Pye et al., 2009). Dry deposition is calculated by a standard resistance-in-series model (Wang et al., 1998; Zhang et  
al., 2001). Wet deposition includes wet convective updrafts and large-scale precipitation (Liu et al., 2001). The scavenging  
100 efficiencies of different types of aerosols in wet deposition depend on their hydrophilicity without considering the effect of

aging processes. For example, the scavenging efficiencies of POA and SOA are set to be 50% and 80%, respectively (Chung and Seinfeld, 2002; Miao et al., 2021).

For pollutants other than organic precursors, anthropogenic emissions in China and the rest of the world are provided by the Multi-resolution Emission Inventory model for Climate and air pollution research (MEIC) and the Community Emissions  
105 Data System (CEDs), respectively (Geng et al., 2024; Hoesly et al., 2018). Open biomass burning emissions are provided by the Fire Inventory from the National Center for Atmospheric Research (NCAR) version 2.5 (FINNv2.5) (Wiedinmyer et al., 2023). Soil NO<sub>x</sub> emissions are calculated by the Berkeley-Dalhousie Soil NO<sub>x</sub> Parameterization (BDSNP) (Hudman et al., 2012). Biogenic emissions are calculated by the Model of Emissions of Gases and Aerosols from Nature (MEGAN v2.1) (Guenther et al., 2012).

## 110 2.2 Full-volatility-range OA scheme

The standard GEOS-Chem model includes two OA schemes named as “simple OA” and “complex OA”. The simple OA scheme treats POA as non-volatile and applies fixed yields to estimate SOA, whereas the complex OA scheme treats POA as semivolatile with more sophisticated parameterization and SOA yields (Pai et al., 2020). The complex OA scheme is firstly  
115 described by Pye and Seinfeld (2010), which considers SOA from VOCs, IVOCs, and S/LVOCs. In this scheme, all IVOC emissions are allocated to one lumped species with a SOA yield referring to naphthalene. IVOC emissions are estimated as 66 times of naphthalene emissions, which are approximated from benzene by the approach suggested by Pye and Seinfeld (2010). For S/LVOCs, their emissions are 1.27 and 1 times of the OC emissions for anthropogenic sources and open biomass burning, provided by CEDs and FINNv2.5, respectively, following an update suggested by Pai et al. (2020). The 49% and  
120 51% of the S/LVOC emissions are allocated to the two lumped species with C\* of 1646 and 20 μg m<sup>-3</sup>, respectively (Shrivastava et al., 2006). The complex OA scheme also has an option to treat POA as non-volatile, which is no longer recommended because of the large SOA underestimation (Schroder et al., 2018).

The full-volatility-range OA scheme is built based on the complex OA scheme in GEOS-Chem (Pye et al., 2010; Pai et al., 2020). The graphical overviews of these two schemes are shown in Figure S1, and their OA tracers are listed in Table S1 in the Supplement. First, a volatility-basis-set (VBS) approach with updated NO<sub>x</sub>-dependent SOA yields is used to simulate the  
125 SOA production from the oxidation of isoprene, terpenes, and aromatics. The updates in SOA yields and vapor wall-loss corrections aim to better represent the low-loading gas-particle partitioning (Zhang et al., 2014; Chen et al., 2024). Vapor wall-loss correction factors of 1.2, 1.9, and 2.2 are used for the oxidation of aromatics under high-NO<sub>x</sub> conditions, the oxidation of aromatics under low-NO<sub>x</sub> conditions, and the high-NO<sub>x</sub> pathway of isoprene SOA formation, respectively. The heterogeneous uptake of isoprene epoxydiols (IEPOX) on aerosols is included, which applies parameterizations on aerosol  
130 acidity and nucleophile concentration-sensitive reactive uptake coefficients (Marais et al., 2016). To avoid double counting in low-NO<sub>x</sub> conditions, the traditional low-NO<sub>x</sub> pathway for isoprene SOA formation is turned off (Pai et al., 2020).

Additionally, the heterogeneous reactions of glyoxal and methylglyoxal on aerosols are included, for which fixed reactive uptake coefficients are applied (Marais et al., 2016).

135 Unlike the default scheme for which tracers only represent limited volatility bins, the full-volatility-range OA scheme deals with continuously binned OA precursors and their oxidation processes (Figure 1). IVOCs are represented by four lumped species that have decadal saturation concentrations ( $C^*$ ) ranging from  $10^3$  to  $10^6 \mu\text{g m}^{-3}$ , and the SOA production from the oxidation of IVOCs is simulated by the VBS approach. The SOA yields of IVOC species are listed in Table S2 in the Supplement. Under high- $\text{NO}_x$  conditions, the SOA yields derived from the photooxidation of *n*-alkanes are applied to the lumped IVOC species to represent the increasing SOA yields with the decreasing volatility of IVOC species (Presto et al., 140 2010; Zhao et al., 2014). We apply the same SOA yields for the IVOC species with  $C^*$  of  $10^3$  and  $10^4 \mu\text{g m}^{-3}$  due to the lack of experiments for IVOC species with carbon numbers over 17 in their formulae. Under low- $\text{NO}_x$  conditions, the IVOC SOA is treated as nonvolatile mass with a constant SOA yield of 73% (Chan et al., 2009; Pye and Seinfeld, 2010). In addition, the IVOC SOA yields are corrected for vapor wall loss with a correction factor of 1.2 for both low- and high- $\text{NO}_x$  pathways (Zhang et al., 2014). For S/IVOCs, the gas phase has four lumped species that have decadal  $C^*$  ranging from  $10^{-1}$  to  $10^2 \mu\text{g m}^{-3}$ , whereas the particle-phase portion is represented by five lumped species with decadal  $C^*$  of  $10^{-2}$  to  $10^2 \mu\text{g m}^{-3}$ . Then the 145 gas-phase S/LVOCs undergo photooxidation with a rate constant of  $2 \times 10^{-11} \text{ cm}^3 \text{ molec}^{-1} \text{ s}^{-1}$  in the scheme to produce oxidation products with 100 times lower  $C^*$  than their precursors (Pye and Seinfeld, 2010). These low-volatility products follow gas-particle partitioning to form SOA. After emission and dilution, the S/LVOCs that remain in or recondense into the particle phase without undergoing any chemical processing are classified as POA.

150 Anthropogenic emissions of the 9-bin lumped organic species are provided by the newly-developed MEIC-global-FVOC inventory (Xu et al., submitted). MEIC-global-FVOC estimates the emissions of organic species by a bottom-up approach under the framework of MEIC based on activity rates, technology distributions, unabated emission factors for different regions, and available control technologies. The emission factors for volatility-binned S/LVOCs and IVOCs from  $C^*$  of  $10^{-2}$  to  $10^6 \mu\text{g m}^{-3}$  are harmonized from recent emission characterization experiments for various sources by advanced 155 measurement techniques, such as two-dimensional gas chromatography-time-of-flight mass spectrometry ( $\text{GC} \times \text{GC-TOF-MS}$ ), thermal desorption-gas chromatography/mass spectrometry (TD-GC-MS), proton-transfer-reaction time-of-flight mass spectrometer (PTR-ToF-MS). Major source sectors in this inventory include coal combustion, oil combustion, biomass combustion, fugitive and industrial processes, and volatile chemical products (VCPs) for industrial and domestic uses. The emission factors of S/LVOCs for biomass combustion of wood and crop for developing regions such as China, 160 India, and African countries are further constrained by local emission factors of OC to reduce the biases in mass closure of S/LVOCs in analytical detection and the regional differences in stove types and combustion conditions. Moreover, empirical scaling factors of 4 and 6 are applied to the residential biofuel combustion-related S/LVOC emissions in Western and Eastern Europe, respectively. Without these scaling factors, the simulated concentrations of biomass burning-related OA (BBOA) are much lower than the observation-derived BBOA concentrations (Figure S2 in the Supplement).



165 Waste treatments and open biomass burning are not represented by MEIC-global-FVOC. We follow our previous work to  
apply emission ratios to NMVOC and OC to derive the total amounts of IVOC and S/LVOC emissions of these sources,  
respectively, and then distribute the emissions to volatility bins based on assumed volatility distributions (Chen et al., 2024).  
The emissions from waste treatments that are proportional to the waste-related NMVOC and OC emissions from CEDS are  
added as additional anthropogenic emissions. This source is important in populated urban areas in developing regions (e.g.,  
170 Africa and India), where incineration of municipal solid waste occurs without any pollution control (Sharma et al., 2019;  
McDuffie et al., 2020). The empirical emission ratios for waste treatments are listed in Table S3 in the Supplement. The total  
anthropogenic IVOC and S/LVOC emissions in 2018 are 37.4 Tg and 25.4 Tg, respectively (spatial distributions shown in  
Figure S3a in the Supplement). For open biomass burning, the emissions of IVOC and S/LVOC are derived from the  
NMVOC and OC emissions from FINNv2.5 with empirical scalars listed in Table S3 (Chen et al., 2024; Wiedinmyer et al.,  
175 2023). The total IVOC and S/LVOC emissions from open biomass burning are 36.7 Tg and 38.5 Tg in 2018, respectively.  
The spatial distributions of IVOC and S/LVOC emissions from open biomass burning are shown in Figure S3b in the  
Supplement. In addition, biogenic emissions for isoprene and terpenes are calculated by MEGAN v2.1, and the biomass  
burning emissions for VOC precursors such as isoprene, terpenes, and aromatics are provided by FINNv2.5.

Configurations of the model runs are listed in Table S4 in the Supplement. Simulation runs with the full-volatility-range OA  
180 scheme and the FVOC emissions are named as “Base”, while the default complex OA scheme-based simulation runs are  
named as “Default”. One sensitivity run (“Default\_emis”) applied the updated emission amounts of FVOCs with the original  
volatility distributions and SOA formation parameterization in the default complex OA scheme. The emission-related  
concentration differences are therefore calculated as “Default\_emis” – “Default”, and the scheme-related concentration  
differences are calculated as “Base” – “Default\_emis”. Other simulation runs such as “Zero\_Biof”, “Zero\_Anthro”,  
185 “Zero\_BioBurn”, “Zero\_Biog”) excluded OA precursor emissions from residential and biofuel, anthropogenic, biomass  
burning, and biogenic sources, respectively. In addition, we applied the wet deposition parameterization developed by Luo et  
al. (2020) in the simulation (“WDLuo”), which modifies rainout and washout parameters from the GEOS-chem default wet  
deposition, to explore the sensitivity of the revised OA simulation to wet-deposition settings.

### 2.3 Ambient observations for model evaluations

190 The locations of ambient observations are shown in Figure S4 in the Supplement. The OC data set consists of annual mean  
concentrations of OC (2016-2021) in PM<sub>2.5</sub> from 335 sites in various monitoring networks such as the Interagency  
Monitoring of Protected Visual Environments (IMPROVE) and the Chemical Speciation Network (CSN) in U.S., the  
European Monitoring and Evaluation Programme (EMEP), the aerosol composition network led by China National  
Environmental Monitoring Centre (CNEMC), and the International Network to study Deposition and Atmospheric  
195 composition in Africa (INDAAF). In addition, 58 annual mean concentrations obtained from year-long measurements  
between 2016 and 2021 at research sites in Asia, Africa, and Latin America are included (Table S5 in the Supplement).



The POA and SOA data sets include source-apportionment results from ground, shipborne, and airborne measurements by Aerodyne aerosol mass spectrometers (AMS or ACSM) (Tables S6-S8 in the Supplement). Primary and secondary OA factors were resolved, including hydrocarbon-like (HOA), cooking-related (COA), biomass-burning-related (BBOA), coal-combustion-related OA factors (CCOA), and oxygenated OA factors (OOAs). We consider the sum of HOA, BBOA, and CCOA as the observation-derived POA and the sum of OOAs as the observation-derived SOA. The COA factor is not included in the model-observation comparisons because of the lack of cooking emissions in the model. In total, 165 worldwide campaign-average surface concentrations (2015-2022) as well as the season-mean concentrations from 21 sites (2015-2019) in the Aerosol, Clouds and Trace Gases Research Infrastructure (ACTRIS) are used in model validation. All of the observation-derived POA and SOA concentrations are compared to the simulated campaign-mean or season-mean concentrations for the base year of 2018 for computation efficiency. The interannual variations of OA formation are expected to be small because of minor changes in anthropogenic emissions of organic precursors during this period in most regions (Xu et al., 2026). Anthropogenic emissions in China had a rapid decline from 2013 to 2017, followed by a slow decline afterwards. By using the middle year of observations as the simulation reference, the potential biases in model-observation comparisons are counteracted. The interannual variations of meteorology modify annual mean OA concentration within  $\pm 10\%$  and have a minor influence on the trends (Ridley et al., 2018; Chen et al., 2024), leading to little impact on the model-observation comparisons.

Shipborne measurements we used in this study consist of four cruise tracks that occurred from April to May and October to November in 2011 and 2012 over the Atlantic Ocean between  $53^\circ$  N and  $53^\circ$  S (Huang et al., 2018). Marine and non-marine factors are resolved by the positive matrix factorization (PMF) analysis of AMS measurements during the four cruises, which showed high correlations with time series of source tracers and mass spectral patterns of identified sources. Airborne data are taken from the Wintertime Investigation of Transport, Emissions, and Reactivity (WINTER) and the Korea-United States Air Quality (KORUS-AQ) campaigns. The former was conducted in the northeastern United States from February to March in 2015 (Schroder et al., 2018), and we used the data from RF01 to RF07 for the model evaluation. The KORUS-AQ campaign was conducted over South Korea and the Yellow Sea in East Asia from May to June in 2016 (Nault et al., 2018). Model results sampled for the locations and specific date and time of each aircraft and cruise campaign are used in the comparisons.

### 3 Results and discussion

#### 3.1 Model validation

Figure 2 shows the concentration differences between our updated OA simulation and the simulation with the default complex OA scheme, which are further attributed to concentration changes arising from the precursor emission update and the full-volatility-range OA scheme. The updated OA simulation results in higher OA concentrations in most continental



regions, especially suggesting an increase of 3 to 10  $\mu\text{g m}^{-3}$  of annual-mean surface OA concentrations in polluted regions. The largest increases are present in East Asia and Central Africa, due to the net increases in both POA and SOA.

230 Under the newly-developed MEIC-global-FVOC emissions, the simulated POA shows a slight change in most regions (Figure 2b), suggesting most of the S/LVOC emissions are captured by traditional OC emissions. By contrast, the simulated SOA mass concentrations show 20-60% increases in East Asia, Europe, southern America, and Africa compared to the default complex OA scheme, mainly due to the increased IVOC emissions (i.e., from 6.0 to 74.1 Tg globally). The simulated POA and SOA concentrations in South Asia are however lower than the default complex OA scheme, which is attributed to  
235 relatively higher S/LVOC emissions in CEDS in that region than in MEIC-global-FVOC emissions. Overall, the precursor emission update leads to net increases of simulated OA concentrations in most continental regions.

The full-volatility-range OA scheme further increases both POA and SOA worldwide, with more increases in POA than SOA (Figure 2c). The default complex OA scheme assumes that all S/LVOC emissions fall into two surrogate bins with  $C^*$  of 1646 and 20  $\mu\text{g m}^{-3}$  (Shrivastava et al., 2006). The revised OA scheme improves the representation of S/LVOC volatilities  
240 and tends to distribute more emissions into lower-volatility bins than the default scheme does, leading to increased POA mass in most continents. Then fewer S/LVOCs remain in the gas phase and hence their contributions to the SOA formation are reduced. Meanwhile, the revised OA scheme improves the representation of IVOC volatilities and their SOA yields, leading to increased SOA formation from IVOCs. In most regions, the increased SOA formation from IVOC updates is partially offset by the decreased SOA formation from S/LVOC updates, resulting in net increases of surface SOA  
245 concentrations. However, net decreases of SOA concentrations occur in some parts of Africa, South America, and North America, which are greatly affected by extreme wildfires. Because open biomass burning emissions have a greater fraction of S/LVOCs in their volatility distribution than anthropogenic emissions (Figure S5 in the Supplement), the SOA reduction from S/LVOC updates is greater than the enhancement from IVOC updates in those areas.

Figure 3 shows the comparisons of observed and simulated campaign-mean concentrations of OC, POA, and SOA in North  
250 America, Europe, and Asia. The model generally reproduces OC, POA, and SOA across regions. For POA, the normalized mean bias (NMB) values range from -0.15 to -0.01, suggesting a reasonable global estimation of the S/LVOC emissions (Figure 3b). For SOA, the NMB value is -0.14 for Asia (Figure 3c), but lower in North America (-0.28) and Europe (-0.38), suggesting unresolved underestimations in the two regions. Uncertain IVOC emissions can contribute to the underestimation of SOA in North America. The MEIC-global-FVOC shows about 20% lower VCP-related IVOC emissions in the United  
255 States than local inventories (Xu et al., 2026). Uncertain residential emissions plausibly contribute to the underestimation of SOA in Europe. There is a lack of local emission factors of S/LVOCs and IVOCs for residential biofuel burning in Europe, and emission activities of residential biofuel use are difficult to quantify there, especially in Eastern Europe (Denier van der Gon et al., 2015; Bertelsen and Vad Mathiesen, 2020). Previous studies show over 50% of OA underestimations in Europe and have attributed their underestimations to the uncertain residential wood combustion and biogenic emissions (Chebaicheb  
260 et al., 2024; Tsimpidi et al., 2025). We applied empirical scalars of 1 to 6 to increase the residential biofuel-related S/LVOC

emissions in some parts of Europe (Figure S2). Such residential biofuel corrections significantly improve the simulations of OC (NMB: -0.47 to -0.22), POA (NMB: -0.50 to -0.15), and SOA (NMB: -0.54 to -0.38) in Europe (Figure S6 in the Supplement), with more improvements in cold seasons (Figure 4). Additionally, insufficient mechanistic representation of SOA formation pathways may cause the underestimation at high SOA concentrations (Figure 3c). For example, aqueous and heterogeneous processes are important SOA formation pathways during haze episodes in China but remain poorly parameterized in current CTM models (Zheng et al., 2021; Gkatzelis et al., 2021). In India, high chlorine emissions may affect the SOA formation (Gunthe et al., 2021). However, neither the chlorine emissions nor the chlorine-driven pathways are well understood and sufficiently considered in the model (X. Liu et al., 2024).

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It is worth mentioning that the observations for OC, POA, and SOA are still limited in high-population-density regions such as Southeast Asia, Africa, and Latin America. Panels a-c in Figure 5 show the model-observation comparisons of surface OC concentrations in those regions. The model performances are reasonable for Southeast Asia and Africa, whereas the modelled OC concentrations are much lower than the observed concentrations in Latin America. The observations in Latin America are all from urban sites, suggesting an urgent need to understand the anthropogenic sources and local emission factors. We further compared the model results to the shipborne observations. Panels e-f in Figure 5 show the simulated and observed non-marine SOA concentrations in the Atlantic Ocean (Huang et al., 2018). High concentrations of over  $2 \mu\text{g m}^{-3}$  are observed in the mid-latitudes of the Northern Hemisphere and near the equator, which were influenced by long-range transport of anthropogenic pollution or open biomass burning. For other regions, the background non-marine SOA concentrations are typically below  $0.5 \mu\text{g m}^{-3}$ . The model results can reproduce such variability over the Atlantic Ocean, providing a continent-wide validation of the model as well as the emission estimates. Some overestimation of the modeled non-marine SOA concentrations is present near the equator, which may be explained by underrepresented open biomass burning emissions there (Wiedinmyer et al., 2023).

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Figure 6 shows the modelled vertical profiles of POA and SOA concentrations compared to the observation-derived profiles in the northeastern United States (WINTER campaign) and in East Asia (KORUS-AQ campaign). The model reproduces the observed vertical variations and concentrations of POA and SOA in the northeastern United States. For KORUS-AQ in East Asia, the model captures the vertical variations of POA and SOA, although it underestimates both POA and SOA at low altitudes. Such underestimation may result from underrepresented local emissions in the global inventory (Brewer et al., 2023; Park et al., 2021). The modelled POA concentrations are generally over 50% greater than the observations 500 m above the surface. This model bias suggests regional bias in the volatility distributions of S/LVOC emissions, which may be related to widely-used residential coal in Asia for which emission factors are limited (Shen et al., 2022). When comparing to a global data set of OA vertical profiles from aircraft campaigns, previous studies have shown that the GEOS-Chem model can capture the observed variability in OA with acceptable model biases (Pai et al., 2020; Brewer et al., 2023). These model studies did not reproduce the AMS-derived vertical profiles of POA and SOA concentrations. The traditional OA scheme with nonvolatile POA assumption would overestimate POA aloft, while the default complex OA scheme usually



underestimates POA (Schroder et al., 2018). The full-volatility-range OA scheme in this study shows more robust  
295 performance across different source regions and various chemical regimes for SOA production.

### 3.2 Surface concentrations and vertical profiles of OA

Global distributions of simulated annual-mean surface concentrations of OA, POA, and SOA from the full-volatility-range  
OA scheme are shown in panels a-c of Figure 7. High annual-mean OA concentrations that exceed  $10 \mu\text{g m}^{-3}$  occur in China  
and India as well as several places in Southeast Asia, Sub-Saharan Africa, and South America. By contrast, the annual-mean  
300 OA concentrations are usually below  $5 \mu\text{g m}^{-3}$  in most places in North America and Europe. In general, the SOA mass  
concentrations are greater than POA on the model-grid scale, and the SOA mass fractions may increase from about 30-50%  
in source regions to 70-90% in remote regions (Figure S7a in the Supplement).

Figure 8 shows the main sources of OA in different regions. In China, India, Europe, and Western Africa, anthropogenic  
sources are the predominant contributors to the OA mass, among which 50-70% of the mass is SOA. Because residential  
305 burning is the largest contributor to S/LVOCs in those regions (Xu et al., 2026), control measures on this sector would be  
most effective for mitigating the SOA pollution. Chen et al. (2024) show that single sector control strategy is insufficient to  
lower the OA levels down to  $5 \mu\text{g m}^{-3}$  in China. Multi-sector control measures and clean energy replacements are urgently  
needed to achieve tightened air quality standards. In Southeast Asia, Southern Africa, and some parts of North America,  
open biomass burning is the predominant contributor to the OA mass, whereas biogenic emissions are responsible for about  
310 30-60% of the OA mass in most parts of South America and Oceania. Under climate change, the OA concentrations in these  
regions may increase because of the enhanced wildfires and biogenic emissions (Pfannerstill et al., 2024; Cunningham et al.,  
2024).

Population exposure distributions of POA and SOA show significant differences across regions, highlighting the potential  
difference in POA and SOA impacts (Figure 8). For example, consistent steeper slopes of POA than SOA are present in  
315 India and China, suggesting greater reduction of population exposure per unit mass mitigation for POA. In addition,  
incomplete combustion is often associated with residential burning, the predominant source of POA in India and China.  
Studies have shown that this type of POA could have higher oxidative potential (i.e., a driver for acute health risks) than  
SOA (Bhattu et al., 2024; F. Liu et al., 2024). We therefore infer that the POA mitigation may bring more health co-benefits  
than SOA in India and China. This is very different from Europe and the United States, where SOA slopes are similar to  
320 POA's, and greater oxidative potential has been attributed to SOA (Daellenbach et al., 2020). The two factors together  
support the stronger association of long-term SOA exposure with cardiopulmonary mortality than POA reported by  
epidemiological studies (Pye et al., 2021; Pond et al., 2022). Moreover, the slopes of POA and SOA at high exposures in  
Southern Africa are driven by open biomass burning, and thus the majority of the population is not affected. By contrast,  
populations in Southeast Asia face significantly higher exposure to widespread open biomass burning, specifically by the  
325 SOA generated from these emissions.



The simulated zonal-mean distributions of OA, POA, and SOA concentrations are shown in panels d-f of Figure 7. The OA concentrations decrease from surface to high altitudes, reflecting the centralized precursor emissions and SOA formation near the surface. Below 600 hPa, the OA concentration is highest at the equator region, explained by high emissions of open biomass burning there (Figure S3). The Northern Hemisphere has much greater OA concentrations than the Southern Hemisphere, resulting from high anthropogenic emissions at mid-latitudes. Vertical inversions of OA concentrations occur in upper altitudes in polar regions, as is consistent with previous model findings (Tilmes et al., 2019; Lou et al., 2020). Such inversions are mainly driven by the long-range transport of SOA from mid-latitudes to high latitudes (Figure 7f). The average mass fractions of SOA may increase from over 60% at the surface to over 85% at 200 hPa (Figure S7b). Although POA contributes only 10-15% of the OA mass in upper altitudes, POA exhibits much stronger absorption than SOA (Li et al., 2025; Wang et al., 2025). Overall, a greater and more widespread absorption effect from OA is expected compared to previous estimates.

### 3.3 Global OA budgets

Figure 9 shows the simulated global OA burden and its comparisons to previous findings. Many previous studies treated POA as nonvolatile species and reported high POA burdens of 0.65 to 0.94 Tg (Hodzic et al., 2016; Jo et al., 2023; Lou et al., 2020; Pai et al., 2020). The nonvolatile assumption would cause large overestimations of the POA concentrations (Schroder et al., 2018; Brewer et al., 2023). The studies that considered POA as semivolatile and applied simplified parameterizations for the gas-phase partition of S/LVOCs reported much lower POA burdens of 0.03-0.24 Tg (Pye and Seinfeld, 2010; Tsimpidi et al., 2016). This discrepancy highlights the importance of a reasonable representation of the S/LVOC volatilities in modelling OA (Tsimpidi et al., 2017). Our simulations are constrained by comprehensive global data sets of POA and SOA. The estimated POA, SOA, and the total OA burdens are 0.5 Tg, 2.0 Tg, and 2.5 Tg, respectively (Table 1). Compared to the default simulations (Pye and Seinfeld, 2010), the POA burden is increased by 14.7-fold, and the SOA burden is increased by 25%. Our estimated OA burden is greater than most previous findings, but similar to the burden estimated by Lou et al. (2020), albeit with lower primary and greater secondary components.

Globally, the SOA production is  $105.6 \text{ Tg yr}^{-1}$  and represents 82% of the total OA production. This is consistent with previous top-down estimates (Heald et al., 2010; Spracklen et al., 2011). Biogenic SOA contributes to about 23% of the SOA burden (0.5 Tg), similar to the previous findings of 0.3 to 0.7 Tg (Pye and Seinfeld, 2010; Tsimpidi et al., 2017; Hodzic et al., 2016; Jo et al., 2023; Pai et al., 2020). The remaining 77% of the global SOA burden is contributed by anthropogenic and open biomass burning SOA, for which S/LVOCs and IVOCs are the main precursors and their emissions are most uncertain. Our simulations suggest S/LVOCs and IVOCs together account for 1.2 Tg of the global SOA burden, which is greater than previous findings of 0.2 Tg to 1.0 Tg. The higher SOA burden suggests that the climate impacts of SOA may have been overlooked in previous studies. Open biomass burning contributes to 46% of the SOA burden, highlighting the potential climate-driven changes of the SOA burden and distributions in the future.



The estimated mean lifetimes of OA, POA, and SOA are 7.1, 7.5, and 7.0 days (Figure 10a-b). The shorter lifetime of SOA than that of POA is consistent with several studies, resulting from greater wet-deposition scavenging efficiency of SOA. Wet deposition is crucial to the global OA budget, especially in remote regions (Luo et al., 2020; Gao et al., 2022). We conduct test simulations with the wet-deposition parameterization developed by Luo et al. (2020). The Luo parameterization applies a greater wet removal efficiency to reduce biases in simulated nitric acid, nitrate, and ammonium, but simultaneously decreases the estimated global OA burden by 0.8 Tg (Table S9). The global burdens of POA and SOA would decrease by 31% and 34%, respectively. Such reductions are less near sources but greater in remote regions (Figure 10c-d). The Luo parameterization may also lead to much shorter lifetimes for POA and SOA (i.e., from 7 days to 5 days in our simulations) (Figure 10a-b). Moreover, several previous studies have considered additional chemical loss pathways (e.g., via photolysis and fragmentation reactions) for SOA in the model to balance the SOA burden after implementing uncertain and empirical updates to enhance the SOA production. This implementation may lower the SOA lifetimes to less than 4 days (Hodzic et al., 2016; Lou et al., 2020). Recent studies suggest lower OA photolysis rates compared to the ones applied in the model (O'Brien and Kroll, 2019; Guan et al., 2025). Nevertheless, there is still a lack of measurements to support sophisticated parameterizations on the chemical production and removal processes for different types of OA.

#### 4 Conclusion

In this study, we developed a new modeling framework to adopt full-volatility-range emissions of organic precursors for OA simulation in GEOS-Chem. This framework facilitates the model representation of gas-particle partitioning and SOA formation of organic precursors over a wide range of volatility and thus improves the model skills in the characterization of POA and SOA on a global scale. The new simulations show greater POA and SOA burdens across continents and suggest potentially stronger and more widespread impacts of the OA components on air quality, health, and radiation, led by both of the emission updates and the revised OA scheme. The results highlight the difference in dominant source types and population exposure distributions of POA and SOA in different regions. In India and China, POA mitigation, especially on incomplete residential burning, may still bring great health co-benefits. Long-term health-oriented air quality improvements globally need more focus on SOA, for which multi-sector control measures and clean energy replacements are necessary.

The model results are validated by a comprehensive data set including surface, airborne, and shipborne measurements. One caveat is that the PMF-derived POA and SOA concentrations are not identical to the modelled POA and SOA. We think the comparisons between the sum of HOA, CCOA, COA, and BBOA to the modeled POA and the sum of OOAs to the modeled SOA are meaningful because (1) the PMF POA factors cover the main anthropogenic source types whose emissions are represented by the new inventory except cooking; (2) the PMF OOA factors represent SOA from various precursors with different photochemical ages as well as aqueous SOA (Jimenez et al., 2009; Chen et al., 2015; Zheng et al., 2023), and the modelled SOA is the sum of biogenic SOA, aromatic SOA, IVOC SOA, S/LVOC SOA, and key aqueous SOA which covers all important precursors and their photooxidation and aqueous processes. The mass fraction of COA on a regional or global



390 scale should be small. The PMF-derived SOA includes heterogenous aging of POA which may explain a part of the underestimation of modeled SOA but expected to be small given the good comparisons aloft and in remote regions.

Although we have updated the OA scheme with continuous volatility bins and better SOA yields, the SOA scheme remains as a volatility-lumped and yield-based approach. Model accuracy is limited when exploring local temporal variations (Woody et al., 2016; Miao et al., 2020; Oak et al., 2022; Chen et al., 2024). Advanced measurements on local emission  
395 factors of full-volatility-range organic precursors from anthropogenic sources as well as the simultaneous quantification of the total organic carbon mass to constrain the S/LVOC emissions are crucial but limited, particularly for Europe and India. OA source apportionment data are rare in Southeast Asia, Africa, and Latin America, calling for more ambient measurements there. There is still room for improving the yield parameterization. The IVOC-SOA yields in this study depend on only the IVOC precursor volatilities, which should include justifications on their molecular structure and the NO<sub>x</sub>  
400 level (Tkacik et al., 2012; Lu et al., 2020; Manavi and Pandis, 2022). Moreover, the nonlinear response of SOA formation to global warming is largely unknown. The temperature dependence of organic emissions needs to be considered in the model (Wu et al., 2024; Pfannerstill et al., 2024). Our results highlight the high contribution of open biomass burning to the global SOA burden. Significant changes in SOA production are expected in the future because of the reduction of anthropogenic emissions driven by tightened regional air quality standards (e.g., in China) and the intensified wildfires. The difference in  
405 global annual emissions of OC from open biomass burning is over 100% among commonly used emission inventories and much higher regionally (Carter et al., 2020; Wiedinmyer et al., 2023). It is important to further constrain open biomass burning emissions and understand the chemical conversion of biomass burning plumes. Finally, the simulated OA burden is sensitive to the parameterization of wet deposition, calling for more measurements of the deposition fluxes of OA and its components.

#### 410 **Data availability**

The airborne measurement data and PMF results of the WINTER and KORUS-AQ campaigns are downloaded from UCAR/NCAR - Earth Observing Laboratory (<https://doi.org/10.5065/D6MC8XT8>) and are provided by the dataset of Brewer et al. (2023) (<https://doi.org/10.7910/DVN/IDWE39>), respectively. IMPROVE and CSN data are downloaded from <http://views.cira.colostate.edu/fed/>. EMEP and INDAAF data are downloaded from <https://ebas-data.nilu.no/> and  
415 <https://indaaf.obs-mip.fr/>, respectively. Surface measurement data that gathered from literatures are listed in the Supplement. Other data presented in this paper are available upon request to the corresponding author.

#### **Author contributions**

Q.C. designed the study. R.X., J.L., and G.G. provided the MEIC-global-FVOC emission inventory. R.M. and R.X. prepared the data set of surface OC measurements. R.M., S.X., H.W., Y.Z., and S.L. prepared the data set of global AMS and ACSM



420 measurements. K.R.D., I.E.H., and A.S.H.P. provided the measurement data from ACTRIS network. S.H. provided the data of shipborne measurements. R.M. developed the model scheme and performed the model simulations. R.M, Q.C., R.X., and I.E.H. analyzed the model data. R.M, Q.C., R.X., J.J., I.E.H., M.S., C.G., G.B., V.A.K. and A.P.T. discussed the model results. R.M. and Q.C. wrote the paper with comments from all the other authors.

### Competing interests

425 At least one of the (co-)authors is a member of the editorial board of Atmospheric Chemistry and Physics.

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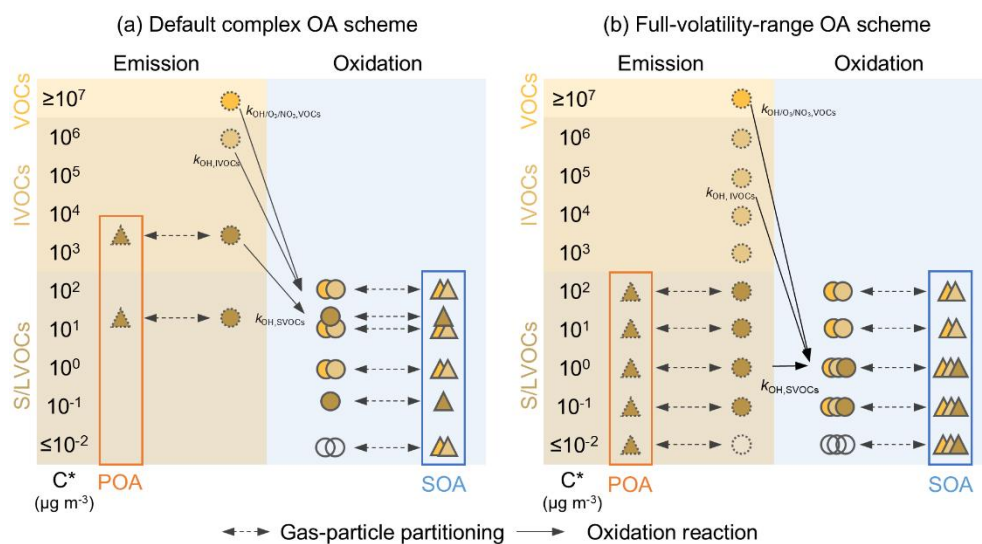


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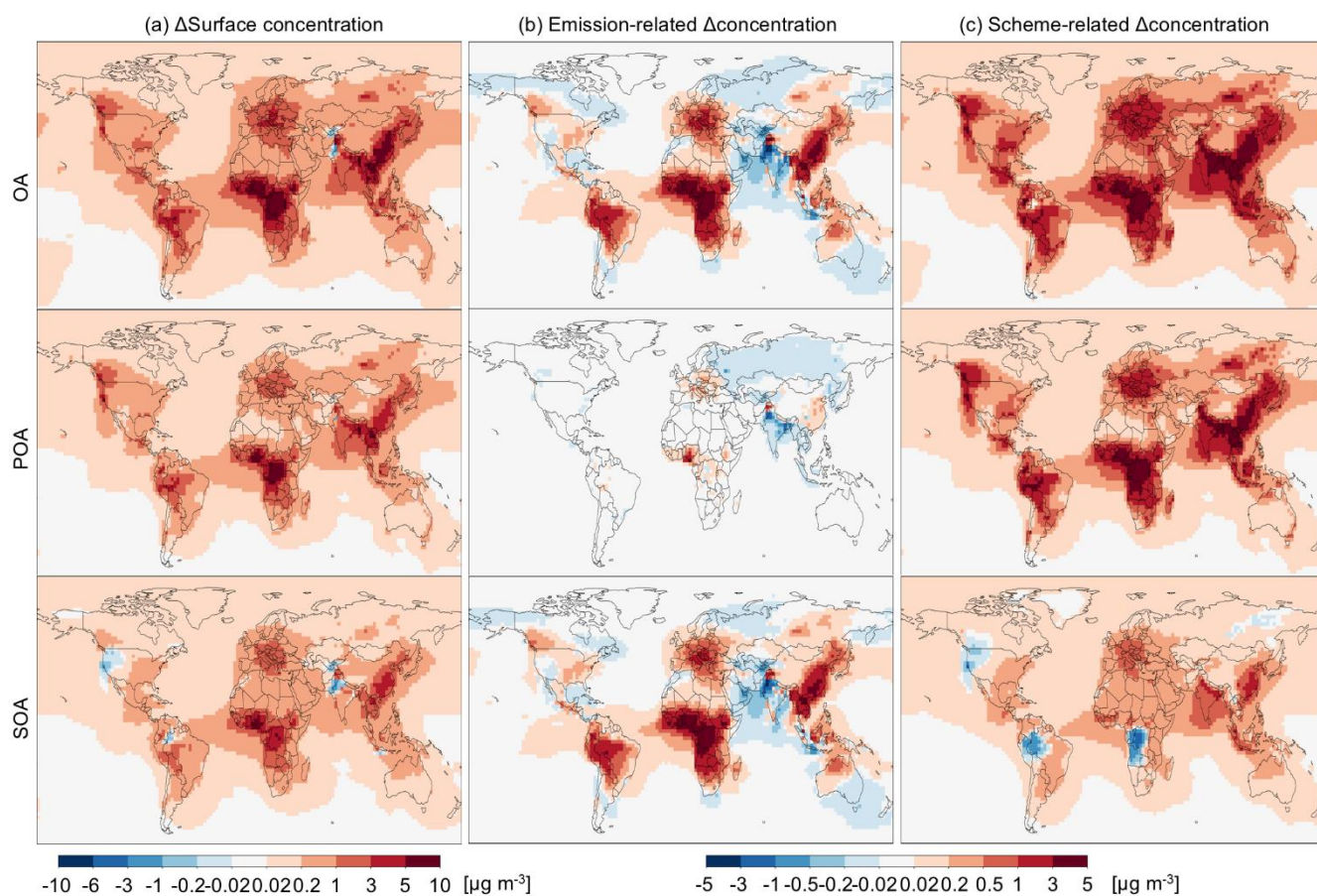


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Figure 1. Schematic diagram of the POA and SOA formation from VOC, IVOC, and S/LVOC precursors in the (a) default complex OA and (b) full-volatility-range OA schemes. Colored circles and triangles represent model tracers of gaseous precursors, oxidation products, and OA components. For  $C^* \leq 10^{-2} \mu\text{g m}^{-3}$ , OA components mostly remain the particle phase at ambient conditions and thus their gaseous tracers (open circles) are ignored in the model scheme.



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795 **Figure 2.** The differences of annual-mean surface concentrations of OA, POA, and SOA between different simulation runs in GEOS-Chem (Table S4), including (a) the overall changes calculated as “Base” – “Default”, (b) the emission-related changes calculated as “Default\_emis” – “Default”, and (c) the scheme-related changes calculated as “Base” – “Default\_emis”.



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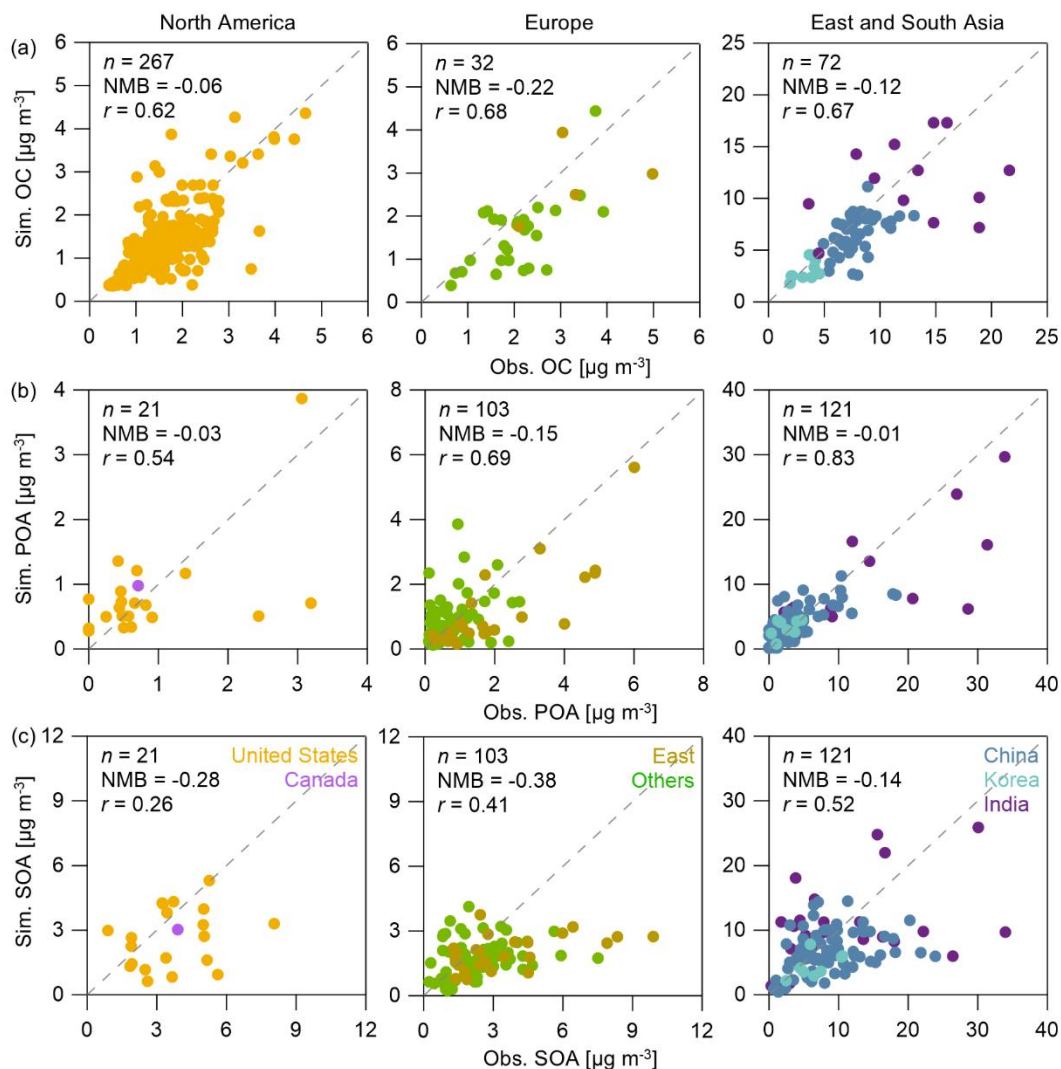
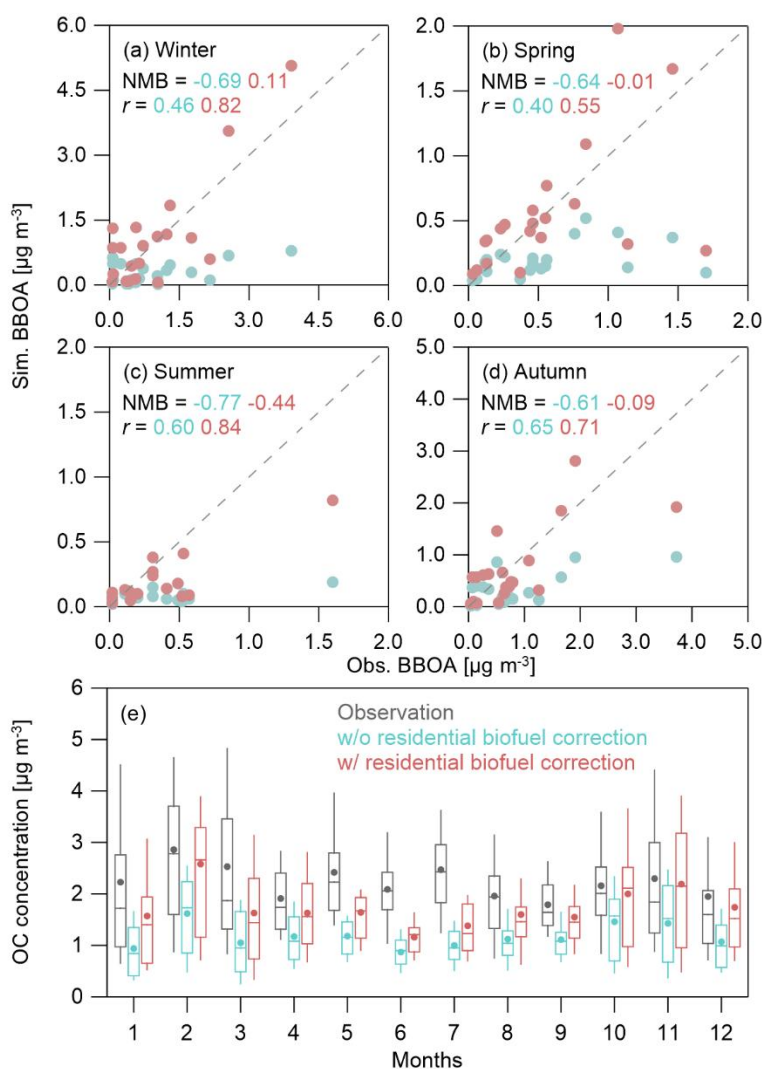


Figure 3. Scatter plots of the observed and simulated (“Base”) concentrations of (a) OC, (b) POA, and (c) SOA in North America, Europe, and East and South Asia. East European countries include Poland, Romania, Estonia, Lithuania, Greece, and Cyprus. Other European countries include Czech, Germany, Switzerland, Finland, Norway, Spain, Italy, France, Ireland, Netherland, and United Kingdom. The OC data are annual mean surface concentrations taken from various monitoring networks. The POA and SOA data are the campaign-mean concentrations derived from the source-apportionment results of the surface AMS or ACSM measurements.

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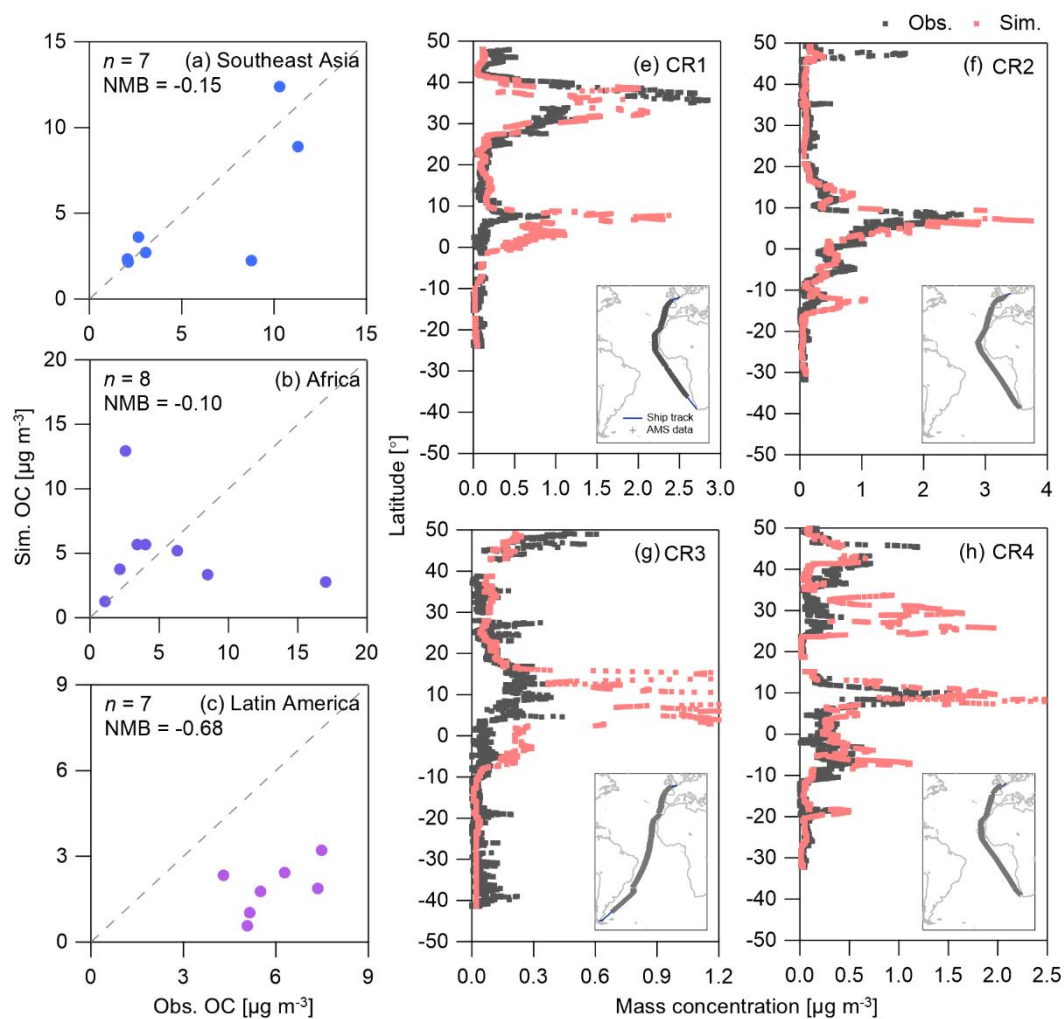


815 **Figure 4.** (a-d) Scatter plots of the observation-derived and simulated (“Base”) concentrations of BBOA in Europe in the four seasons. (e) The box-and-whisker plots of the observed and simulated monthly OC concentrations in Europe. The upper and lower edges of the boxes, the whiskers, the middle lines, and the solid dots denote the 25th and 75th percentiles, the 10th and 90th percentiles, the median values, and the mean values of OC concentrations, respectively.

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**Figure 5.** (a-c) Scatter plots of the observed and simulated (“Base”) mass concentrations of OC in Southeast Asia, Africa, and Latin America. (e-h) Comparisons of the observation-derived and simulated latitudinal profile of non-marine SOA from four shipborne campaigns, CR1 to CR4, in the Atlantic Ocean.

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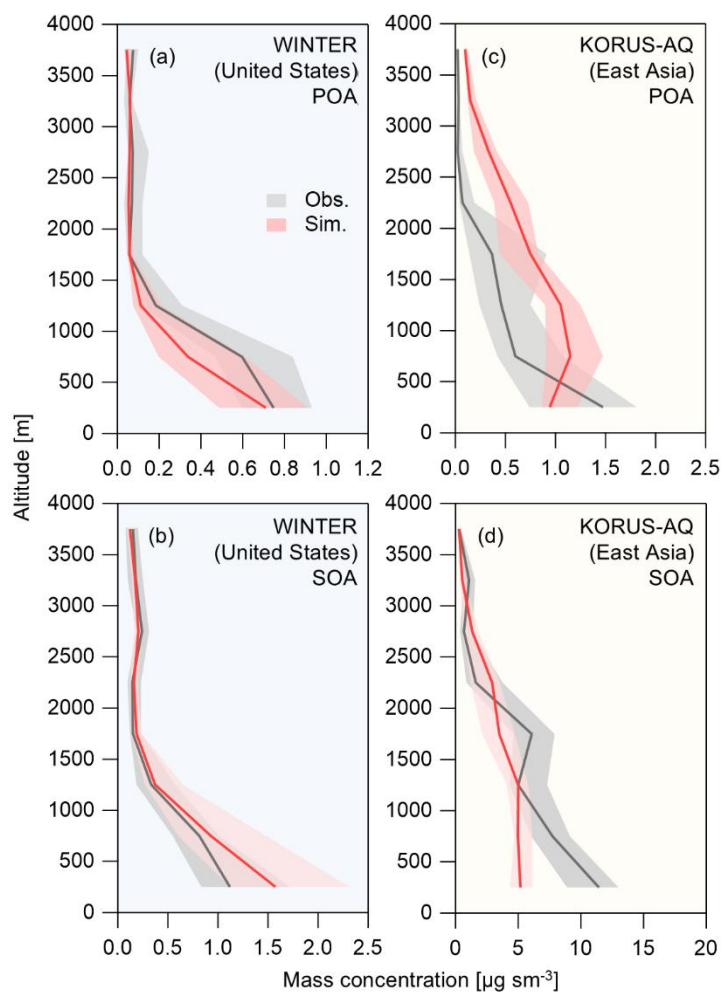


Figure 6. Comparisons of the observation-derived and simulated (“Base”) vertical profiles of POA and SOA concentrations for the WINTER and KORUS-AQ aircraft campaigns.

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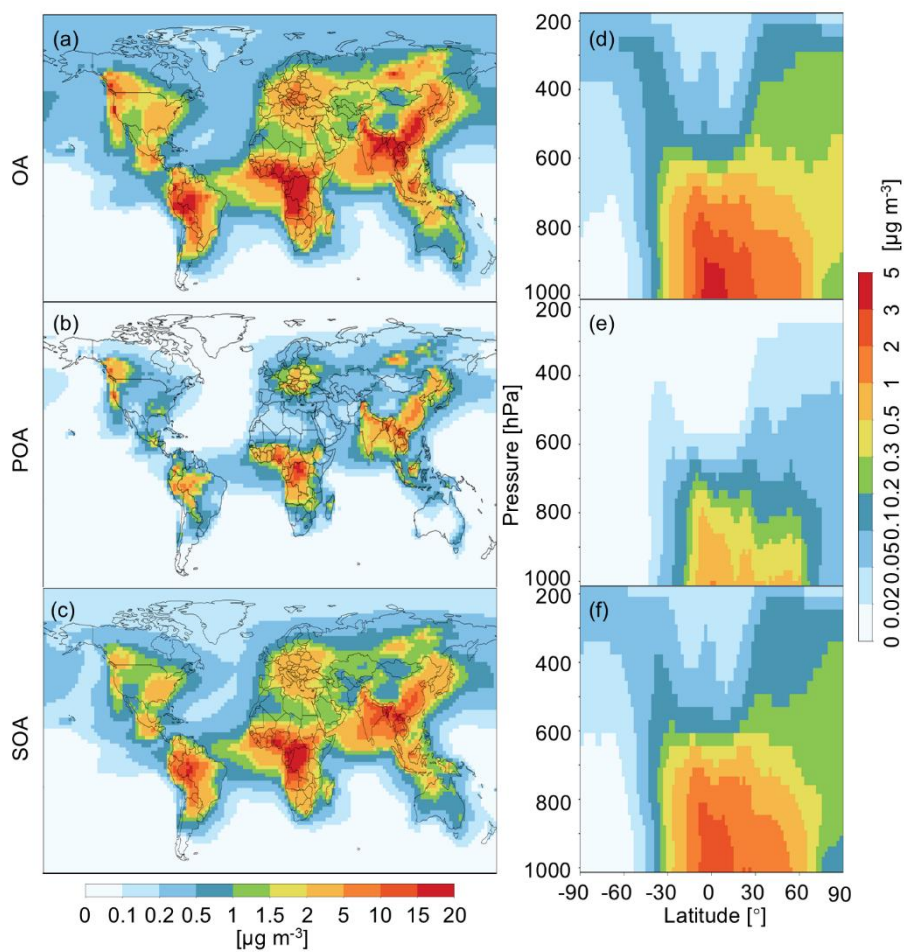
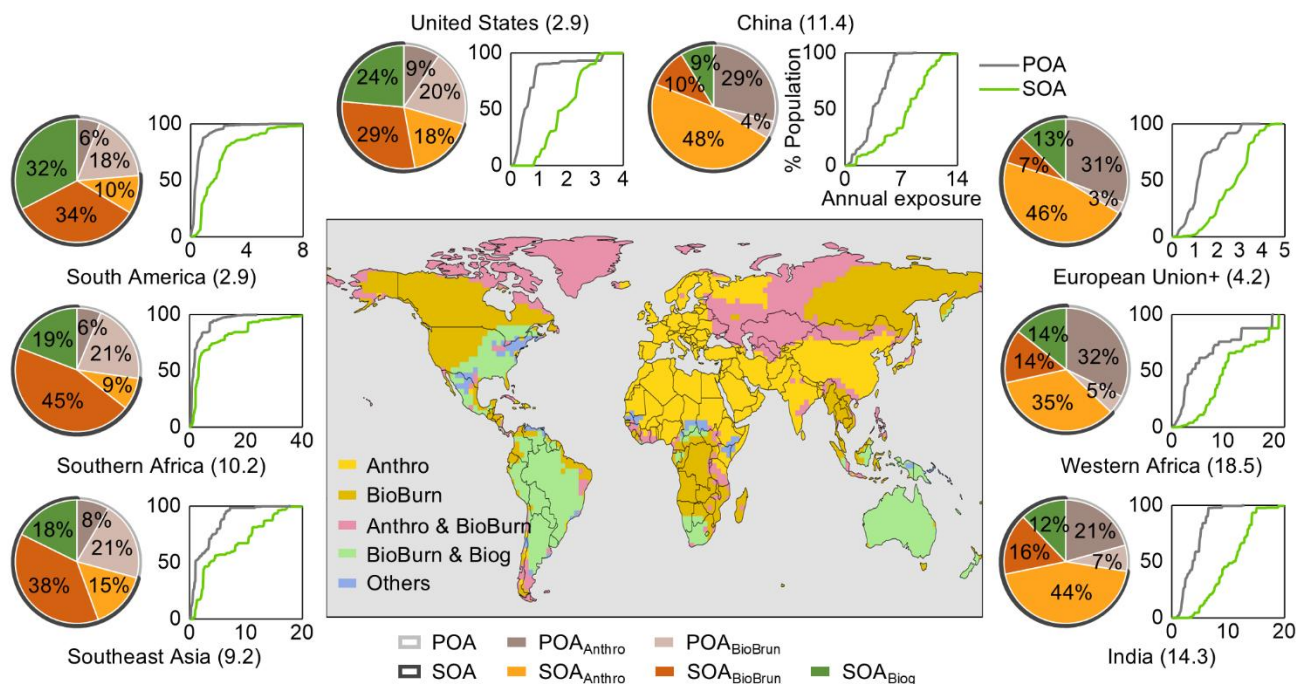


Figure 7. Simulated annual-mean (a-c) surface and (d-f) zonal mean concentrations of OA, POA, and SOA in the year of 2018 from the full-volatility-range OA runs (“Base”).

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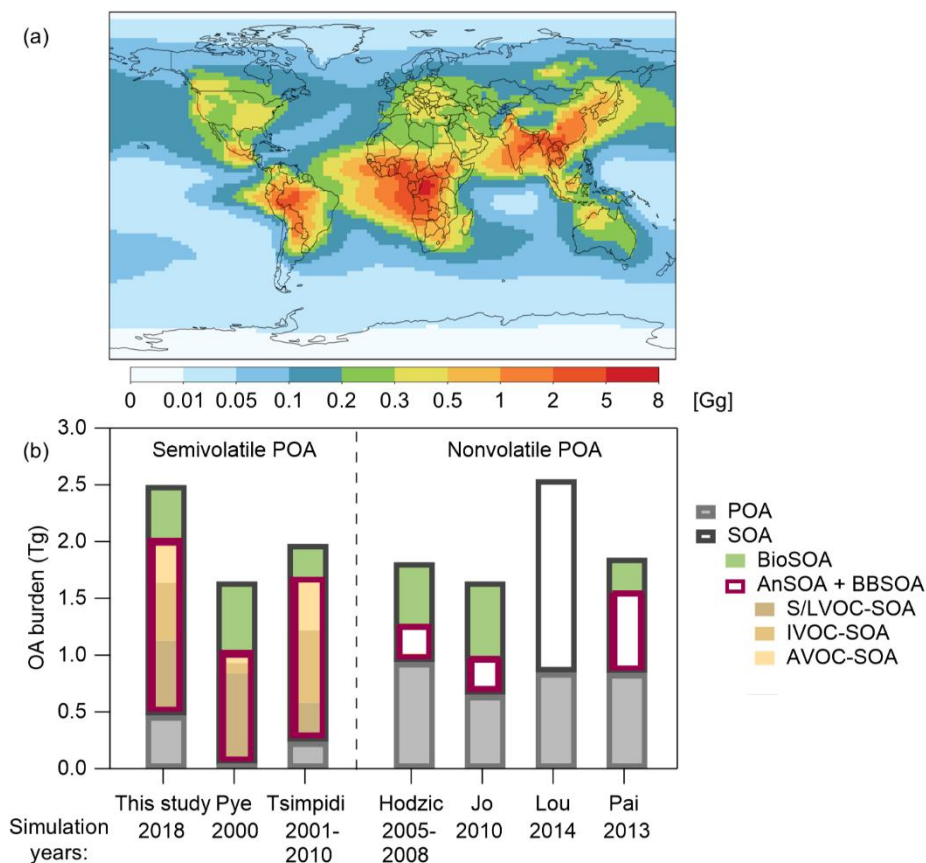


860 **Figure 8. Sources that contribute to over 30% of annual-mean surface OA concentrations in 2018. Pie charts illustrate the mass**  
**fractions of different types of POA and SOA in population-weighted OA concentrations, and cumulative distributions represent**  
**the population percentages exposed to POA and SOA concentrations (in the unit of  $\mu\text{g m}^{-3}$ ) in different regions and countries. The**  
**numbers in parentheses indicate population-weighted OA concentrations. “Anthro”, “BioBurn”, and “Biog” represent**  
**anthropogenic, open biomass burning, and biogenic sources, respectively. European Union+ encompasses the 27 member states of**  
 865 **European Union, along with Norway, Switzerland, and Liechtenstein. Southern America includes Argentina, Bolivia, Brazil, Chile,**  
**Colombia, Ecuador, Guyana, Paraguay, Peru, Suriname, Uruguay, and Venezuela. Western Africa includes Benin, Burkina Faso,**  
**Cape Verde, The Gambia, Ghana, Guinea, Guinea-Bissau, Ivory Coast, Liberia, Mali, Mauritania, Niger, Nigeria, Senegal, Sierra**  
**Leone, and Togo. Southern Africa includes Angola, Botswana, Comoros, Congo, Eswatini, Lesotho, Madagascar, Malawi,**  
**Mauritius, Mozambique, Namibia, Seychelles, South Africa, Tanzania, Zambia, and Zimbabwe.**

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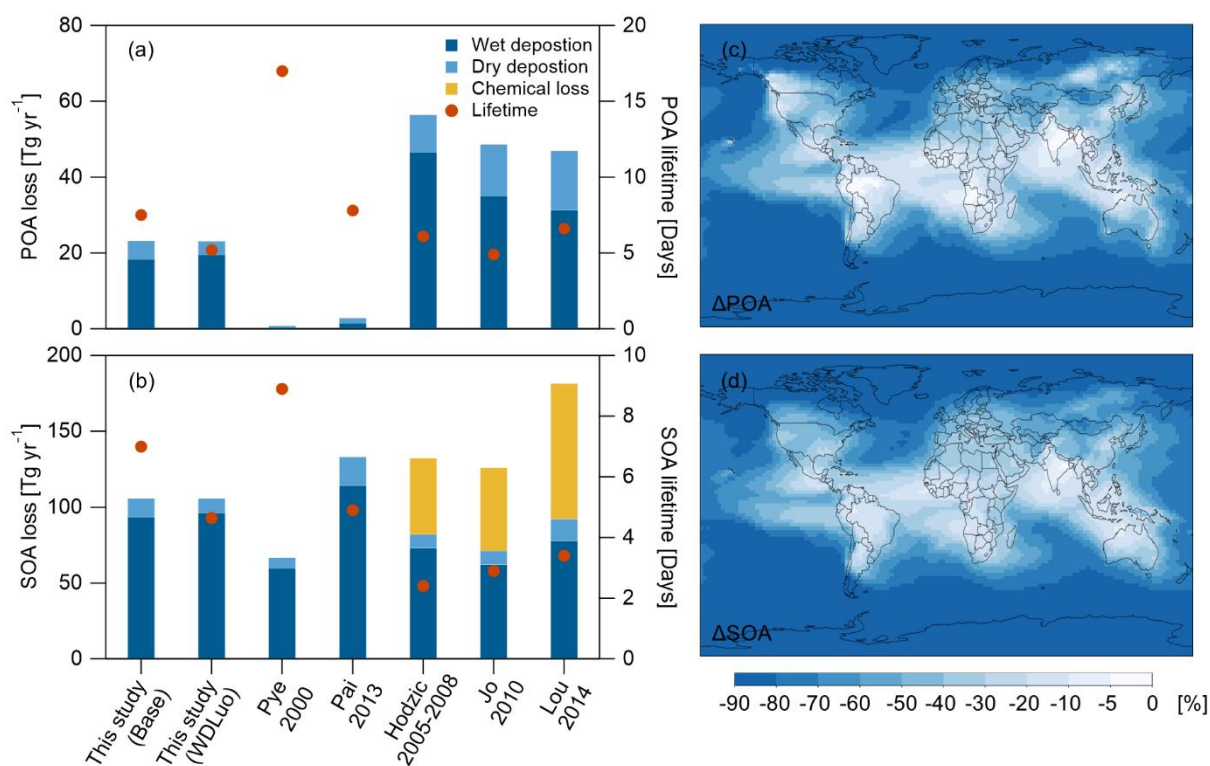
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880 **Figure 9. (a) Simulated global OA burden in 2018 (“Base”). (b) Comparisons of global OA burden estimates, including the “Revised Simulation” from Pye and Seinfeld (2010), “NY\_DPH run” from Hodzic et al. (2016), “CAM-Chem simulation” from Jo et al. (2023), “FRAG50\_PHO” from Lou et al. (2020), and “Simple scheme” Pai et al. (2020). The simulation years for all the studies are shown.**



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Figure 10. (a-b) Comparisons of the estimated global loss and global mean lifetime of POA and SOA among studies. (c-d) The differences in POA and SOA burdens between the simulations with default wet deposition parameterization and the parameterization from Luo et al. (2020) (“WDLuo”– “Base”).

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**Table 1. Global budgets for OA and its components for the year 2018 (“Base”).**

	Net production (Tg yr <sup>-1</sup> )	Dry deposition (Tg yr <sup>-1</sup> )	Wet deposition (Tg yr <sup>-1</sup> )	Burden (Tg)	Lifetime (days)
<b>POA</b>	<b>23.0</b>	<b>4.8</b>	<b>18.3</b>	<b>0.5</b>	<b>7.5</b>
<b>SOA</b>	<b>105.6</b>	<b>12.3</b>	<b>93.3</b>	<b>2.0</b>	<b>7.0</b>
S/LVOC-SOA	21.0	3.6	17.4	0.7	11.5
IVOC-SOA	31.8	3.2	28.6	0.5	5.8
AVOC-SOA*	19.4	1.9	17.5	0.4	7.2
Terpene-SOA	12.4	1.6	10.8	0.2	4.5
Isoprene-SOA*	21.0	2.0	19.1	0.3	5.7
<b>OA</b>	<b>128.6</b>	<b>17.1</b>	<b>111.5</b>	<b>2.5</b>	<b>7.1</b>
Anthropogenic OA	27.6	3.7	23.8	0.7	8.6
Biogenic OA	37.0	3.6	33.4	0.5	5.3
Open biomass burning OA	64.1	9.8	54.3	1.3	7.5

\*: included the SOA formed from glyoxal and methylglyoxal.