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#### **Key Points:**

- Fifty-eight percent of wintertime organic aerosol in the northeastern U.S. is secondary organic aerosol from pollution sources
- Observed photochemical growth rate of organic aerosol is well reproduced by a simple parameterization based on past summer measurements
- Model shows that secondary organic aerosols from pollution sources are widespread making up 14% of submicron particles near surface

**Supporting Information:** 

Supporting Information S1

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# Widespread Pollution From Secondary Sources of Organic Aerosols During Winter in the Northeastern United States

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**Abstract** Secondary organic aerosol (SOA) from pollution sources is thought to be a minor component of organic aerosol (OA) and fine particulate matter beyond the urban scale. Here we present airborne observations of OA in the northeastern United States, showing that 58% of OA over the region during winter is secondary and originates from pollution sources. We observed a doubling of OA mass from SOA formation in aged emissions, with unexpected similarity to OA growth observed in polluted areas in the summer. A regional model with a simple SOA parameterization based on summer measurements reproduces these winter observations and shows that pollution SOA is widespread, accounting for 14% of submicron particulate matter in near-surface air. This source of particulate matter is largely unaccounted for in air quality management in the northeastern United States and other polluted areas.

**Plain Language Summary** Organic aerosol is a major contributor to fine particulate matter concentrations. The wintertime sources of organic aerosols in polluted areas have remained uncertain because of a lack of regional-scale measurements. We made aircraft-based observations of organic aerosols over the northeastern United States during winter. We observed that a majority of organic aerosols consist of secondary organic aerosols that form in the atmosphere from gases emitted from pollution sources. Prior to our measurements, the expectation was that wintertime pollution sources of secondary organic aerosols are minor outside urban areas. Our results show that their influence is ubiquitous over the entire eastern United States, and a better understanding of these sources can help in developing effective policies to reduce wintertime air pollution.

# 1. Introduction

Organic aerosol (OA) constitutes about half of the fine particulate matter in populated areas worldwide (Snider et al., 2016; Q. Zhang et al., 2007) and is thus responsible for a large fraction of the human health burden of particulate matter exposure (Cohen et al., 2017; Lelieveld et al., 2015). OA is directly emitted, known as primary OA (POA), or is formed through the atmospheric oxidation of precursor gases (volatile organic compounds, VOCs) producing an array of lower volatility compounds, which condense or react on particles to form secondary organic aerosol (SOA). VOCs originate from biogenic and anthropogenic sources. They react with hydroxyl radicals (OH) and other oxidants in the atmosphere and evolve through myriad intermediate products and reactions to generate compounds that can form SOA. This complex chemical system has not been fully characterized, and many atmospheric chemistry models, with older parameterizations, underestimate observed pollution SOA by a factor of 10 (Gentner et al., 2017; Hallquist et al., 2009; Hodzic et al., 2009; Tsigaridis et al., 2014; Volkamer et al., 2006). Recently proposed models have closed the measurement/model gap for pollution SOA during summer, although with significant uncertainties about the key precursors (Cappa et al., 2016; Ma et al., 2017).

©2019. American Geophysical Union. All Rights Reserved. The sources of POA and SOA precursors and the chemical processes leading to the formation of SOA vary seasonally, particularly in polluted midlatitude regions such as the northeastern United States. In summer, emissions of biogenic VOCs (BVOCs) are elevated and their oxidation to form SOA is rapid. Thus, BVOCs are a dominant regional source for summertime SOA over the northeastern United States (Spracklen et al., 2011). In winter, loss of deciduous foliage, less insolation, and low temperatures significantly decrease BVOC emissions (Guenther et al., 1995) and biogenic SOA is expected to be low. Wintertime OA comes mainly from pollution sources, with major contribution of POA from residential wood combustion (Bond, 2004; Butt et al., 2016). Colder temperatures favor the particle phase, decreasing the evaporation of POA to semivolatile organic compounds (SVOCs; Donahue et al., 2006; Robinson et al., 2007) but also increasing the condensation of less oxidized products (Stolzenburg et al., 2018), thus impacting SOA formation. SOA has lower volatility than POA and its concentrations are weakly sensitive to temperature changes (Cappa & Jimenez, 2010). Reduced insolation and water vapor concentrations in winter decrease the main source of OH and slow the formation of SOA, although other sources of OH (Ren et al., 2006) and oxidants like chlorine radicals (Thornton et al., 2010) can become important in winter.

Empirical source apportionment of OA is performed with either tracer-based methods that use emissions ratios of POA to tracers like elemental carbon (Turpin & Huntzicker, 1995) or other organic molecules (Schauer et al., 1996), or with factor analysis of the OA mass spectra time series (Q. Zhang et al., 2011). Summertime studies in polluted areas have found consistent estimates for the POA and SOA fractions with these methods (Aiken et al., 2009; Docherty et al., 2008). Wintertime OA source apportionment studies over the northeastern United States have shown that the fraction of OA from pollution SOA is high (~50%) at the urban scale (Jimenez et al., 2009; Polidori et al., 2006; Rattigan et al., 2010; Schroder et al., 2018; Weimer et al., 2006), but at the regional scale much of the OA is thought to be POA from residential wood combustion (Schichtel et al., 2017). At the rural sites in the Interagency Monitoring of Protected Visual Environments (IMPROVE) network, 7-21% of the wintertime organic carbon was estimated to be secondary using the elemental carbon-tracer method (Yu et al., 2007). Similar fractions of SOA were calculated with tracer-based methods for other rural and suburban sites in the eastern and midwestern United States (Chen et al., 2010; Kleindienst et al., 2007; Lewandowski et al., 2008; Piletic et al., 2013; Sunder & Hopke, 2007; Y. Zhang et al., 2009). Air quality models with older SOA parameterizations also predict low contributions from pollution SOA (Foley et al., 2010; Ridley et al., 2018). In contrast, our observations of OA in winter demonstrate that pollution SOA accounts for 58% of the OA, which can be well simulated with a simple parameterization based on past summer observations in polluted areas. Furthermore, our model simulations show that the influence of pollution SOA is ubiquitous over the region.

# 2. Observations and Model Used in This Study

#### 2.1. WINTER Measurements

Between 1 February and 15 March 2015, we collected extensive observations of OA and other chemical species during the Wintertime Investigation of Transport, Emissions, and Reactivity (WINTER) campaign on 13 research flights (RF, Figure S1 in the supporting information) with the U.S. National Science Foundation/National Center for Atmospheric Research C-130 research aircraft. The flights took place in daytime and nighttime, and the majority (71%) of measurements were made below an altitude of 1 km. Concentrations of OA and inorganic species (sulfate, SO<sub>4</sub><sup>2-</sup>, nitrate, NO<sub>3</sub><sup>-</sup>, and ammonium, NH<sub>4</sub><sup>+</sup>) in submicron particulate matter (PM<sub>1</sub>, particulate matter of diameter  $< 1 \mu m$ ) were measured with a highresolution time-of-flight aerosol mass spectrometer (HR-ToF, Aerodyne Research Inc.; DeCarlo et al., 2006; Schroder et al., 2018). The instrument setup, operation, and calibration during the WINTER campaign are described in detail by Schroder et al. (2018). Briefly, nonrefractory species are flash vaporized in the instrument at 600 °C under high-vacuum and ionized by 70-eV electrons. The ions are orthogonally extracted and analyzed by HR-ToF mass spectrometry. One-minute integrated measurements are used in our analysis. Detection limits for the 1-min measurements were 194  $ng/sm^3$  (OA), 57  $ng/sm^3$  (SO<sub>4</sub><sup>2-</sup>), 30 ng/sm<sup>3</sup> (NO<sub>3</sub><sup>-</sup>), and 5 ng/sm<sup>3</sup> (NH<sub>4</sub><sup>+</sup>). The instrument accuracy ( $2\sigma$ ) was estimated as 38% (OA) and 35% (inorganics; Bahreini et al., 2009). The instrument was not operational on flight RF05. POA and oxidized OA (OOA) concentrations were inferred using positive matrix factorization (PMF) of the aerosol mass spectrometer 1-min observations for six flights (RF01-RF04, RF06, and RF07) as described by Schroder et al. (2018). The PMF factors represent fossil fuel and cooking POA (referred to as hydrocarbon-like OA, HOA), wood combustion POA (biomass burning OA, BBOA), and OOA—which has been shown to be a good surrogate for SOA (Jimenez et al., 2009; Schroder et al., 2018). CO was measured by vacuum ultraviolet fluorescence (Aero-Laser AL-5002), with an accuracy of 3%. The CO instrument was not operational on flight RF06.

#### 2.2. GEOS-Chem Model

We use the GEOS-Chem chemical transport model v10-01 (www.geos-chem.org) driven by assimilated meteorological fields from the National Aeronautics and Space Administration Global Modeling and Assimilation Office's GEOS-5 Forward Processing system (Reinecker et al., 2008). We use the one-way nested-grid configuration with a resolution of 0.5° latitude by 0.625° longitude over North America (10- $60^{\circ}$ N,  $60-130^{\circ}$ W) with initial and boundary conditions from a  $4^{\circ} \times 5^{\circ}$  global simulation (Kim et al., 2015; Wang et al., 2004). GEOS-Chem includes gas-phase chemistry of HO<sub>x</sub>-NO<sub>x</sub>-VOC-O<sub>3</sub>-SO<sub>2</sub>-BrO<sub>x</sub> coupled with the chemistry of  $SO_4^{2-}$ ,  $NO_3^{-}$ ,  $NH_4^+$  particles, and OA (Bey et al., 2001; Fisher et al., 2016; Mao et al., 2010; Park et al., 2003, 2004; Parrella et al., 2012; Pye et al., 2009; Travis et al., 2016). The simulation is performed for 1 February to 15 March 2015 with a spin-up period of 6 months for the global simulation and 15 days for the high-resolution North America simulation. For comparison to observations, the model is sampled at the time and location of the measurements. We use GEOS-Chem to calculate the OH equivalent age of the emissions, as described in the supporting information (Text S1). Jaeglé et al. (2018) and Shah et al. (2018) describe in detail the GEOS-Chem simulations for the WINTER campaign. Jaeglé et al. (2018) found a 65% overestimate of CO emissions in the U.S. Environmental Protection Agency's 2011 National Emissions Inventory (NEI 2011) during WINTER. Thus, for the GEOS-Chem simulation in this study we have lowered CO emissions by 40% (i.e., 1/1.65).

GEOS-Chem simulates the emissions of POA from pollution and fire sources and of SOA formation from biogenic, pollution, and fire emissions (Heald et al., 2011; Kim et al., 2015; Park et al., 2003; Pye et al., 2010; Pye & Seinfeld, 2010). Pollution emissions over the United States are from the NEI 2011 inventory. For the WINTER simulations, the NEI 2011 POA emissions are reduced by 50% based on initial comparisons with the WINTER observations (Schroder et al., 2018). SOA formation from pollution sources follows the SIMPLE (SIMPLified parameterization of combustion SOA) method described in Hodzic and Jimenez (2011). In this parameterization, a surrogate SOA precursor gas is emitted in proportion to CO emissions from nonpoint sources, with a ratio of 80 g (kg CO)<sup>-1</sup>. The surrogate precursor gas is oxidized by OH with a first-order rate constant of  $1.25 \times 10^{-11}$  cm<sup>3</sup> per molecule per second and forms nonvolatile SOA with a yield of 100%. Details of the implementation of the SIMPLE parameterization in GEOS-Chem are described in the supporting information (Text S2).

# 3. Results and Discussion

#### 3.1. OA Observations During the WINTER Campaign

The mean OA concentration observed during the WINTER flights below 1-km altitude was  $1.48 \pm 0.93 \mu g/$  sm<sup>3</sup> (campaign mean  $\pm$  standard deviation; Figure 1a). (1 sm<sup>3</sup> equals 1 m<sup>3</sup> at 273.15 K and 1,013.25 hPa.) OA accounted for 38% of the observed PM<sub>1</sub>, more than the individual contributions of SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, or NH<sub>4</sub><sup>+</sup>. Higher OA concentrations (3–4  $\mu g/$ sm<sup>3</sup>) were observed near pollution sources in the Ohio River Valley (Region 1, Figure 1), New York City (Region 2), and the Washington-Baltimore area (Region 3). We find that a large fraction of the observed OA is oxidized. The median OOA fraction (OOA/OA) for the six analyzed flights was 0.58 (Figure 1c). OOA/OA varied moderately during the campaign (interquartile range of 0.17), indicating that SOA is a major constituent of OA not only in urban areas, as previously observed (Jimenez et al., 2009; Polidori et al., 2006; Rattigan et al., 2010; Weimer et al., 2006) but over a much larger regional scale.

The observations show a pattern in the variation of OOA/OA. OOA/OA is lower (0.5) close to urban sources in the Ohio River Valley and higher (0.6–0.8) for observations farther downwind of New York and Washington-Baltimore (Figure 1). We expect OOA/OA to increase with distance from a source as VOCs are slowly oxidized during downwind transport. We now examine this observed change in OA as a function of photochemical oxidation of emitted VOCs.





Figure 1. Spatial distribution of organic aerosol (OA) concentrations and of the contribution of oxidized OA (OOA-a proxy for secondary OA) during WINTER (1 February to 15 March 2015). OA concentrations (a) observed and (b) modeled for WINTER flights below 1 km altitude. The GEOS-Chem model is sampled along the flight tracks. Observed and modeled concentrations are binned onto the  $0.5^{\circ}$  latitude  $\times 0.625^{\circ}$  longitude GEOS-Chem grid. The campaign mean ± standard deviation of OA concentration is shown above each panel. (c) Fraction of OA composed of OOA (observations) and SOA (GEOS-Chem) for six flights (RF01-RF04, RF06, and RF07). The box represents the 25th, 50th, and 75th percentile values, the whiskers extend to 1.5 times the interquartile range beyond the 25th and 75th percentile values, and the circles show the outliers (<2% of the data). Also shown are the OOA/OA and SOA/OA fractions for the Ohio River Valley (1), downwind of New York City (2), and downwind of Washington-Baltimore area (3). Observed and modeled vertical profiles of OA (d), sum of observed HOA (hydrocarbon-like OA) and BBOA (biomass burning OA) compared to GEOS-Chem POA (primary OA) (e), observed OOA compared to GEOS-Chem SOA (f) for the WINTER campaign. Panel (d) includes all 12 WINTER flights when OA observations were available, while panels (e) and (f) include the six flights for which positive matrix factorization was available. The observation-model pairs are grouped into altitudes bins of width 0.2-1 km, and the mean ± standard deviation are shown for the observations (black diamonds and error bars) and the model (orange line and shading). WINTER = Wintertime Investigation of Transport, Emissions, and Reactivity; HOA = hydrocarbon-like organic aerosol; BBOA = biomass burning organic aerosol; POA = primary organic aerosol; SOA = secondary organic aerosol; OOA = oxidized organic aerosol; AMS = aerosol mass spectrometer.

#### 3.2. Atmospheric Evolution of OA Downwind of Emission Regions

In order to examine how observed OA and SOA evolve downwind of emission regions, we first control for the variability in OA caused by spatial differences in emissions strengths, atmospheric transport, and dilution. We do so by considering the enhancement ratios of OA and carbon monoxide, CO, ( $\Delta$ OA/ $\Delta$ CO) for the WINTER observations (DeCarlo et al., 2010; Kleinman et al., 2007, 2008). Indeed, CO is expected to be coemitted (or emitted from sources closely collocated) with POA and SOA precursors from combustion and other pollution sources. We express the degree of photochemical oxidation of VOCs as the OH equivalent age, which is the equivalent exposure time of the pollution emissions to a reference OH concentration of  $1.5 \times 10^6$  molecules per cubic centimeter. The OH equivalent age is 0 for fresh emissions and increases monotonically as emissions age in the atmosphere. Therefore, the growth of  $\Delta$ OA/ $\Delta$ CO in aged emissions reflects the amount of new OA and SOA formed from the oxidation of VOCs as the plume dilutes into the regional background. We calculate the OH equivalent age from the GEOS-Chem simulated concentrations of two species along the WINTER flights (see Text S1). We group the aircraft observations by their OH





Figure 2. Photochemical evolution of OA and OOA observed during WINTER flights. (a)  $\Delta OA/\Delta CO$  and (b)  $\Delta OOA/\Delta CO$  as function of the OH equivalent age (photochemical age at an OH concentration of  $1.5 \times 10^{\circ}$  molecules per cubic centimeter) for the WINTER observations below 1-km altitude. Panel (a) includes observations from the 11 flights in which the aerosol mass spectrometer and CO instruments were operational, and panel (b) includes observations from 5 of the 11 flights for which positive mass factorization was available. Observations are grouped into 12 OH equivalent age bins, and the resulting  $\Delta OA/\Delta CO$  and  $\Delta OOA/\Delta CO$ slopes are calculated using reduced major axis regression (see Text S1). The regression slopes (filled circles) and their 95% confidence intervals (error bars) for each age bin are shown. The slope of the best fit lines (solid black) are indicated.  $\Delta POA/\Delta CO$  is calculated without binning the observations by equivalent age and is marked with the dashed line. POA is the sum of HOA and biomass burning organic aerosol factors from positive matrix factorization. The best fit lines for the GEOS-Chem  $\Delta OA/\Delta CO$  and  $\Delta$ SOA/ $\Delta$ CO are also shown (orange). The blue shaded area shows the range of  $\Delta OA/\Delta CO$  ratios observed in past spring and summer studies in the northeastern United States and other polluted areas (de Gouw & Jimenez, 2009). WINTER = Wintertime Investigation of Transport, Emissions, and Reactivity; OA = organic aerosol; POA = primary organic aerosol; HOA = hydrocarbon-like organic aerosol; OOA = oxidized organic aerosol.

equivalent age and calculate  $\Delta OA/\Delta CO$  for each age bin using linear least squares regression (Kleinman et al., 2008).

During WINTER, we find that  $\Delta OA/\Delta CO$  increased substantially in aged emissions, because of the formation of new SOA (Figure 2a). The observed  $\Delta OA/\Delta CO$  increased from 45 to 85  $\mu$ g·sm<sup>-3</sup>·ppmv<sup>-1</sup> as the OH equivalent age of the emissions increases from 2 hr to 1 equivalent day (eq. day), implying a (best fit) growth rate of  $\Delta OA/\Delta CO$  of 57  $\mu g \cdot sm^{-3} \cdot ppmv^{-1}$  eq. day<sup>-1</sup>. Note that because wintertime OH concentrations are much lower than the  $1.5 \times 10^6$  molecules per cubic centimeter, 1 eq. day corresponds to about 3 days of atmospheric processing. The  $\Delta OA/\Delta CO$  at the time of emissions (the y intercept of the best fit line) is  $28 \ \mu g \cdot sm^{-3} \cdot ppmv^{-1}$  and is similar to the  $\Delta POA/\Delta CO$  inferred from PMF.  $\Delta OOA/\Delta CO$  increased at a rate of 66  $\mu$ g·sm<sup>-3</sup>·ppmv<sup>-1</sup> eq. day<sup>-1</sup>, mirroring the growth in  $\Delta OA/\Delta CO$  (Figure 2b). Similar OOA growth rates were observed in a New York City plume that was sampled on one WINTER flight (Schroder et al., 2018). Figure 2 shows that these SOA formation rates are not limited to concentrated urban plumes but are prevalent over the entire region. WINTER observations of oxidized VOCs (oVOCs) with the HR-ToF chemical ionization mass spectrometer (Lee et al., 2018; HRToF) show significant amounts of secondary, more oxidized, low volatility, anthropogenic oVOCs, which are consistent with the high concentration of pollution SOA (Figure S2).

Figure 2 also shows a comparison of the WINTER campaign to past spring and summer observations of  $\Delta OA/\Delta CO$  in the northeastern United States and other polluted regions (de Gouw & Jimenez, 2009; DeCarlo et al., 2010; Hayes et al., 2013, 2015; Kleinman et al., 2007, 2008). Note that we have controlled for the differences in emissions, dilution, and oxidation by normalizing to CO enhancements and using an OH equivalent age. de Gouw and Jimenez (2009) summarized several OA measurements in polluted urban areas and found mean  $\Delta OA/\Delta CO$ in fresh emissions of 9  $\mu g \cdot sm^{-3} \cdot ppmv^{-1}$  and increasing to a mean of 75  $\mu$ g·sm<sup>-3</sup>·ppmv<sup>-1</sup> for aged emissions. The WINTER POA/ $\Delta$ CO in fresh emissions is a factor of 2-3 higher than the summertime values likely because of the large POA emissions from residential wood combustion during winter. Indeed, the U.S. NEI (U.S. Environmental Protection Agency, 2018) and the WINTER observations (Schroder et al., 2018) indicate that residential wood combustion makes up about three quarters or more of the wintertime POA emissions in the northeastern United States.

The growth in  $\Delta OA/\Delta CO$  observed during WINTER is remarkably similar to the growth observed in past summertime studies in polluted urban areas. The influence of BVOCs was minor during those studies (Hayes et al., 2015) as was the case during WINTER. But other seasonal differences appear to compensate each other resulting in a similar net  $\Delta OA/\Delta CO$  formation rate in winter and summer. During winter, less evaporation of POA may reduce SOA formation from SVOCs but increased formation from intermediate volatility organic compounds (IVOCs)

(Donahue et al., 2006; Robinson et al., 2007) and more condensation of less oxidized gases facilitates SOA formation (Stolzenburg et al., 2018). Laboratory-based studies find that residential wood combustion emits large amounts of aromatic VOCs, which is an additional source of SOA precursors in winter (Bruns et al., 2016), although many field studies do not report a net increase of  $\Delta OA/\Delta CO$  in biomass burning plumes (Cubison et al., 2011; Shrivastava et al., 2017). Gasoline vehicles emit several times more VOCs at colder temperatures and may therefore produce more SOA in winter but their CO emissions are also higher in winter (Platt et al., 2017).



**Figure 3.** Mean GEOS-Chem simulated concentrations of (a) OA and (b)  $PM_1$  below 1-km altitude for 1 February to 15 March 2015. The black contour lines in panel (a) show the percentage contribution of pollution SOA to OA, and the black contour lines in panel (c) show the percentage contribution of OA to  $PM_1$ . Note the different color scales for panels (a) and (b). (c) The mean (1 February to 15 March 2015) simulated composition of  $PM_1$  below 1-km altitude over the northeastern United States (dashed rectangle in panel b). The components are primary OA (POA), pollution SOA, biogenic SOA, sulfate (SO<sub>4</sub><sup>2-</sup>), nitrate (NO<sub>3</sub><sup>-</sup>), and ammonium (NH<sub>4</sub><sup>+</sup>). OA = organic aerosol; SOA = secondary organic aerosol.

#### 3.3. Wintertime Distribution of Pollution SOA Over the Northeastern United States

We find that the modeled OA, POA, and SOA concentrations agree with the observations (Figure 1), demonstrating that sources, transport, and deposition of OA are well represented in the model and that SOA formed photochemically from pollution sources is a major source of OA during WINTER. The photochemical growth of modeled  $\Delta OA/\Delta CO$  and  $\Delta SOA/\Delta CO$  with OH equivalent age is similar to the observations but does display some minor differences due to the model underestimate in the observed variability of CO concentrations (Figures 2, S3, and S4). A detailed comparison of the GEOS-Chem simulation for other gas and aerosol species for WINTER showed that the model reproduces the observed ozone (O<sub>3</sub>), nitrogen oxides (NO<sub>x</sub>), total reactive nitrogen (NO<sub>y</sub>), and inorganic aerosol species (Jaeglé et al., 2018; Shah et al., 2018). In the northeastern United States, in the bottom 1 km of the atmosphere, the GEOS-Chem model predicts that pollution SOA accounts for 56% of OA and that OA contributes to 25% of the PM<sub>1</sub> mass, as much as SO<sub>4</sub><sup>2-</sup> (Figure 3). Thus, the model predicts that pollution SOA accounts for 35% of the observed PM<sub>1</sub> concentrations in the region in winter (Figure S5). GEOS-Chem predicts that on average 40% of winter-time OA at these sites is SOA.

The distribution of OA concentrations simulated for 1 February to 15 March 2015 shows that OA is not confined to the populated areas where most emissions occur, but is widespread over the northeastern United States (Figure 3a). This is because a large fraction (35–60%) of the OA over the region is from pollution SOA, which forms gradually during the regional transport of VOC emissions away from the sources. The lifetime of OA against deposition to the Earth's surface is a few days (Tsigaridis et al., 2014), long enough to allow regional-scale transport. In the southeastern United States, higher OH concentrations (by a factor of 3) and a larger contribution of fossil fuel sources result in a greater SOA/OA fraction (60–70%). Current air quality models, which often simulate too little SOA from pollution sources, may thus systematically underestimate OA by factors of 2–3 and underestimate their contribution to PM<sub>1</sub>. Often, these SOA underestimates are not apparent due to an overestimate in the concentrations of POA (de Gouw & Jimenez, 2009; Schroder et al., 2018).

The lower regional SOA/OA fractions reported by previous tracer-based apportionment studies suggest that the POA/tracer emissions ratios used in those studies may be too high. Indeed, we reduced the wintertime NEI OA emissions by 50% to better match the WINTER measurements of HOA + BBOA. Moreover, it is difficult to estimate POA/tracer ratios from ambient measurements in polluted regions, because of the significant fractions of SOA/OA present even during winter, as shown here.

### 4. Conclusions

We have shown here that OA is widespread over the wintertime northeastern United States and that about half of wintertime OA is from secondary OA formed from the oxidation of precursor gases emitted from pollution sources. Past decreases in OA over the United States might have been driven by decreases in POA emissions (Ridley et al., 2018), but our results imply that further decrease in wintertime OA can be achieved by reductions in pollution SOA precursors. Our results suggest the need for further speciation of wintertime VOCs and simulation experiments to evaluate the efficiency of SOA formation from known anthropogenic VOC emissions, such as aromatics and alkanes, under conditions of the wintertime atmosphere. Moreover, there is a need for measurements of SVOCs and IVOCs that are missing from most models and are thought to be important contributors to pollution SOA (Hayes et al., 2015; Hodzic et al., 2010; Robinson et al., 2007). Wintertime PM<sub>1</sub> pollution is a problem in Europe, China, and India and is characterized by high SOA (Crippa et al., 2014; Sun et al., 2016). Our results suggest that pollution sources of SOA may be important in these areas, too. Efforts to mitigate poor air quality around the world will benefit from the identification and characterization of these pollution sources of SOA precursors.

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