Understanding global secondary organic aerosol amount and size-resolved condensational behavior

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Summary

• Secondary organic aerosols (SOA) are major contributors to ultrafine particle growth to CCN-sizes particles.

• Uncertainty #1: Recent studies show that an additional source of SOA of 100 Tg (SOA) yr⁻¹ is required to match measurements (Spracklen et al., 2011).

• Uncertainty #2: Many models treat SOA as semivolatile with condensation onto the aerosol mass distribution; however, recent closure studies show a significant fraction of SOA condensing to aerosol surface area (Riipinen et al., 2011).

1. Amount of SOA is very uncertain

Top-down estimates suggest a global source ranging from 12-18 Tg (SOA) yr⁻¹, meanwhile atmospheric models using bottom-up estimates suggest SOA formation of 12-70 Tg (SOA) yr⁻¹. Therefore the uncertainty in the global SOA source is very uncertain with ranges of 12-18 Tg (SOA) yr⁻¹ (Spracklen et al., 2011) (Figure 1).

By comparing GLOMAP model simulations to AMS measurements, Spracklen et al. (2011) were able to close the measurement-model gap by adding 100 Tg (SOA) yr⁻¹ of SOA correlated with anthropogenic CO emissions to the model simulations. In this way, we will use this additional 100 Tg (SOA) yr⁻¹ of additional SOA (correlated with anthropogenic CO) in the model simulations.

2. SOA condensation to surface area or mass

Freshly nucleated (~1 nm) particles must grow to CCN sizes in order to affect climate (Figure 2). To model the growth of these particles the condensation of SOA to the particles must be assessed. Many models treat SOA solely as semi-volatile (C* > 0.2). However, recent closure studies with field measurements show that a significant fraction of SOA condenses to the aerosol surface area, which suggests a very low volatility (C* ≈ 10⁻³ mg m⁻³) (Figure 3).

3. Model Description

• 3D atmospheric model: GEOS-Chem (www.geos-chem.org)

• GEO-10 m topographical fields

• 4×5° horizontal resolution, 30 vertical layers (from the surface to 0.01 mb)

4. Additional SOA increases CCN number

Figure 4. Simulations for model year 2001 of the percent change of N10 in the BL between the additional SOA correlated to the anthropogenic CO emissions and the base case emissions both using the surface area condensation scheme (SURF-XXSOA SURF-BASE). The additional SOA yields a 27% global BL decrease in N10.

Figure 6. Simulations for model year 2001 of the percent change of N10 in the continental boundary layer (BL) between condensation of SOA proportional to the surface area compared to the mass distribution (SURF-XXSOA – MASS-BASE). The surface area condensation scheme yields a 3% global BL increase in N10 over the mass distribution scheme.

5. Surface area increases CCN number

Figure 5. Simulations for model year 2001 of the percent change of N80 in the BL between the additional SOA correlated with anthropogenic CO emissions and the base case emissions both using the surface area condensation scheme (SURF-XXSOA SURF-BASE). The additional SOA yields a 30% global BL increase in N80, however in some regions with high anthropogenic CO emissions the increase in N80 exceeds 100%.

6. Surface area condensation with the additional SOA performs best

Figure 7. Simulations for model year 2001 of the percent change of N80 in the BL between the additional SOA compared with anthropogenic CO emissions and the base case emissions both using the surface area condensation scheme (SURF-XXSOA SURF-BASE). The additional SOA yields a 30% global BL increase in N80, however in some regions with high anthropogenic CO emissions the increase in N80 exceeds 100%.

7. References


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