

1 **Using a global aerosol model adjoint to unravel the footprint of spatially-**
2 **distributed emissions on cloud droplet number and cloud albedo**

3 **V.A. Karydis¹, S.L. Capps², R.H. Moore³, A. Russell⁴, D. K. Henze⁵ and A. Nenes^{1,2*}**

4 ¹ Earth and Atmospheric Sciences, Georgia Institute of Technology, Atlanta, GA

5 ² Chemical and Biomolecular Engineering, Georgia Institute of Technology, Atlanta, GA

6 ³ NASA Langley Research Center, Hampton, VA

7 ⁴ Civil and Environmental Engineering, Georgia Institute of Technology, Atlanta, GA

8 ⁵ Department of Mechanical Engineering, University of Colorado, Boulder, CO

9 *Corresponding author: athanasios.nenes@gatech.edu

10 **Abstract**

11 The adjoints of the GEOS-Chem Chemical Transport Model and a comprehensive cloud
12 droplet parameterization are coupled to study the sensitivity of cloud droplet number
13 concentration (N_d) over US regions and Central Europe to global emissions of anthropogenic fine
14 aerosol precursors. Simulations reveal that the N_d over the midwestern and southeastern US is
15 mostly sensitive to SO₂ emissions during August, and to NH₃ emissions during February. Over
16 the western US, N_d is mostly sensitive to SO₂ and primary organic aerosol emissions. In Central
17 Europe, N_d is most sensitive to NH₃ and NO_x emissions. As expected, local emissions strongly
18 affect N_d ; long-range transport, however, is also important for the western US and Europe.
19 Emissions changes projected for the year 2050 are estimated to have the largest impacts on cloud
20 albedo and N_d over Central Europe during August (42% and 82% change, respectively) and
21 western US during February (12% and 36.5% change, respectively).

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24 **1. Introduction**

25 Cloud droplets form upon pre-existing atmospheric aerosols, and their modulation from
26 anthropogenic emissions has profound impacts on cloud radiative properties, the hydrological
27 cycle and climate. The impact of aerosol on regional climate can be even stronger, owing to the
28 variability of aerosol and regional climate sensitivity [*Shindell and Faluvegi, 2009*]. Attribution
29 of climate forcing from sector-specific emissions is based on sequential perturbation calculations
30 at the cost of a climate run per sector investigated [*Shindell et al., 2009*]. This approach is
31 frequently limited by addressing only one sector (e.g., transport [*Fuglestvedt et al., 2008*]) or by
32 invoking approximations required to reduce the computational burden (e.g., constant oxidant
33 levels [*Koch et al., 2007*]). Adjoint modeling provides an efficient sensitivity analysis alternative
34 to brute-force approaches and does not require perturbations in emissions. This is accomplished
35 by propagating a differential variation of a model output (e.g., aerosol concentration) backwards
36 through the model to express its sensitivity with respect to inputs of interest (e.g., emissions)
37 without perturbing the model state.

38 The GEOS-Chem chemical transport model (CTM) is a widely used global model for
39 studying chemistry-aerosol-climate interactions [*Leibensperger et al., 2012a; b*]. *Henze et al.*
40 [2007] developed the adjoint of the code and since has been used e.g., to study black carbon over
41 the Tibetan Plateau [*Kopacz et al., 2010*]. Here the GEOS-Chem adjoint is coupled together with
42 the adjoint of a comprehensive droplet parameterization [*Karydis et al., 2012*] to quantify the
43 influences of global emissions of primary organic aerosol (POA) and inorganic aerosol
44 precursors (SO_2 , NO_x , and NH_3) on the predicted cloud droplet number concentration (N_d) and
45 cloud albedo (A) within three regions of the continental US and Central Europe during August
46 and February for 2008 and 2050.

47 2. Model framework description

48 The GEOS-Chem model (v.9.1.1, <http://geos-chem.org>) is used to simulate the global
49 aerosol distributions during February and August 2008, with 4° by 5° horizontal resolution and
50 47 pressure levels from the surface to 0.01 hPa. A detailed description of the model
51 configuration and emissions is provided in the Supplementary Material. The adjoint of the
52 GEOS-Chem model [Henze *et al.*, 2007] computes $\frac{\partial M_{a,tot}}{\partial E_{i,j}}$, the sensitivity of total aerosol mass
53 concentrations, $M_{a,tot}$, to specific emissions E_i (where i is the emission type) for each grid cell
54 j . In this study, $\frac{\partial M_{a,tot}}{\partial E_{i,j}}$ is calculated monthly for $M_{a,tot}$ on the final day of the month.

55 Calculation of N_d is carried out with the Kumar *et al.* [2009] parameterization; droplets
56 are formed in ascending cloud parcels that contain soluble and insoluble particles competing for
57 water vapor. The N_d adjoint [Karydis *et al.*, 2012] provides the sensitivity of N_d with respect to
58 the parameterization input (updraft velocity, uptake coefficient and the number,
59 hygroscopicity/adsorption parameters of each aerosol lognormal mode). The two adjoint
60 frameworks elucidate the relationship of emissions to N_d by combining the local sensitivity of N_d
61 to aerosol number concentration, $\left. \frac{\partial N_d}{\partial N_a} \right|_r$ (converted into local sensitivity of N_d to aerosol mass
62 concentration, $\left. \frac{\partial N_d}{\partial M_a} \right|_r$, following the aerosol mass to number conversion as presented by Karydis
63 *et al.* [2011]), with the sensitivity of total aerosol mass concentrations to anthropogenic
64 emissions from each grid cell of the model, $\frac{\partial M_a}{\partial E_{i,j}}$, obtained from the GEOS-Chem adjoint. The
65 result is the “footprint” (or impacts expressed as the sensitivity) of anthropogenic emissions from

66 each grid cell on the droplet number over the same region, $\frac{\partial N_{d,tot}}{\partial E_{i,j}} = \left(\sum_{r=1}^R \frac{\partial N_d}{\partial M_a} \Big|_r \right) \left(\frac{\partial M_{a,tot}}{\partial E_{i,j}} \right)$,

67 where R is the total number of cells in the region. The relative importance of anthropogenic
68 emissions from each grid cell are presented as fully normalized sensitivities,

69 $R_{i,j} = \frac{E_{i,j}}{N_d} \frac{\partial N_d}{\partial E_{i,j}} = \frac{\partial \ln N_d}{\partial \ln E_{i,j}}$, where $E_{i,j}$ is the emission of type i considered for grid cell j and

70 N_d is the N_d over the region of study. $R_{tot,j}$ expresses the contribution of total anthropogenic

71 emissions from grid cell j on N_d and $R_{i,tot}$ expresses the total (worldwide) contribution of

72 anthropogenic emissions of type i (NH₃, SO₂, NO_x, or POA) on N_d . A detailed description of

73 the sensitivity calculations is provided in the Supplementary Material.

74 The sensitivity results can be used to estimate the change in future N_d and cloud albedo,

75 especially since adjoint sensitivities tend to be more robust than traditional source-receptor

76 relationships determined from perturbing emissions across broad spatial scales [Turner *et al.*,

77 [2012]. We consider projected aerosol precursor emissions following the Representative

78 Concentration Pathway 4.5 (RCP4.5; supplementary material Table S2), where total radiative

79 forcing is stabilized before 2100 by employment of a range of technologies and strategies for

80 reducing greenhouse gas emissions [Clarke *et al.*, 2007]. Using the projected emissions change

81 between 2010 and 2050, the change in N_d , ΔN_d , for each region is calculated for February and

82 August. The first order change in cloud albedo (ΔA) is calculated from ΔN_d by integrating the

83 susceptibility of A to N_d , $\frac{\partial A}{\partial N_d} = \frac{A(1-A)}{3N_d}$ [Twomey, 1991] over each region between 2010 and

84 2050 values. Cloud albedo at 2050, A_{2050} , is then given by $A_{2050} = \frac{C}{1+C}$, where

85
$$C = \left(1 + \frac{\Delta N_d}{N_{d,2010}} \right)^{1/3} \left(\frac{A_{2010}}{1 - A_{2010}} \right)$$
 and A_{2010} , $N_{d,2010}$ are the local values of albedo and N_d at 2010,

86 respectively. ΔA is then equal to $A_{2050} - A_{2010}$. Values of A_{2010} are obtained by differencing NASA
 87 CERES satellite total sky and clear sky albedos for February and August [Acker and Leptoukh,
 88 2007] as described in Moore *et al.* [2012]. These calculations give an upper limit on albedo
 89 change since liquid water feedbacks which mitigate microphysical changes are not considered
 90 [Stevens and Feingold, 2009]. Given the lack of explicit microphysics in this study, a required
 91 assumption is that the aerosol number size distribution shape remains unchanged in the future.
 92 This may induce some error in the predicted sensitivity; as differences in aerosol microphysical
 93 processes may change the relationship between emissions and CCN concentrations. Toward this,
 94 Manktelow *et al.* [2009] found that the aerosol number potential of N. American and Asian
 95 emissions was a factor of 3 to 4 times larger than that of European emissions, resulting in a 70%
 96 more efficient production of CCN.

97 **3. Results and Discussion**

98 Figure 1 presents the fully normalized sensitivity of regional N_d to anthropogenic
 99 emissions from each grid cell. Figure 2a and 2b depict the total worldwide contribution of
 100 anthropogenic emissions on N_d as well as the fractional contribution of these anthropogenic
 101 emissions from within each region for present and future (based on RCP4.5 scenario) conditions,
 102 respectively. An analysis of each region and species follows.

103 *Midwestern US:* N_d is mostly sensitive to SO_2 ($R_{\text{SO}_2, \text{tot}} = 8.6 \times 10^{-2}$) and NO_x ($R_{\text{NO}_x, \text{tot}} = 3.4 \times 10^{-2}$)
 104 during August, and to NH_3 emissions during February ($R_{\text{NH}_3, \text{tot}} = 6.3 \times 10^{-2}$). Moreover, the high
 105 impact of NH_3 emissions on N_d during February together with the low impact of NO_x emissions,
 106 confirms that NH_3 is the limiting reactant in the formation of ammonium nitrate during the

107 winter in this area. POA emissions have relatively low impact ($\sim 0.5 \times 10^{-2}$) on N_d , (Figure 2)
108 owing to the lower magnitude of the primary organic emissions, compared to inorganic
109 precursors over the midwestern US. The importance of NH_3 emissions in February and SO_2
110 emissions in August to aerosol formation over the midwestern US was also confirmed by
111 *Tsimpidi et al.* [2007]. *Henze et al.* [2009] also predicted similar trends for the sensitivity of
112 inorganic aerosols to NH_3 and SO_2 , however, the predicted effect on N_d is less pronounced as
113 only a small fraction of aerosols is activated into droplets (<5%) over the polluted continental
114 environments such as midwestern US. The lower impact of NO_x emissions on N_d during
115 February is in accordance with *Tsimpidi et al.* [2008], who attributed this behavior to the
116 increase of oxidants levels (after a NO_x emission reduction during winter) which promoted
117 secondary aerosol production and compensated for the decrease of nitrates. In August, sulfate is
118 the dominant inorganic aerosol component; therefore, aerosol concentrations are more sensitive
119 to SO_2 emission changes from electricity generating units, followed by NO_x emissions, while
120 NH_3 emissions have little impact (Figure 2). N_d is predicted to decrease by 75.62 cm^{-3} (9.87%)
121 and 26.05 cm^{-3} (3.4%) due to SO_2 and NO_x emission changes by 2050, respectively (Table S3).
122 During February, SO_2 emission decreases will increase the free NH_3 in the atmosphere, resulting
123 in an increase in ammonium nitrate concentration. Therefore, sulfate is replaced by nitrate in the
124 aerosol phase resulting in a minimal negative response of N_d to SO_2 emission changes ($R_{\text{SO}_2, \text{tot}} = -$
125 0.28×10^{-2}). The negative sensitivity arises from sufficiently low temperatures and high
126 concentrations of HNO_3 at which the addition of one mole of SO_4^{-2} to the aerosol can actually
127 reduce the total mass by displacing two moles of NO_3^- [*Moya et al.*, 2002; *West et al.*, 1999].
128 Overall, during August, N_d and A are predicted to decrease in 2050 by $\sim 100 \text{ cm}^{-3}$ (13.2% change
129 from 2010 levels) and $\sim 4 \times 10^{-3}$ (4.2%), respectively (supplementary material, Table S3). During

130 February, ΔA and ΔN_d are predicted to be very low (Table S3). In this case, $R_{SO_2,tot}$ and $R_{NO_x,tot}$
131 are very small, while $R_{NH_3,tot}$ is high (and NH_3 emissions are expected to slightly increase in the
132 future) (Figure 2a) so emissions changes have a small impact on N_d and A .

133 *Southeastern US:* N_d over the southeastern US is more sensitive to SO_2 electricity generation
134 emissions during both seasons (5.9×10^{-2}) followed by NH_3 emissions from agriculture during
135 February (5.1×10^{-2}) and NO_x emissions from electricity generating units during August (2.8×10^{-2})
136 (Figure 2). Consistent with *Capps et al.* [2012], the low amounts of nitric acid and ammonia
137 not “bound” to sulfate leads to low levels of aerosol ammonium nitrate. This means that the
138 aerosol cannot buffer SO_2 emissions shifts. N_d is predicted to decrease by 39 cm^{-3} (5.9%) and 57
139 cm^{-3} (10%) due to SO_2 emission changes by 2050 during August and February, respectively, and
140 by 16 cm^{-3} (2.5%) due to NO_x emissions changes during August (Table S3). Anthropogenic POA
141 emissions have an important impact during both February and August ($R_{NH_3,tot} = 2.2 \times 10^{-2}$ and
142 1.1×10^{-2} , respectively). Nevertheless, the predicted N_d change attributed to POA emission
143 changes by 2050 is negligible (less than 0.1%) since the latter is not expected to change
144 significantly over US (Table S2). Overall, N_d and A are predicted to decrease in 2050 by ~ 53.3
145 cm^{-3} (8.3%) and 3.5×10^{-3} (2.5%), respectively, during August and by 56.7 cm^{-3} (10%) and
146 5.1×10^{-3} (2.8%), respectively, during February (Table S3).

147 *Western US:* Anthropogenic SO_2 emissions are found to be the most important contributor to
148 cloud droplet formation over the western US during both February and August ($R_{SO_2,tot} =$
149 12.8×10^{-2} and 6×10^{-2} , respectively) (Figure 2). SO_2 emissions contribution to N_d over western
150 US is similar (during August) or even higher (during February) than the corresponding
151 contribution to N_d over midwestern and southeastern US, despite the fact that typically SO_2
152 emissions are lower in western US. This can be explained by the high sensitivity of N_d to SO_2

153 from long range transport, especially from eastern China, resulting in a significant decrease of N_d
154 in 2050 (52.6 cm^{-3} or 20.5%) due to changes on SO_2 emissions outside the western US. SO_2
155 emissions from shipping also contribute significantly to N_d . POA anthropogenic emissions also
156 significantly impact the N_d over the West, especially during February ($R_{\text{POA},\text{tot}} = 6.7 \times 10^{-2}$).
157 Similarly to southeastern US though, the predicted ΔN_d in 2050 due to POA emission changes is
158 low (Table S3). Anthropogenic NH_3 emissions during August and anthropogenic NO_x emissions
159 during February do not impact N_d over the West ($R_{\text{NO}_x,\text{tot}} = 0.5 \times 10^{-2}$ and 1.7×10^{-2} , respectively)
160 (Figure 2). Although highly concentrated centers of agricultural or urban activity emit significant
161 amounts of NO_x and NH_3 , respectively, pristine surrounding land causes dilution that limits the
162 apparent impact on aerosol formation. Overall, N_d and A are predicted to decrease in 2050 by
163 $\sim 52.6 \text{ cm}^{-3}$ (12.2%) and 2.6×10^{-3} (4%), respectively, during August and by 93.5 cm^{-3} (36.5%)
164 and 17.1×10^{-3} (12.3%), respectively, during February (Table S3). The strong effect of long-range
165 transport of pollutants is mostly evident during February where the 57% of the predicted ΔN_d in
166 2050 is attributed to changes on anthropogenic emissions from outside the western US.

167 *Central Europe:* N_d is mostly sensitive to NH_3 ($R_{\text{NH}_3,\text{tot}} = 11.7 \times 10^{-2}$) and NO_x ($R_{\text{NO}_x,\text{tot}} = 10.6 \times 10^{-2}$)
168 2) emissions during both August and February ($R_{\text{NH}_3,\text{tot}} = 5.7 \times 10^{-2}$ and $R_{\text{NO}_x,\text{tot}} = 7.1 \times 10^{-2}$). POA
169 emissions impact N_d negligibly over Central Europe during both seasons ($R_{\text{POA},\text{tot}}$ as low as
170 0.2×10^{-2}). These conclusions are consistent with *Megaritis et al.* [2012] who found that NH_3
171 emissions had the most significant impact on aerosol formation over Europe, while the impact of
172 POA emissions was the smallest. Ammonium nitrate is a major aerosol component over Europe;
173 therefore, emissions of its gas phase precursors (NH_3 and NO_x) exhibit the strongest impact on
174 the inorganic aerosol population and N_d . However, according to RCP4.5 scenario, emissions of
175 NH_3 over central Europe are not expected to change significantly by 2050 (0.06% increase). On

176 the other hand, changes on NO_x emissions result to significant changes on ΔN_d (315 cm^{-3} or 50%
177 and 74 cm^{-3} or 10% during August and February, respectively). SO_2 also contributes with a
178 similar magnitude to N_d over central Europe during August ($R_{\text{SO}_2, \text{tot}} = 6.7 \times 10^{-2}$). N_d is predicted
179 to decrease by 230 cm^{-3} (37%) due to SO_2 emission changes by 2050 and is strongly influenced
180 by long-range transport of SO_2 from Midwestern and northeastern US. The predicted ΔN_d due to
181 changes of anthropogenic SO_2 emissions from outside the central Europe during August is 230
182 cm^{-3} (23%). During February, when sulfate levels are lower, N_d is less sensitive to SO_2 emissions
183 ($R_{\text{SO}_2, \text{tot}} = 0.8 \times 10^{-2}$), likely due to the limited H_2O_2 availability over Europe during this season
184 [Megaritis et al., 2012]. Overall, ΔN_d decrease by 510 cm^{-3} (82%) and ΔA by 31.86×10^{-3} (42%,)
185 during August. The large $R_{\text{SO}_2, \text{tot}}$ and $R_{\text{NO}_x, \text{tot}}$ predicted over Europe during this season (Figure 2),
186 together with the large future reductions of these emissions (e.g., SO_2 ; Figure 1) results in the
187 strong response of N_d and A to future emission changes, which is also influenced by the long-
188 range impact of SO_2 emission reductions from the US (Figure 1). During February, ΔN_d and ΔA
189 are predicted to be 77 cm^{-3} (10.7%) and 4.2×10^{-3} (3.2%), respectively, and are mostly attributed
190 to NO_x emissions (Table S3).

191

192 4. Conclusions

193 In this work, the adjoints of the GEOS-Chem CTM and Kumar et. al [2009] droplet
194 parameterization are used to determine the source region and relative impact of aerosol precursor
195 emissions on N_d and albedo. Assessments are carried out for specific regions in February and
196 August of 2008 and 2050 (using the RCP4.5 scenario). Among all species, the influence of
197 sulfate emissions in August is most similar across regions, comprising at least 50% (20%) of the
198 total influence in the US regions (Central Europe). For each region, the percent contribution

199 from sulfate emissions within the region is the lowest of all the species tracked, which implicates
200 long-range transport of significant sulfate burdens. Central Europe N_d in August is far more
201 influenced by NH_3 and NO_x emissions than any region over the US due to the large contribution
202 of ammonium nitrate aerosol to the regional aerosol. Nevertheless, for all four regions, 65-85%
203 of the influence from NH_3 and NO_x emissions came from within the region in August. In
204 February, the influence of NO_x emissions on Central Europe N_d remains much higher than on the
205 N_d over US regions. However, the impact of NH_3 emissions on N_d , during February, is similar for
206 Central Europe, midwestern, and southeastern US. Additionally, the maximum percent
207 contribution of NO_x emissions within the region (nearly 100%) is attained in the western and
208 southeastern US in February. The strongest internal influence is from POA in both August and
209 February, owing to the shorter lifetime of the species. With the exception of the western US, the
210 relative influence of POA on N_d is minor. The influence of different emissions mixes on N_d is
211 most apparent from the distinction between the US and European response to NO_x and NH_3
212 emissions. Based on the RCP4.5 scenario and the 2008 adjoint, A in Central Europe during
213 August and in western US during February is most strongly affected by future emission changes,
214 owing to the effects of long-range transport of anthropogenic emissions (mainly SO_2) and NO_x
215 reductions over Central Europe and the western US. The significant decrease of N_d over these
216 regions results in higher anthropogenic emissions contributions to N_d in 2050 as the maximum
217 supersaturation and droplet activation fractions are increased [Karydis et al., 2012]. In other
218 regions and seasons, the anthropogenic emissions contributions to N_d are lower in 2050, mainly
219 due to the significant decrease of SO_2 and NO_x emissions over the US and Europe (figure 2b).
220 The above results reveal N_d sensitivities that would not have been predicted from earlier forward
221 sensitivity analyses, i.e. the importance of SO_2 long-range transport on N_d over western US or the

222 increasing contribution of SO₂ to N_d over central Europe in the future, and can be used from
223 policy makers to effectively design future emission control strategies.

224 Understanding relative impacts in coupled complex models is a challenge. Using the
225 coupled adjoint sensitivity modeling framework developed here has allowed us to quantify the
226 sensitivity of cloud droplet formation and cloud albedo to different aerosol precursor emissions
227 and to unravel the importance of sectoral, spatial, and seasonal emissions.

228 **Acknowledgments**

229 We acknowledge support from NASA-ACMAP and NOAA. SLC gratefully acknowledges the
230 NSF GRF and NASA ESSF. DKH Acknowledges support from a NASA New Investigator
231 grant. We also thank two anonymous reviewers for comments that improved the manuscript.

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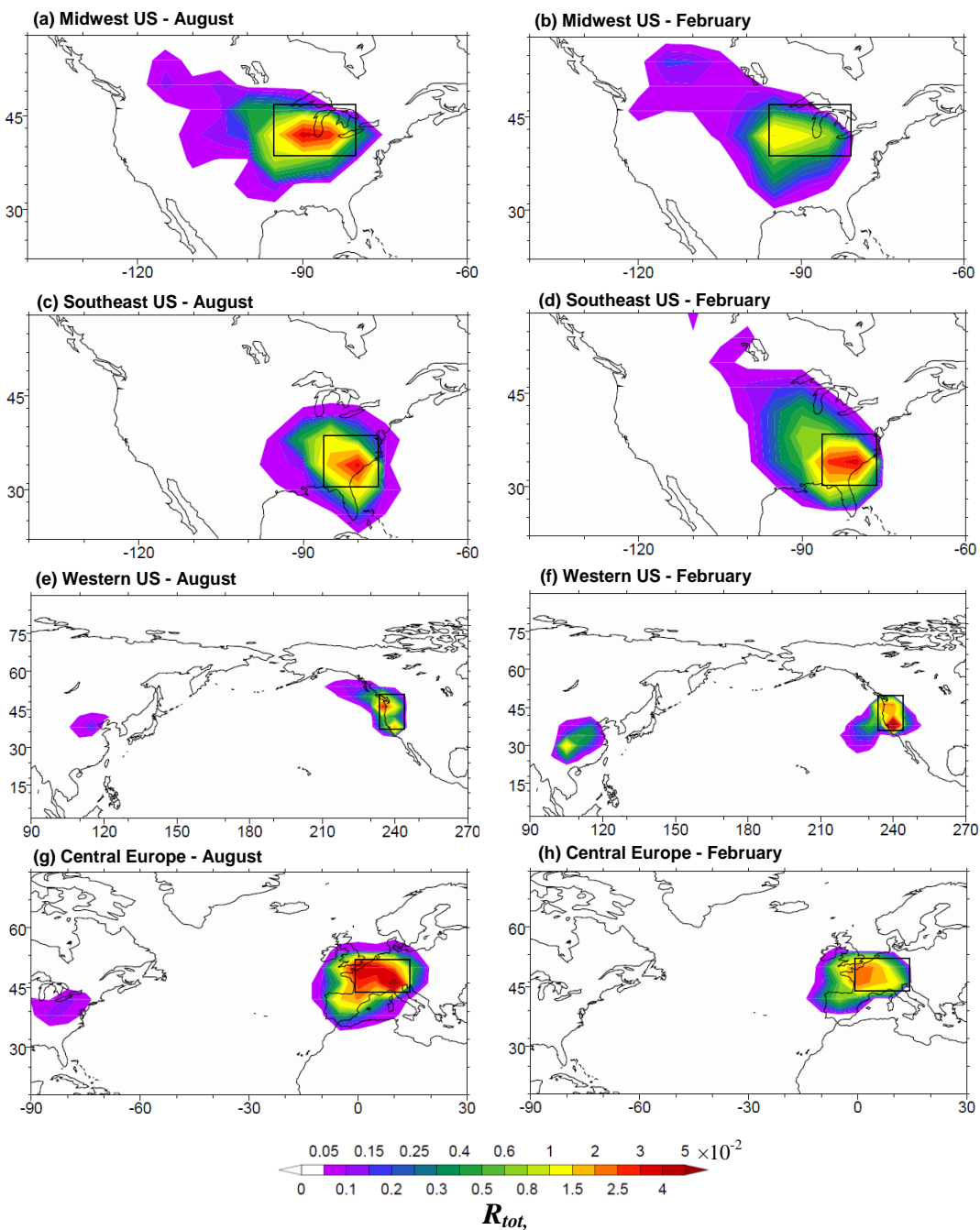
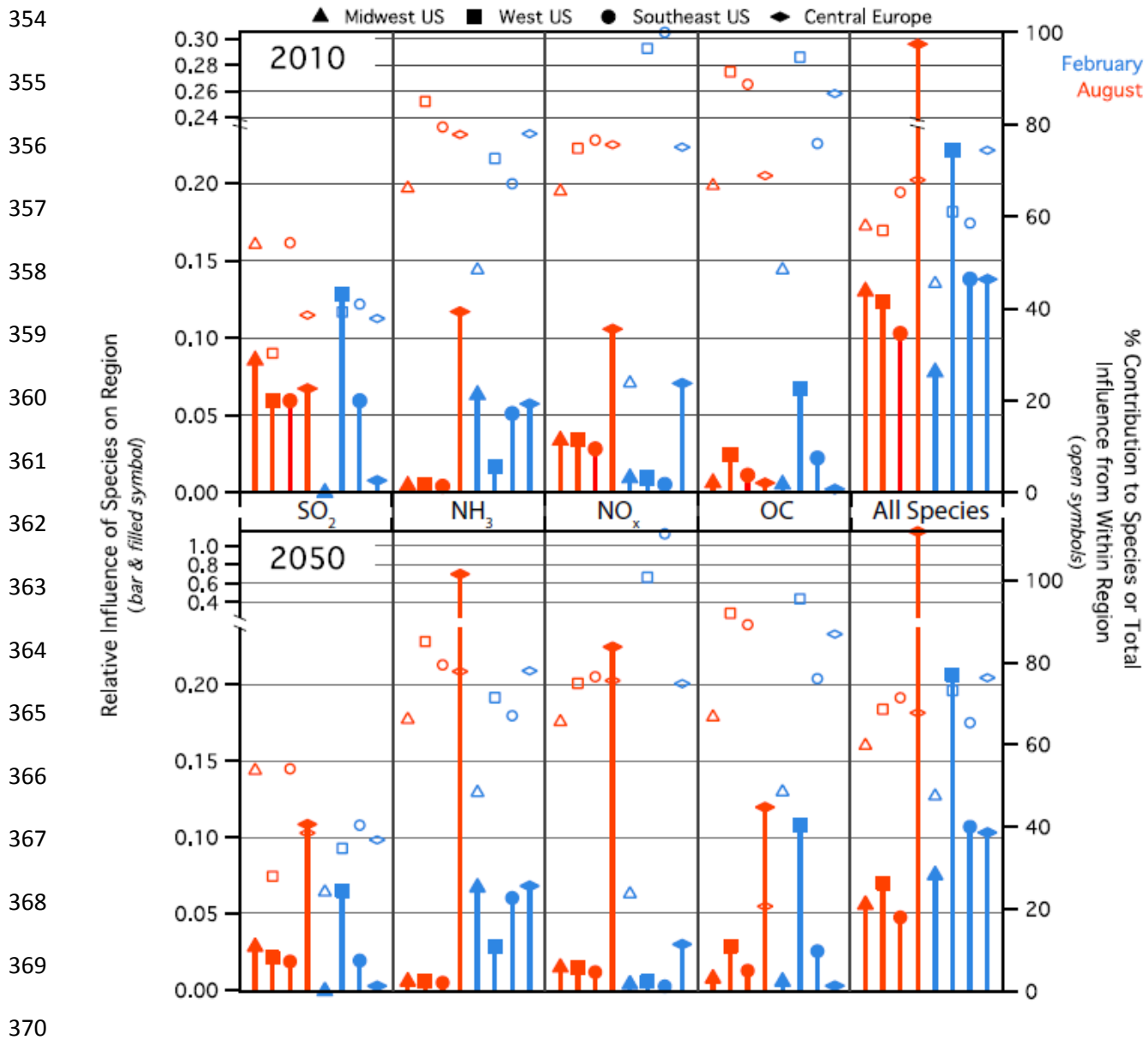


Figure 1: Contribution of total anthropogenic emissions to cloud droplet number concentration over Midwest US (a, b), Southeast US (c, d), Western US (e, f), and Central Europe (g, h) during August (left column) and February (right column) of 2008. The regions of study are denoted by the squares.



371 **Figure 2:** Total contribution of anthropogenic SO₂, NO_x, NH₃, and OC emissions to N_d over
 372 Midwest US, Southeast US, Western US, and Central Europe during August (orange) and
 373 February (blue) of 2010 (top panel) and 2050 (bottom panel). The fractional contribution of
 374 anthropogenic emissions from within each region on N_d is also shown (open symbols).

Supplementary Material

1. Model framework description

The GEOS-Chem model is driven using assimilated meteorological data from the Goddard Earth Observing System (GEOS-5) of the NASA Global Modeling and Assimilation Office (GMAO) [Fountoukis and Nenes, 2007] and includes gas-phase chemistry coupled with heterogeneous reactions, inorganic aerosol thermodynamics, and oxidative aging of carbonaceous aerosols [Bauer et al., 2004]. NH₃ emissions are from the Global Emission Inventory Activity (GEIA) inventory [Bouwman et al., 1997]. Biogenic VOCs follow the Model of Emissions of Gases and Aerosols from Nature (MEGAN) inventory [Guenther et al., 2006]. NO_x and SO_x emissions are obtained from the Emissions Database for Global Atmospheric Research (EDGAR) inventory [Olivier and Berdowski, 2001]. These global inventories are overwritten with regional inventories for the US (NEI99), Europe (EMEP), southeast Asia [Streets et al., 2003], and Mexico (BRAVO) [Kuhns et al., 2005]. Biomass burning emissions are from van der Werf et al. [2006].

Aerosol mass concentrations from GEOS-Chem follow a prescribed number size distribution (Table S1) according to Karydis et al. [2012] and distributed in three externally mixed modes: *i*) sulfate, ammonium, nitrate, organics, and black carbon from anthropogenic and biomass burning sources, *ii*) sulfate, nitrate, and sea salt from marine processes, and, *iii*) mineral dust. Sensitivity to secondary organic aerosol (SOA) is not considered, as GEOS-Chem does not produce adjoint sensitivities of SOA mass to volatile organic carbon emissions. Following

Karydis et al. [2011], the number concentration of each aerosol mode is given by $N_a = \frac{m_{tot}}{V \rho_{tot}}$

where m_{tot} is the total mass concentration of the aerosol mode, V is the volume of the dry

aerosol, and ρ_{tot} is the density of the aerosol given by $\rho_{tot} = \frac{m_{tot}}{\sum_{y=1}^Y \frac{m_y}{\rho_y}}$. m_y is the mass

concentration of each component within the aerosol mode (e.g. sulfate, nitrate, ammonium, OC, BC), ρ_y is its density and Y is the number of aerosol components. The hygroscopicity of each aerosol mode is calculated based on the aerosol composition, provided by GEOS-Chem, and is given by $\kappa = \frac{VH_{f,tot}\rho_{tot}V_{f,s}M_w}{M_s\rho_w}$, where $V_{f,s}$ is the soluble volume fraction, M_w and M_s is the

5 molecular weight of water and soluble fraction respectively, ρ_w is the density of water, and,

$VH_{f,tot}$ is the total Van't Hoff factor of soluble fraction given by $VH_{f,tot} = \frac{\sum_{y=1}^{YS} \frac{m_y VH_{f,y}}{M_y}}{\sum_{y=1}^{YS} \frac{m_y}{M_y}}$, where

YS is the number of aerosol soluble components, $VH_{f,y}$ is the Van't Hoff factor of each soluble aerosol component and M_y is the molecular weight of each soluble aerosol component. Other important parameters for N_d calculations are presented by *Karydis et al.* [2012].

10

2. Computing sensitivities of N_d to anthropogenic aerosol emissions and ΔN_d due to future anthropogenic emissions changes

Figure 1 of the main text presents the fully normalized sensitivity (unitless) of regional N_d to anthropogenic emissions from each grid cell:

$$15 \quad R_{i,j} = \sum_{r=1}^R \left(\left(\frac{N_{a,r}}{N_{d,r}} \frac{\partial N_{d,r}}{\partial N_{a,r}} \right) \sum_{y=1}^Y \left(\frac{M_{a,r,y}}{N_{a,r}} \frac{\partial N_{a,r}}{\partial M_{a,r,y}} \right) \right) \left(\frac{E_{i,j}}{\sum_{r,y=[1,1]}^{[R,Y]} M_{a,r,y}} \frac{\partial \sum_{r,y=[1,1]}^{[R,Y]} M_{a,r,y}}{\partial E_{i,j}} \right) = \frac{E_{i,j}}{\sum_{r=1}^R N_{d,r}} \frac{\partial \sum_{r=1}^R N_{d,r}}{\partial E_{i,j}} = \frac{\partial \ln N_d}{\partial \ln E_{i,j}}$$

where $E_{i,j}$ is the emission of type i considered for grid cell j , $N_{d,r}$ and $N_{a,r}$ are the are the aerosol number and droplet number concentrations for regional grid cell r , respectively, $M_{a,r,y}$ is the aerosol mass concentration of species y for regional grid cell r , and N_d is the total droplet number concentration over the region of study. The first term,

$\sum_{r=1}^R \left(\left(\frac{N_{a,r}}{N_{d,r}} \frac{\partial N_{d,r}}{\partial N_{a,r}} \right) \sum_{y=1}^Y \left(\frac{M_{a,r,y}}{N_{a,r}} \frac{\partial N_{a,r}}{\partial M_{a,r,y}} \right) \right)$, is the fully normalized adjoint sensitivity of total droplet

number to aerosol mass concentration, over the region of study, from the droplet activation

parameterization; the second term, $\left(\frac{E_{i,j}}{\sum_{r,y=[1,1]}^{[R,Y]} M_{a,r,y}} \frac{\partial \sum_{r,y=[1,1]}^{[R,Y]} M_{a,r,y}}{\partial E_{i,j}} \right)$, is the fully normalized adjoint

sensitivity of aerosol mass concentration, over the region of study, to anthropogenic emissions of

5 type i considered for grid cell j , calculated by GEOS-Chem adjoint. The above sensitivity

calculations are made only for the aerosol mode containing sulfate, ammonium, nitrate, organics,

and black carbon, since currently GEOS-Chem does not produce adjoint sensitivities to sea salt

and dust emissions.

Figure 2a of the main text depicts the total worldwide contribution of anthropogenic emissions to

10 present N_d (unitless): $R_{i,tot} = \sum_{j=1,n} \frac{\partial \ln N_d}{\partial \ln E_{i,j}}$, where i is the emission type and n is the total number

of grid cells.

Figure 2b of the main text depicts the total worldwide contribution of anthropogenic emissions to

future N_d (unitless): $R_{i,tot} = \sum_{j=1,n} \left(\frac{\partial \ln N_d}{\partial \ln E_{i,j}} \frac{(1 + \Delta E_{i,j})}{(1 + \Delta N_d)} \right)$, where $\Delta E_{i,j}$ and ΔN_d are the fractional

change of emissions $\left(\frac{E_{i,j,2050} - E_{i,j,2010}}{E_{i,j,2010}} \right)$ and droplet number $\left(\frac{N_{d,2050} - N_{d,2010}}{N_{d,2010}} \right)$ from Table S2

15 and Table S3, respectively.

Table S3 presents the change in N_d (cm^{-3}) due to future emissions changes:

$$\left(\frac{\partial N_d}{\partial M_a}\right)\left(E_{i,j}\frac{\partial M_a}{\partial E_{i,j}}\right)\Delta E_{i,j} = \frac{\partial N_d}{\partial E_{i,j}}(E_{i,j,2050} - E_{i,j,2010}).$$

The first term, $\left(\frac{\partial N_d}{\partial M_a}\right)$, is the un-normalized

adjoint sensitivity of droplet number to aerosol mass concentration, over the region of study,

from the droplet activation parameterization, while the second term, $\left(E_{i,j}\frac{\partial M_a}{\partial E_{i,j}}\right)$, is the semi-

5 normalized adjoint sensitivity of aerosol mass concentration, over the region of study, to anthropogenic emissions of type i considered for grid cell j , calculated by GEOS-Chem adjoint.

The third term, $\Delta E_{i,j}$, is the fractional change of emissions from Table S2.

3. R_{ij} and emissions contributions to N_d for each region

10 *Midwestern US*: During August, emissions from within the Midwest contribute ~60% of the N_d , with values of $R_{tot,j}$ up to 3.4×10^{-2} over Illinois (Figure 1a of main text). Emissions from South and Southwest Canada notably contribute to N_d (up to 0.15×10^{-2}). During February, the predicted impact of anthropogenic emissions on N_d over the Midwest US covers a larger region, with the impact of emissions from Canada being more important.

15 *Southeastern US*: Emissions from within the region have the largest impact (60-65%) on N_d in the region ($R_{tot,j}$ up to 3.4×10^{-2}) during both February and August (Figure 1c,1d, and 2 of main text). Emissions from the Midwest also impact regional N_d during both seasons ($R_{tot,j}$ up to 1×10^{-2}). This impact is mostly due to SO_2 emissions during summer and NH_3 emissions during winter. N_d is affected by emissions (mainly NH_3) from south-central Canada during February ($R_{\text{NH}_3,tot}$ up
20 to 0.1×10^{-2}).

Western US: N_d is strongly affected by regional emissions (over California during February, $R_{tot,j} = 6.5 \times 10^{-2}$, and over Washington state during August, $R_{tot,j} = 2.9 \times 10^{-2}$; Figure 1e, 1f of main text). Long-range transport of anthropogenic emissions from Asia [e.g., Jaffe *et al.*, 2003; Wolff, 1984] also impacts significantly cloud droplet formation over the western US, especially during
5 February ($R_{tot,j}$ up to 1.5×10^{-2} for Eastern China) and accounts approximately for one third of the total sensitivity of emission impacts on N_d over the western US.

Central Europe: During August, anthropogenic emissions from northern Italy (Po Valley), have the largest impact on N_d over Central Europe ($R_{tot,j}$ up to 5.3×10^{-2} , Figure 1g of main text). Anthropogenic emissions from the northeastern and midwestern US also contribute about 4% to
10 the N_d over Europe ($R_{tot,j}$ up to 0.2×10^{-2}). Due to the prevalent winds during February, N_d is more sensitive to anthropogenic emissions over France with $R_{tot,j}$ up to 2.4×10^{-2} (Figure 1h of main text).

Table S1. Size distribution parameters applied to aerosol populations from GEOS-Chem model

Aerosol type	Aerosol components	Density (g cm⁻³)	Median diameter (μm)	Geometric standard deviation	Number fraction	Reference
Anthropogenic, Biomass Burning	Sulfate	1.77	0.1	1.9	1	Chuang et al. [1997]; Radke et al. (1988)
	Nitrate	1.725				
	Ammonium	1.75				
	Organic Carbon	1.2				
	Black Carbon	1.5				
Marine	Sulfate	1.77	0.018	1.4	0.81	Lance et al. [2004]
	Nitrate	1.725	0.075	1.6	0.18	
	Sea Salt	2.2	0.62	2.7	0.01	
Mineral Dust	Mineral Dust	2.6	0.16	2.1	0.93	D’Almeida et al. [1987]
			1.4	1.9	0.07	
			10	1.6	3×10 ⁻⁶	

Table S2: Fractional emission change (2010-2050) for SO₂, NH₃, NO_x, and primary OC based

5 on RCP4.5 scenario. Negative values correspond to a decrease from 2010 to 2050.

Region	SO₂	NH₃	NO_x	Primary OC
Western Europe, Northern America, Australia, Japan	-0.72	0.06	-0.62	0.02
Eastern Europe, Russia	-0.69	0.03	-0.50	-0.10
Asia	-0.64	0.12	-0.24	-0.52
Middle East, Africa	-0.05	0.20	0.04	0.00
Latin and Central America	-0.49	0.06	-0.23	-0.42

Table S3: Change (2010-2050) in droplet number (cm^{-3}) and shortwave cloud albedo from emissions changes based on RCP4.5 scenario. Negative values correspond to a decrease from 2010 to 2050. Numbers in parentheses represent the percentile change (%) of droplet number and cloud albedo.

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Region	ΔN_d from SO_2	ΔN_d from NH_3	ΔN_d from NO_x	ΔN_d from primary OC	Total ΔN_d	ΔA from Total ΔN_d ($\times 10^{-3}$)
Midwestern US	-75.62	0.34	-26.05	0.18	-101.15	4.08
August	(-9.87)	(0.04)	(-3.4)	(0.03)	(-13.2)	(4.2)
Midwestern US	2.47	4.64	-7.13	0.15	0.12	-0.001
February	(0.42)	(0.77)	(-1.18)	(0.02)	(0.02)	(-0.005)
Southeastern US	-39.02	0.22	-15.78	0.24	-53.34	3.47
August	(-5.92)	(0.04)	(-2.45)	(0.08)	(-8.3)	(2.5)
Southeastern US	-57.24	4.22	-4.37	0.67	-56.71	5.12
February	(-9.99)	(0.74)	(-0.77)	(0.12)	(-9.9)	(2.8)
Western US	-35.03	0.24	-18.17	0.36	-52.59	2.58
August	(-8.12)	(0.06)	(-4.22)	(0.09)	(-12.2)	(4.0)
Western US	-89.64	1.28	-6.25	1.08	-93.53	17.13
February	(-34.99)	(0.5)	(-2.44)	(0.43)	(-36.5)	(12.3)
Central Europe	-229.29	33.55	-314.99	0.70	-510.03	31.86
August	(-37)	(5.42)	(50.83)	(0.11)	(-82.3)	(41.9)
Central Europe	-9.16	5.75	-73.54	0.09	-76.86	4.21
February	(-1.28)	(0.8)	(-10.24)	(0.01)	(-10.7)	(3.2)

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