A parameterization of sub-grid particle formation in sulphur-rich plumes for global- and regional-scale models.

R. G. Stevens[1], J. R. Pierce[1,2]

[1]Dalhousie University, Halifax, NS, Canada

[2]University of Colorado, Fort Collins, CO, USA

DALHOUSIE UNIVERSITY *Inspiring Minds*

Acknowledgements: We thank Marat Khairoutdinov for access to the System for Atmospheric Modelling. We also thank the Electric Power Research Institute and Natural Sciences and Engineering Research Council of Canada Postgraduate Scholarship D for funding this work.

Abstract

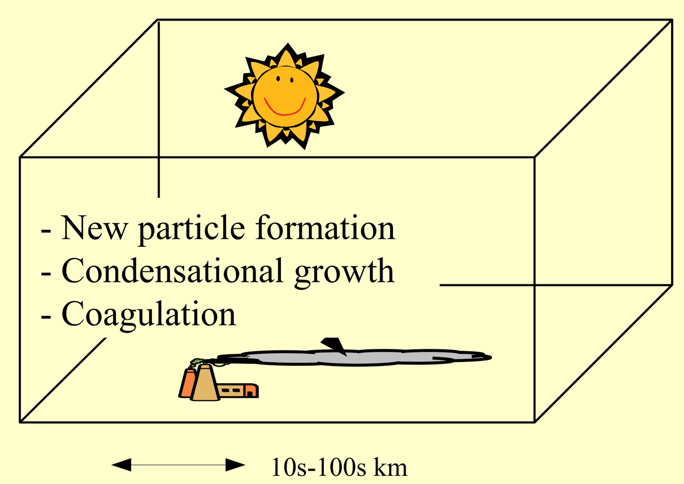
New-particle formation in the plumes of coal-fired power plants and other anthropogenic sulphur sources may be an important source of particles in the atmosphere. It remains unclear, however, how best to reproduce this formation in global and regional aerosol models with grid-box lengths that are 10s of kilometres and larger. The predictive power of these models is thus limited by the resultant uncertainties in aerosol size distributions. We focus on sub-grid sulphate aerosol processes within coal-fired power plant plumes: the sub-grid oxidation of SO_2 with condensation of H_2SO_4 onto newly-formed and preexisting particles.

Based on the results of the System for Atmospheric Modelling (SAM), a Large-Eddy Simulation/Cloud-Resolving Model (LES/CRM) with online TwO Moment Aerosol Sectional (TOMAS) microphysics, we have developed a computationally efficient, but physically based, parameterization that predicts the characteristics of aerosol formed within coal-fired power plant plumes based on parameters commonly available in global and regional-scale models. Given large-scale mean meteorological parameters, emissions from the power plant, mean background condensation sink, mean background SO_2 and NO_x concentrations, and the desired distance from the source, the parameterization will predict the fraction of the emitted SO_2 that is oxidized to H_2SO_4, the fraction of that H_2SO_4 that forms new particles instead of condensing onto preexisting particles, the mean mass per particle of the newly-formed particles, and the number of newly-formed particles per kilogram SO_2 emitted.

The parameterization we describe here should allow for more accurate predictions of aerosol size distributions and a greater confidence in the effects of aerosols in climate and health studies.

1. The "primary sulfate" conundrum

Global and regional aerosol microphysics models cannot explicitly resolve power-plant plumes near their source. However, the chemistry and aerosol microphysics in these plumes may have a large global effect.



- Global models assume distribution of sub-grid or "primary" sulfate from power plants
- 4 major models use 4 different assumptions
- Greatly affect cloud condensation nuclei

Figure 1 (above)

Power-plant plume within a global or regional model grid box. Aerosol processes may be very different in the plume from the grid-box average.

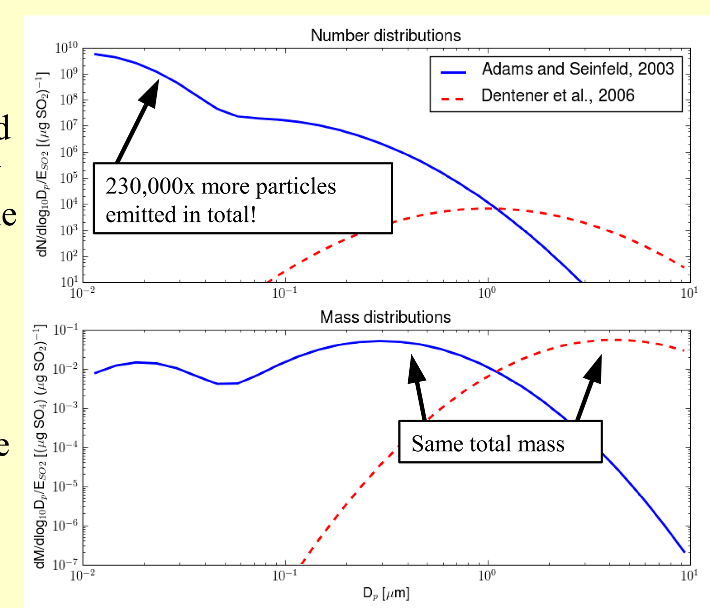


Figure 2 (right)

Two estimates of sub-grid sulfate number and mass size distributions normalized by SO_2 emissions.

2. How does this affect global aerosol predictions?

In order to test how the sub-grid sulfate assumptions may affect global aerosol predictions, we ran two simulation in the GISS-II' GCM with the online aerosol microphysics module TOMAS (Adams and Seinfeld, 2003). All else is held fixed between the two simulations.

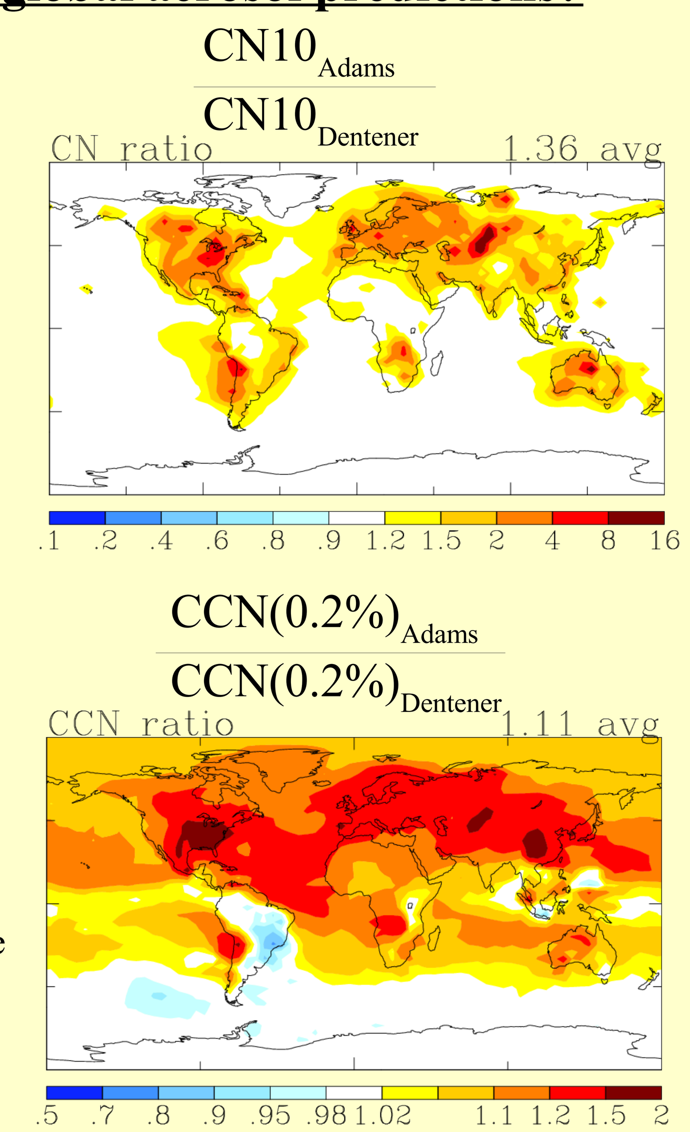


Figure 3 (right)

CN10 and CCN(0.2%) ratios for the two "primary sulfate" simulations. Please note that the colorbar is different for the two plots. There is a significant difference in both CN and CCN globally when changing this single source.

3. Can sub-grid sulfate be improved?

All of the current assumptions about "primary sulfate" in global models are constant for all power-plants under all conditions; however, aerosol microphysical processes depend greatly on source and the existing chemical and physical state of the atmosphere. We have developed a computationally efficient sub-grid sulfate parameterization that predicts the following outputs based on the following inputs:

Inputs:

- SO_2emis and NO_xemis, emissions of SO_2 and NO_x
- $bgSO_2$ and $bgNO_x$, background SO_2 and NO_x concentrations
- CS, background aerosol condensation sink
- Meteorology:
  - blh, boundary layer height
  - v_g, wind speed
  - $dswrf$, sunlight

Outputs:

- f_{ox}, the fraction of SO_2 that oxidizes to form H_2SO_4
- f_{new}, the fraction of H_2SO_4 formed that condenses onto new particles
- N_{new}, the number of newly-formed particles per kg SO_2 emitted
- M_m, the mean mass per particle of newly-formed particles

To do this, we have integrated TOMAS aerosol microphysics into SAM, a LES/CRM model:

TwO-Moment Aerosol Sectional microphysics algorithm (Adams and Seinfeld, 2002)

- Multiple chemical components
- 15 size bins (number and mass)
- Diameters: 3 nm – 10 μm

System for Atmospheric Modelling: (Khairoutdinov and Randall, 2003)

- Large-eddy simulation/cloud resolving model
- Fluid-dynamics model spanning 10s or 100s of km
- Grid box size 10s of meters to several km
- Able to resolve many cloud types and emissions plumes

4. Comparison to observations: the 2006 TEXAQS field campaign

We have evaluated the SAM-TOMAS model against airborne data obtained in the plume of the Parish power generation facility during the 2006 Texas Air Quality Survey (TEXAQS) field campaign. The model provides reasonable predictions of new particle formation and growth within the plume. We also evaluated the model against observations obtained downwind of the Conesville power generation facility during the 2004 International Consortium for Atmospheric Research on Transport and Transformation (ICARTT) campaign (not shown), and found similar results (Stevens et al., 2012).

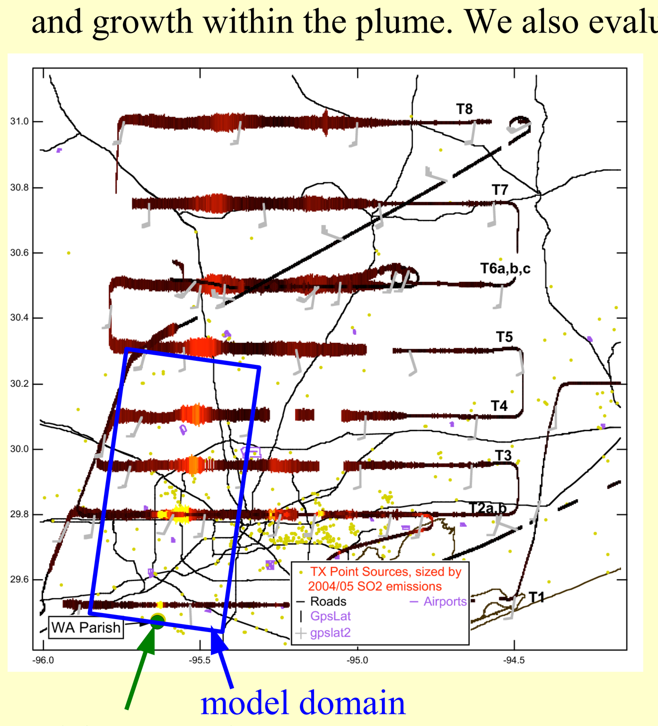


Figure 4 (upper left)
Measured SO_2 concentrations along 2006 TEXAQS flight path

Figure 5 (lower left)
(a) Trace gases and (b) particle number vs. distance downwind from the Parish power-plant, averaged over plume. Dots are aircraft observations; lines are model results. Shaded area and error bars indicate one standard deviation in concentration across plume width.

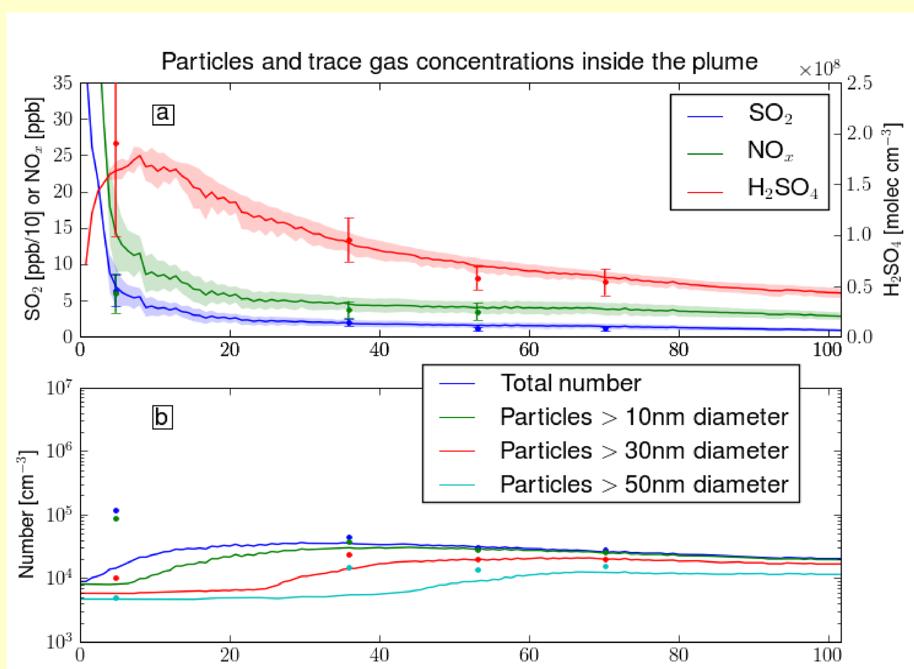
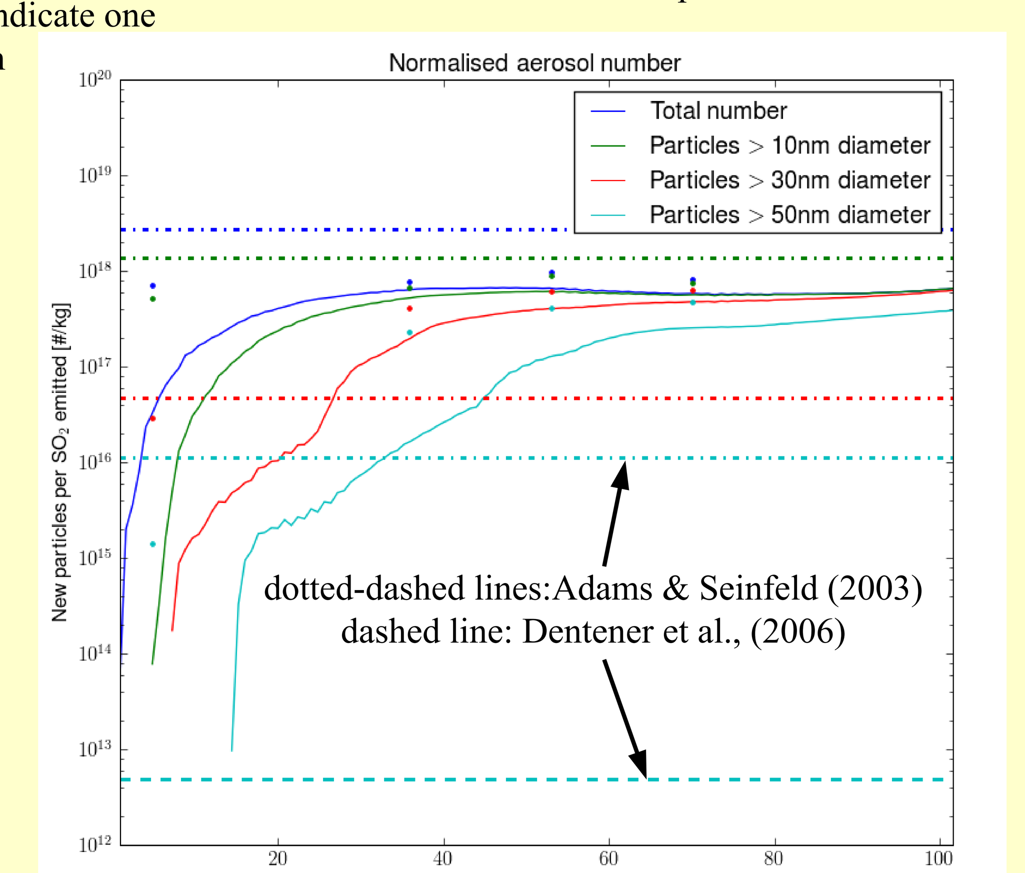


Figure 6 (lower right)
Additional predicted particles per kg SO_2 versus distance from the Parish power plant, summed over the plume. Dots indicate aircraft observations, solid lines indicate model results. Essentially all particles in the Dentener et al. (2006) distribution are larger than 50 nm, so all dashed lines overlap one another.



5. Current parameterization results

We have performed more than 4500 simulations using the SAM-TOMAS model using meteorology, background trace gases and background aerosol concentrations from the GEOS-Chem chemical transport model to create a set of training data. We then used this training data to fit free parameters in a series of empirical equations.

Calculate effective NO_x and SO_2 concentrations within the plume

$$SO_{2,eff}=bgSO_2+1.71\times10^4\frac{SO_2emis}{v_g^{1.22}blh^{0.19}\left(\frac{d}{vg}\right)^{0.77}}\qquad NO_{x,eff}=bgNO_x+9.60\times10^4\frac{NO_xemis}{v_g^{1.23}blh^{0.20}\left(\frac{d}{v_g}\right)^{0.79}}$$

Predict effective OH concentration in plume based on $NO_{x,eff}$ and DSWRF, according to parametrization detailed in Stevens et al. (2012)

Predict the fraction of SO_2 that oxidizes to form H_2SO_4 (f_{ox}) by:

$$f_{ox}=1-\exp\left(-1.65\times10^{-10}OH_{eff}^{0.79}\left(\frac{d}{v_g}\right)^{0.77}\right)$$

$$\frac{SO_{2eff}^{1.92}dswrf^{3.28}}{NO_{x\,eff}^{1.24}CS^{3.48}}>2.99\times10^{14}$$

(96% accuracy)

No: insignificant new-particle formation

- M_m undefined
- $N_{new}=0.$
- $f_{new}=0.$

Yes: new particle formation

mass per particle

$$M_m=1.48\times10^{-27}\frac{f_{ox}^{1.52}SO_{2,eff}^{1.09}}{CS^{-0.62}}\left(\frac{d}{v_g}\right)^{0.97}+4.07\times10^{-23}kg$$

new particles per kg SO_2

$$N_{new}=3.49\times10^{23}f_{ox}^{2.67}SO_{2eff}^{0.27}\exp\left(-4.82\,CS^{0.17}\left(\frac{d}{v_g}\right)^{0.17}\right)$$

fraction of sulphate in new particles

$$f_{new}=\frac{M_mN_{new}}{f_{ox}}\frac{M_{SO_2}}{M_{SO_4}}$$

Figure 7: Parametrized values vs SAM-TOMAS model results for a) f_{ox}, b) M_m, c) N_{new}, and d) f_{new}. Solid lines indicate 1:1 ratios, yellow dashed lines indicate 10:1 ratios, and green dashed lines indicate 2:1 ratios. The horizontal red and blue dashed lines indicate the values from Dentener et al. (2006) and Adams and Seinfeld (2003), respectively.

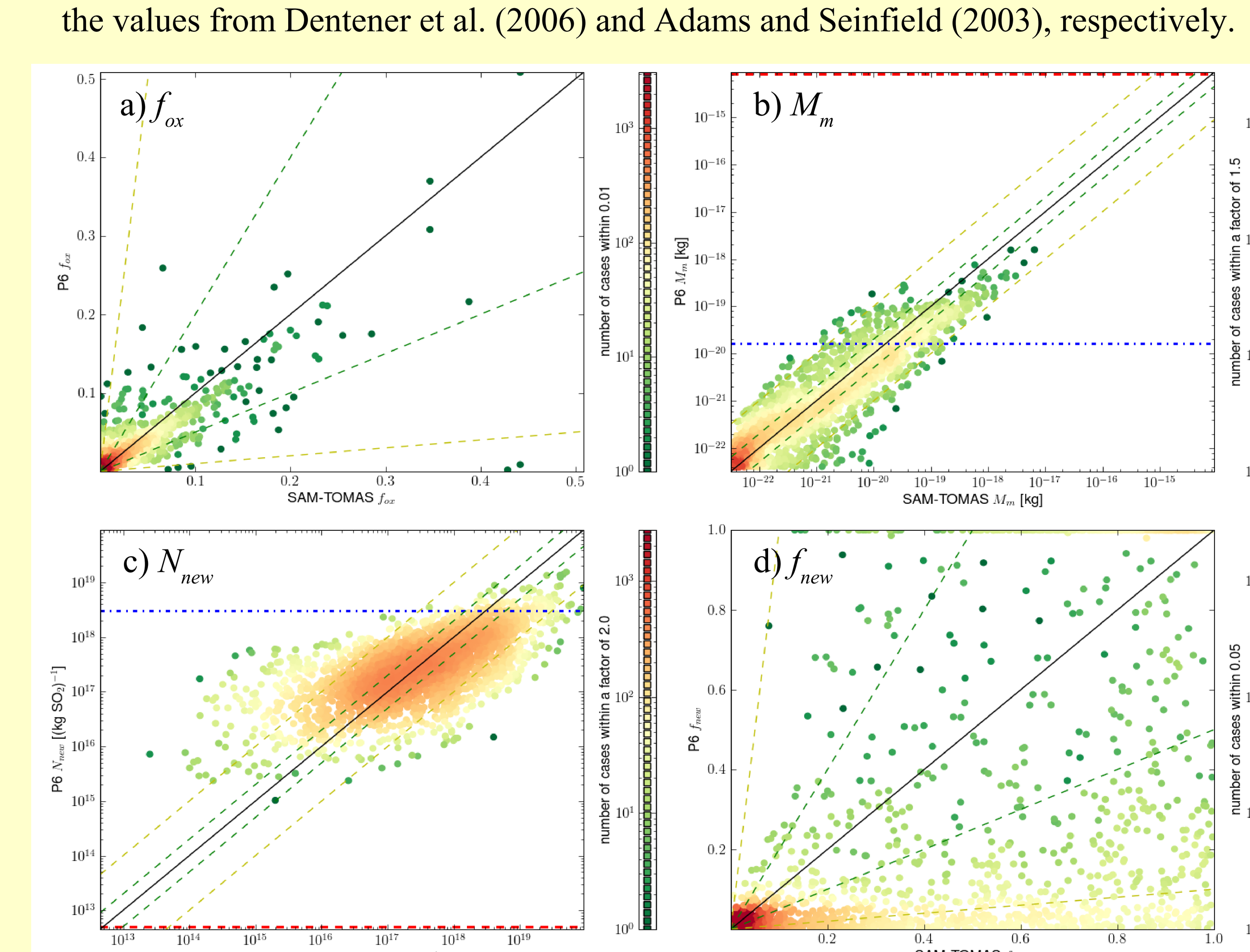


Table 1. Values of correlation coefficients and fractions of data within factors of 2 and 10.

	f_{ox}	M_m	N_{new}	f_{new}
correlation coefficient	0.83	0.86	0.62	0.67
within a factor of 2	50 %	60 %	34%	29%
within a factor of 10	77 %	97%	84%	67%

6. Summary

- We have evaluated the SAM-TOMAS model against observations, and particle size distributions and number concentrations are reproduced well for distances relevant to global and regional-scale models.
- Background aerosol, meteorology, and emissions have large effects on aerosol formation and growth – no single assumed aerosol size distribution will be appropriate in all cases.
- We have created a parametrization that will capture these effects and provide more realistic aerosol size and number information for global and regional-scale models.
- We can currently predict whether or not significant new-particle formation will occur with 96% accuracy.
- For those cases where we correctly predict new-particle formation, we generally predict the mean mass per particle and the number of new particles within a factor of 10.

7. References

1. Adams, P. J., and J. H. Seinfeld, *Geophys. Res. Lett.*, 10.1029/2002GL016303, 2003.
2. Dentener, F., et al., *Atmos. Chem. Phys.*, 6, 4321-4344, 2006.
3. Adams, P. J., and J. H. Seinfeld, *J. Geophys. Res.*, 10.1029/2001JD001010, 2002.
4. Khairoutdinov, M. F., and D.A. Randall, *J. Atmos. Sci.*, 60, 607-625, 2003.
5. Stevens, R. et al., *Atmos. Chem. Phys.*, 12, 189-206, 2012.