FINAL REPORT

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2. Title of Research Grant: Cloud/Aerosol Parameterizations: Application and Improvement of General Circulation Models

3. **Scientific Goal(s) of Research Grant**: **Abstract**

One of the biggest uncertainties associated with climate models and climate forcing is the treatment of aerosols and their effects on clouds. The effect of aerosols on clouds can be divided into two components: The first indirect effect is the forcing associated with increases in droplet concentrations; the second indirect effect is the forcing associated with changes in liquid water path, cloud morphology, and cloud lifetime. Both are highly uncertain. This project applied a cloud-resolving model to understand the response of clouds under a variety of conditions to changes in aerosols. These responses are categorized according to the large-scale meteorological conditions that lead to the response. Meteorological conditions were sampled from various fields, which, together with a global aerosol model determination of the change in aerosols from present day to pre-industrial conditions, was used to determine a first order estimate of the response of global cloud fields to changes in aerosols. The response of the clouds in the NCAR CAM3 GCM coupled to our global aerosol model were tested by examining whether the response is similar to that of the cloud resolving model and methods for improving the representation of clouds and cloud/aerosol interactions were examined.

4. Progress and accomplishments during the period of the grant:

(1) Published two manuscripts analyzing simulations using present day and pre-industrial aerosols in our coupled global NCAR CAM-aerosol model compared to a cloud resolving model.
In the first paper, a present-day case of thin, warm marine-boundary-layer (MBL) clouds is simulated by a cloud-system resolving model (CSRM) and is compared to the same case of clouds simulated by the NCAR general circulation model (GCM). The simulation by the CSRM adopts a higher resolution and more advanced mirophysics as compared to that of the GCM, enabling the CSRM-simulation to act as a benchmark to assess the simulation by the GCM. Explicitly simulated interactions among the surface latent heat (LH) fluxes, buoyancy fluxes, and cloud-top entrainment lead to the deepening-warming decoupling and thereby a transition from stratiform clouds to cumulus clouds in the CSRM. However, in the simulation by the GCM, these interactions are not resolved and thus the transition to cumulus clouds is not simulated. This leads to substantial differences in cloud mass and radiation between simulations by the CSRM and the GCM. When stratocumulus clouds are dominant prior to the transition to cumulus clouds, interactions between supersaturation and cloud droplet number concentration (CDNC) (controlling condensation) and those between rain evaporation and cloud-base instability (controlling cloud dynamics and thereby condensation) determine cloud mass and thus the radiation budget in the simulation by the CSRM. These interactions result in smaller condensation and thus smaller cloud mass and reflected solar radiation by clouds in the simulation by the CSRM than in the simulation by the GCM where these interactions are not resolved. The resolved interactions (associated with condensation and the transition to cumulus clouds) lead to better agreement between the CSRM-simulation and observation than that between the GCM-simulation and observation.

In the 2nd paper, the response of the case of thin, warm marine-boundary-layer (MBL) clouds to preindustrial (PI) and present-day (PD) conditions is simulated by a cloud-system resolving model (CSRM) and compared to that simulated by a GCM.

The percentage increase of liquid-water path (LWP) due to a change from the PI to PD conditions is ~ 3 times larger in the CSRM than that in the GCM due to the formation of cumulus clouds. The formation of

cumulus clouds is controlled by a larger increase in the surface latent-heat (LH) flux in the PD environment than in the PI environment rather than by the change in aerosols. However, the aerosol increase from the PI to PD level determines the LWP response in the stratocumulus clouds, while the impacts of changes in environmental conditions are negligible for stratocumulus clouds. The conversion of cloud liquid to rain through autoconversion and accretion plays a negligible role in the CSRM in the response to aerosols, whereas it plays a role that is as important as condensation in the GCM.

Supplementary simulations show that increasing aerosols increase the sensitivity of the cloud responses to the PI and PD environmental conditions and that aerosol effects on clouds depend on the cloud type; the liquid water path (LWP) of warm cumulus clouds is more sensitive to aerosols than the LWP of stratocumulus clouds.

Lee, S. S., J.E. Penner, and M. Wang, 2009: Comparison of a global-climate model simulation to a cloud-system resolving model simulation for long-term thin stratocumulus clouds, Atmos. Chem. Phys., 9, 6497-6520.

Lee, S. S. and J. E. Penner, 2010: Comparison of a global-climate model to a cloud-system resolving model for the long-term response of thin stratocumulus clouds to preindustrial and present-day aerosol conditions, Atmos. Chem. Phys., 10, 6371-6389.

(2) Published a paper examining whether aerosol chemistry or size or other factors were more important in determining the first aerosol indirect effect.

We used an aerosol composition dataset that encompasses the U.S.A (the IMPROVE network) and explored the impact of changing both the size distribution as well as the composition of the aerosols on the droplet concentrations (N_D) predicted using a warm parcel model. We ran the parcel model through five vertical velocities, aerosol compositions from four seasons, varying aerosol number concentrations, and varying HNO_3 gas concentrations in 23 sensitivity studies. We analyzed calculated droplet concentrations within the U.S.A. and showed that differences in aerosol composition and changing size distribution parameters cause significant differences in N_D between regions. Non-uniform deviations appear between test cases and base cases across a vertical velocity range of 10 to 300 cm/s. Changing the fraction of water-soluble organic carbon (WSOC) to create an initial externally mixed aerosol population was found to be the variable causing greatest changes in droplet concentrations, after that caused by changing initial aerosol number concentrations. Our results show that the aerosol composition and the treatment of WSOC play dominant roles in the prediction of droplet concentration from aerosols.

Roesler, E. and J. E. Penner, 2010: Can global models ignore the chemical composition of aerosols?, Geophys. Res. Lett., 37, L24809, doi:10.1029/2010GL044282.

(3) Published a paper examining how the differences in thunderstorm development and stratocumulus clouds leads to differences in their response to aerosols.

We showed that the morphological differences (specifically the differences in depth) between thunderstorm clouds and stratocumulus clouds lead to differences in the response of precipitation to changes in aerosols. Increasing aerosols are known to suppress the conversion of droplets to rain (i.e., socalled autoconversion). This increases droplets as a source of evaporative cooling, leading to an increased intensity of downdrafts. The acceleration of the intensity of downdrafts is larger in convective clouds due to their larger cloud depths (providing longer paths for downdrafts to follow to the surface) than in stratiform clouds. More accelerated downdrafts intensify the gust front, leading to significantly increased updrafts, condensation and thus the collection of cloud liquid by precipitation, which offsets the suppressed autoconversion. This leads to an enhancement of precipitation with increased aerosols in convective clouds. However, the downdrafts are less accelerated in stratiform clouds due to their smaller cloud depths, and they are not able to induce changes in updrafts as large as those in convective clouds. Thus the offset is not as effective, and this allows the suppression of precipitation with increased aerosols.

Lee, S.S., L.J. Donner, and J.E. Penner, 2010: Thunderstorm and stratocumulus: how does their contrasting morphology affect their interactions with aerosols?, Atmos. Chem. Phys., 10, 6819-6837.

(4) Published a paper examining the meteorological conditions and the response of cirrus clouds to changes in aerosols.

We examined the response of large scale cirrus clouds to changes in aerosols. In these clouds, intensified interactions among the cloud ice number concentration (CINC), deposition and dynamics play a critical role in the ice water path (IWP) increases due to aerosol increases. Increased aerosols lead to increased CINC, providing increased surface area for water vapor deposition. The increased deposition causes depositional heating which produces stronger updrafts, and leads to the increased IWP. The conversion of ice crystals to aggregates through autoconversion and accretion plays a negligible role in the IWP responses to aerosols, as does the sedimentation of aggregates. The sedimentation of ice crystals plays a more important role in the IWP response to aerosol increases than the sedimentation of aggregates, but, not more important than the interactions among the CINC, deposition and dynamics.

Lee, S. S. and J. E. Penner, 2010: Aerosol effects on ice clouds: Can the traditional concept of aerosol indirect effects be applied to aerosol-cloud interactions in cirrus clouds?, Atmos. Chem. Phys., 10, 10345-10358.

(5) Submitted a paper examining how changes in solar radiation change aerosol cloud interactions in thin stratocumulus clouds with liquid water path (LWP) < 50 gm^{-2}.
As solar radiation increases, decoupling within the marine boundary layer (MBL) becomes stronger, leading to less condensation and less LWP and thus the absence of the surface precipitation. This enables the evaporation of rain to affect the cloud-base instability. In these cases without surface precipitation, as rain evaporation increases due to more conversion of cloud liquid to rain in the PI case, the cloud-base instability increases and thus updrafts increase which leads to larger LWP in the PI case than in the PD case. In cases with surface precipitation (at decreased solar radiation), the in-cloud interactions among cloud droplet number concentration (CDNC), supersaturation, and updrafts play an important role in the effect of aerosols on the LWP; these in-cloud interactions produce larger LWP with the PD aerosols than with the PI aerosols.

Lee, S. S. and J. E. Penner, 2009: Impact of solar radiation on aerosol-cloud interactions in thin stratocumulus clouds, Atmos. Chem. Phys. Discuss., 9, 23791-23833.

(6) Submitted a paper describing the factors that determine the effects of aerosols on total cloud mass and how they depend on the liquid water path (LWP).
Increasing aerosols decreases the size of droplets and thus their collection efficiencies, leading to an inefficient conversion of droplets to precipitable raindrops. This, in turn, increases the mass of droplets suspended in the air by decreasing the removal of cloud mass by sedimentation and has been known to be one of the main mechanisms which determines the effect of aerosols on cloud mass. However, Lee et al. (JGR, 2009) showed that this mechanism played a negligible role in the determination of the cloud mass as compared to aerosol-induced feedbacks between microphysics and dynamics in thin stratocumulus clouds with LWP of ~ 50 g m^{-2} or less. This was contrary to studies which have shown that the mechanism associated with the aerosol-induced inefficient conversion plays an important role in the determination of the effect of aerosols on cloud mass. These studies are generally based on clouds with LWP > 50 g m^{-2}. Hence, it is important to understand whether the role of aerosol-induced feedbacks in the effect of aerosols on cloud mass depends on the level of LWP. This study examines the dependence of the role of the conversion of droplets to raindrops and their sedimentation in the determination of the effect of aerosols on cloud mass on the level of LWP. Pairs of numerical experiments for high and low aerosol cases are run for four cases of stratiform clouds with different LWPs. Comparisons among these cases show that the role of the conversion of cloud droplets to precipitable raindrops and sedimentation becomes less important as the level of LWP decreases. Instead, the role of the feedbacks between microphysics and dynamics become more important with the decreasing level of LWP.

The results of this study indicate that the traditional approach to the understanding of the aerosol-cloud interactions and its application to the parameterization of these interactions in climate models can be

misleading. The understanding of feedbacks between microphysics and dynamics induced by aerosol changes and their parameterization can be critical to the correct assessment of the effect of aerosols on clouds and climate.

Lee, S. S. and J. E. Penner, 2009: Factors determining the effect of aerosols on cloud mass and the dependence of these factors on liquid-water path, Atmos. Chem. Phys. Discuss., 9, 19313-19350.
This paper has been revised and re-submitted to Atmospheric Environment:
Lee, S. S. and J. E. Penner, 2011: Dependence of aerosol-cloud interactions in stratocumulus clouds on liquid-water path, Atmos. Environ., submitted.

(7) Published a paper analyzing the response of thin stratocumulus clouds to changes in aerosols using a cloud resolving model.

Thin clouds with mean liquid-water path (LWP) of ~ 50 g m^{-2} cover 27.5 % of the globe and thus play an important role in the Earth's radiation budget. Radiative fluxes at the Earth's surface and top of atmosphere (TOA) are very sensitive to the LWP variation when the LWP becomes smaller than ~ 50 g m^{-2}, so aerosol effects on thin clouds can have a substantial impact on the variation of global radiative forcing if LWP changes. This study examines the aerosol indirect effect (AIE) through changes in the LWP in three cases of thin warm stratocumulus clouds with LWP < 50 g m^{-2}. We use a cloud-system resolving model (CSRM) coupled with a double-moment representation of cloud microphysics. Intensified interactions among the cloud droplet number concentration (CDNC), condensation and dynamics at high aerosol play a critical role in the LWP responses to aerosol increases. Increased aerosols lead to increased CDNC, providing the increased surface area of droplets where water vapor condenses. This increases condensation and thus condensational heating to produce stronger updrafts, leading to an increased LWP with increased aerosols in two of the cases where precipitation reaches the surface. In a case with no surface precipitation, LWP decreases with increases in aerosols. In this case, most of precipitation evaporates just below the cloud base. With decreases in aerosols, precipitation increases and leads to increasing evaporation of precipitation, and thereby, increasing instability around cloud base. This leads to increased updrafts and thus condensation from which increased LWP results..

Lee, S. S., J.E. Penner, and S.M. Saleeby, 2009: Aerosol effects on liquid-water path of thin stratocumulus clouds, J. Geophys. Res., 114, D07204, doi:10.1029/2008JD010513.

8) Published two papers with our global aerosol model coupled to the NCAR climate model.

In the first paper, the simulated aerosol fields from a coupled aerosol/atmospheric circulation model that includes prediction of both sulfate aerosol size and number are evaluated. Sensitivity tests were used to evaluate uncertainties due to the inclusion of primary-emitted particulate sulfate as a means of representing nucleation of particles in sub-grid scale plumes, the use of two empirical boundary layer aerosol nucleation mechanisms, and a 3-mode sulfate aerosol representation. Primary-emitted particulate sulfate contributes significantly to aerosol number at sites located in the boundary layer over Europe, but the absence of constraints on the number of such particles from either observations or fine-resolution models makes this treatment for parameterization of nucleation of new particles undesirable. The use of an empirical boundary layer nucleation mechanism improves the comparison of simulated aerosol number concentrations with observations in the marine boundary layer, suggesting that a treatment of boundary layer nucleation is needed in global aerosol models although more studies are needed to quantify how different nucleation mechanisms and condensable gases other than sulfuric acid affect aerosol number. The 3-mode representation of sulfate aerosol simulates the observed increase in accumulation mode number concentration with altitude in the upper troposphere, and improves the simulated Aitken mode aerosol number concentration there. This indicates the importance of a separate representation of freshly nucleated particles when nucleation is an important source of particle number concentrations.

The second paper calculated the first aerosol indirect effect associated with different forms of boundary layer nucleation treatments. Here, we used the version of the global aerosol model that included the empirical boundary layer nucleation mechanism, the use of primary-emitted sulfate particles to represent sub-grid scale nucleation, as well as binary homogeneous nucleation to explore how nucleation affects the

CCN concentration and the first aerosol indirect effect (AIE). The inclusion of the empirical boundary layer nucleation scheme increases the global average CCN concentrations in the boundary layer by 31.4% when no primary-emitted sulfate particles are included and by 5.3% when they are included. Particle formation with the boundary layer nucleation scheme decreases the first indirect forcing over the ocean, and increases the first indirect forcing over land when primary sulfate particles are included. This suggests that whether particle formation from aerosol nucleation increases or decreases aerosol indirect effects largely depends on the relative change of primary particles and SO2 emissions from the preindustrial to the present day atmosphere. Including primary-emitted sulfate particles significantly increases both the anthropogenic fraction of CCN concentrations and the first aerosol indirect forcing. The forcing from various treatments of aerosol nucleation ranges from -1.22 to -2.03 W/m^2. This large variation shows the importance of better quantifying aerosol nucleation mechanisms for the prediction of CCN concentrations and aerosol indirect effects.

Wang, M., J.E. Penner, and X. Liu, 2009: Coupled IMPACT aerosol and NCAR CAM3 model: Evaluation of predicted aerosol number and size distribution, J. Geophys. Res., 114, D06302, doi:10.1029/2008JD010459.

Wang, M. and J. E. Penner, 2009: Aerosol indirect forcing in a global model with particle nucleation, Atmos. Chem. Phys., 9, 239-260, 2009, www.atmos-chem-phys.net/9/239/2009/.

9) Published a paper on our study of the impact of aerosols on clouds during the May 2003 Intensive Operational Period at the Southern Great Plains.

The Active Tracer High-resolution Atmospheric Model was used to examine the aerosol indirect effect (AIE) for a spring continental stratus cloud based on data during the May 17, 2003 Aerosol Intensive Operation Period (AIOP) at the Atmospheric Radiation Measurement (ARM) Program Southern Great Plains site. Model results for our base case, which uses observed aerosol concentrations, agree reasonably well with the available observations, giving confidence that the basic model is reasonable. Sensitivity tests are performed to explore the response of the clouds to changes in the aerosol number concentration and surface fluxes. During the major part of the simulation, from 6:30 am through 14:00 local time, an increase in the aerosol number concentration (N_a) results in a decrease of the mean cloud droplet size, and an increase of the cloud liquid water path (LWP) until aerosol number concentration levels reach 1200 cm^{-3}. Further increases in aerosol concentration do not increase the liquid water path because the depletion of cloud water by precipitation is negligible above this number concentration. After 14:00, the liquid water path decreases when aerosols increase as long as $N_a < 600$ cm^{-3} and remains unchanged for higher aerosol concentrations. The decrease of LWP is associated with the evaporative cooling below cloud base which leads to more condensation of water vapor, a result that is consistent with afternoon satellite observations of the response of continental clouds to increases in droplet concentrations. A sensitivity test with a stronger surface latent flux increases both the cloud geometrical thickness and cloud water content. On the other hand, a sensitivity test with a stronger surface sensible heat flux leads to a higher cloud base, and a shallower and drier cloud. The response of the cloud geometrical thickness to changes in surface sensible heat flux dominates that of the cloud water content. The cloud fraction is also reduced at the end of the simulation time period. Because the surface heat fluxes will likely change when aerosol and droplet number concentrations change, these sensitivity tests showed that a fully-coupled simulation with a land surface model will be needed to fully assess the response of the cloud to changing aerosol concentrations. Nevertheless, since the thermodynamic boundary layer profiles do not change significantly when aerosol concentrations are changed, our results for changing aerosol concentrations are qualitatively correct.

Guo, H., J. E. Penner, M. Herzog, and S. Xie: 2007, Investigation of the first and second aerosol indirect effects using data from the May 2003 Intensive Operational Period at the Southern Great Plains, J. Geophys. Res., 112, D15206, doi:10.1029/2006JD007173.

(10) Published a paper to examine present day and pre-industrial simulations with our coupled global aerosol model including prediction of new sulfate particles via binary nucleation with the NCAR CAM model. Examined whether the predicted changes in sulfate aerosol particle size affected the estimate of the first indirect effect.

We have previously published our scheme for treating the nucleation of new particles by binary heterogenous nucleation of sulfate and examined the predicted size distribution of the aerosol in the

IMPACT aerosol model (Liu et al., J. G. R., 2005). Here, we employed this model in the coupled NCAR CAM3-IMPACT model and examined whether the predicted size changes the calculated first aerosol indirect effect. We found that the predicted cloud droplet number concentration and size compared well with satellite observations from MODIS. We used the predicted pre-industrial aerosol size distribution in the present day simulations and found that the global average change in the first indirect effect was 0.03 Wm^{-2}, but it was as large as 2 Wm^{-2} in some regions. If we use a size distribution based on an average "observed" size distribution, the changes were larger. We also examined a case where we fixed the aerosol composition by fixing the hygroscopicity. This had a somewhat larger impact on the global average forcing (0.1 Wm^{-2}), showing that composition cannot be ignored in determining the aerosol indirect effect.

Wang, M. and J. E. Penner, 2009: Aerosol indirect forcing in a global model with particle nucleation, Atmos. Chem. Phys., 9, 239-260, www.atmos-chem-phys.net/9/239/2009/.