

# GACP 3<sup>rd</sup> YEAR PROGRESS REPORT AND SUMMARY

TITLE: Global Modeling of Tropospheric Aerosols and Their Contribution to Climate Variation

Name: Catherine C. Chuang

Institution: Atmospheric Science Division (L-103)  
Lawrence Livermore National Laboratory  
Livermore, CA 94550

## GOALS

The goals of this project are (1) to study the global aerosol cycle and to make a more quantitative evaluation of the effects of aerosol emissions on the Earth's radiative balance; and (2) to identify and analyze the responsible processes in aerosol/cloud/climate interactions.

## ACCOMPLISHMENT

### (1) Model Enhancement

We have revised our coupled climate/chemistry models to examine the forcing by all primary aerosol components. The revised model now calculates the following aerosol compounds: sulfate, dust, sea salt, and carbonaceous aerosols (biomass and fossil fuel organic and black carbon). This new version was also used to initiate a study for the direct forcing assessment that covers the years 1950 to present using the varying fossil fuel emissions.

Much of our effort also went to model the effect of absorbing aerosols on clouds. The presence of black carbon in cloud may reduce the cloud albedo. It is important not only to account for the change in droplet concentration associated with carbonaceous aerosols acting as cloud condensation nuclei, but also to account for the absorption that occurs when black carbon becomes associated with drops. We modeled the BC absorbing effect on clouds through modification of the refractive index for a droplet/BC mixture using the effective medium approximation together with the Maxwell Garnett mixing rule. Given the refractive index for the droplet with embedded BC particles, we approximated its single scattering albedo using geometric optics. By comparison with the single scattering albedo tables, we developed a parameterization of the single scattering albedo for a droplet/BC mixture as a function of the drop radius, volume fraction of BC within the drop, and the wavelength of solar radiation (see Figure 1). This parameterization was incorporated into our coupled models to evaluate the impact of absorbing particles on aerosol indirect forcing.

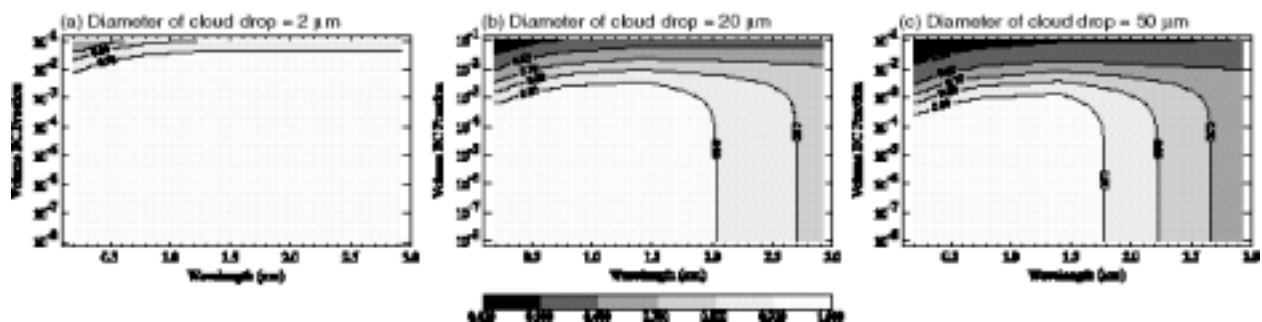
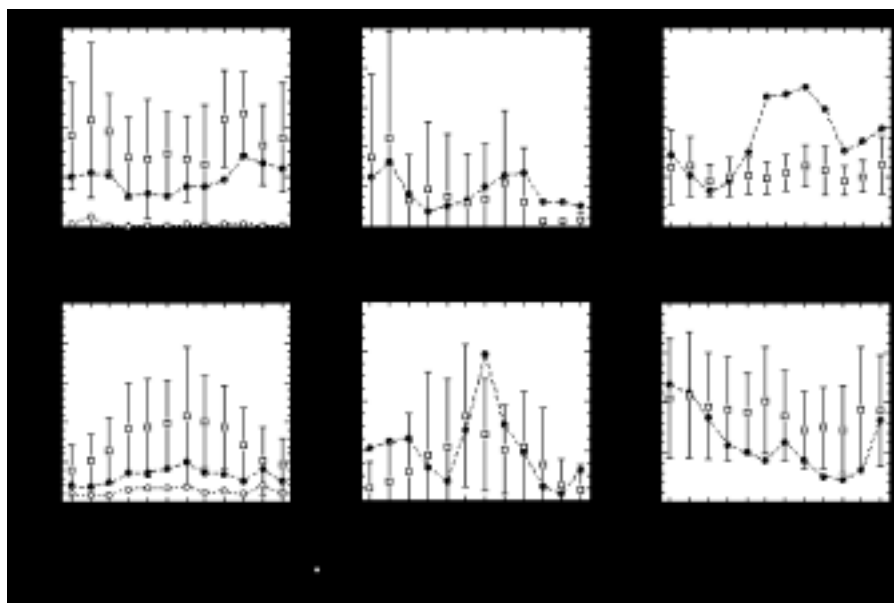


Figure 1. Variations of single scattering albedo of cloud drops as a function of volume BC fraction and wavelength.

(2) *Comparisons to In-Situ Measurements*

Model simulated aerosol concentrations were compared to measurements to validate the adequacy of the model. Figure 2 presents comparisons of simulated seasonal surface concentrations of non-seasalt sulfate, dust, and sea salt to measurements at American Samoa and Barbados [Savoie and Prospero, private communication, 2000]. Most of the simulated concentrations are within one deviation of the mean of the measurements, but discrepancies do exist. Figure 3 shows the observed and simulated surface concentrations for both organic carbon and black carbon at a number of locations [Lioussé et al., 1996; Cooke et al., 1999]. Comparisons of carbonaceous aerosols to observations are more difficult because the measured concentrations are only available on a campaign basis such that the measured values are subject to short term variability. Despite these facts, most simulated values are within a factor of 2 of the measurements.



Figures 2. Simulated concentrations versus measurements at American Samoa and Barbados. Error bars are one standard deviation above and below the mean of the measurements. Also shown are the simulated concentrations of anthropogenic sulfate (open circles).

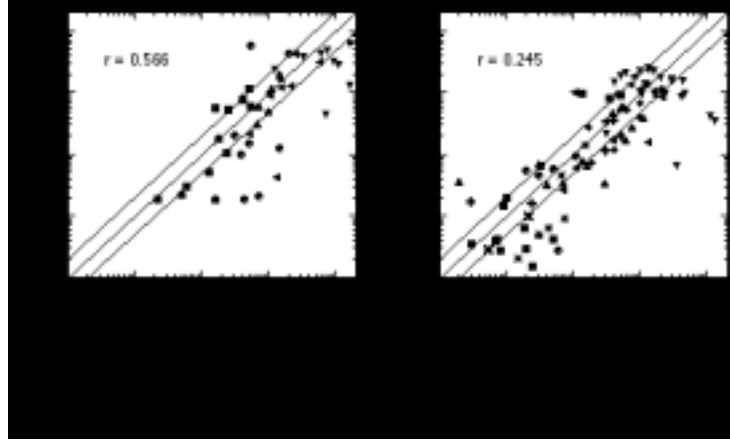


Figure 3. Observed and simulated surface concentrations of OC and BC. Correlation is presented by  $r$ . Two thin lines represent the range above and below the observed values by a factor of 2.

### (3) Impact of Anthropogenic Aerosols on Cloud Number Concentration and Susceptibility

A modified cloud drop parameterization from *Chuang et al.* [1997] was applied into our coupled models to better represent the number concentration of cloud drops ( $N_d$ ) nucleated on aerosols with different size distributions. We found that the increases of  $N_d$  can be up to  $200 \text{ cm}^{-3}$  due to anthropogenic carbonaceous aerosols and up to  $30 - 90 \text{ cm}^{-3}$  due to change of aerosol size distribution associated with the deposition of anthropogenic sulfate on pre-existing particles. Our simulations also indicate that the presence of industrial aerosols has significantly reduced cloud susceptibility, i.e., the sensitivity of cloud albedo to changes in  $N_d$ , in the northern hemisphere in January, and biomass aerosols have reduced susceptibility in the southern hemisphere in July.

Figure 4 presents the susceptibility for warm clouds and those derived from the satellite-retrieved cloud optical thickness  $\tau$  and effective drop radius  $r_e$ . These data were retrieved from 1989 - 1991 AVHRR measurements using the algorithm developed by *Kawamoto et al.* [2001]. The liquid water path is calculated from  $w_{\text{path}} = (2/3)\rho \tau r_e$ . The column  $N_d$  is estimated from  $N_{d,\text{col}} = \tau r_e / (2\pi r_v^3)$ , where  $r_v$  is the volume mean radius of the drop size distribution. Here, we assume a log-normal drop size distribution with a standard deviation  $\sigma = 1.4$  and a mode radius  $r_o = r_e \exp[-2.5 (\ln \sigma)^2]$ . For a vertically homogeneous drop profile,  $N_{d,\text{col}} = N_{d,s} h_s$  and  $w_{\text{path}} = w_L h_s$ , where  $N_{d,s}$  and  $h_s$  are the satellite-retrieved drop concentration and cloud thickness for warm clouds, respectively. A typical value of liquid water content  $w_L = 0.30 \text{ g m}^{-3}$  is used to estimate  $h_s$  and then obtain the satellite-retrieved  $N_{d,s}$ . General features of the satellite-derived cloud susceptibility are similar to those of the model, though the magnitude is higher by about a factor of 2 in most of the regions except over the South Pacific Ocean in July. This discrepancy may be caused by the uncertainty in the prescribed drop size distribution where the satellite-retrieved  $N_{d,\text{col}}$  varies with the assumed value of  $\sigma$ . For  $r_e = 10 \text{ }\mu\text{m}$ , the retrieved  $N_{d,\text{col}}$  would be 27% lower if  $\sigma$  decreases from 1.4 to 1.1 and would be higher by a factor of 3 if  $\sigma = 2.0$ . There are further uncertainties associated with the prescribed liquid water content and the retrieved cloud top temperature. Further investigation is needed to give an overall uncertainty of the estimated cloud susceptibility derived from both the model and satellite measurements.

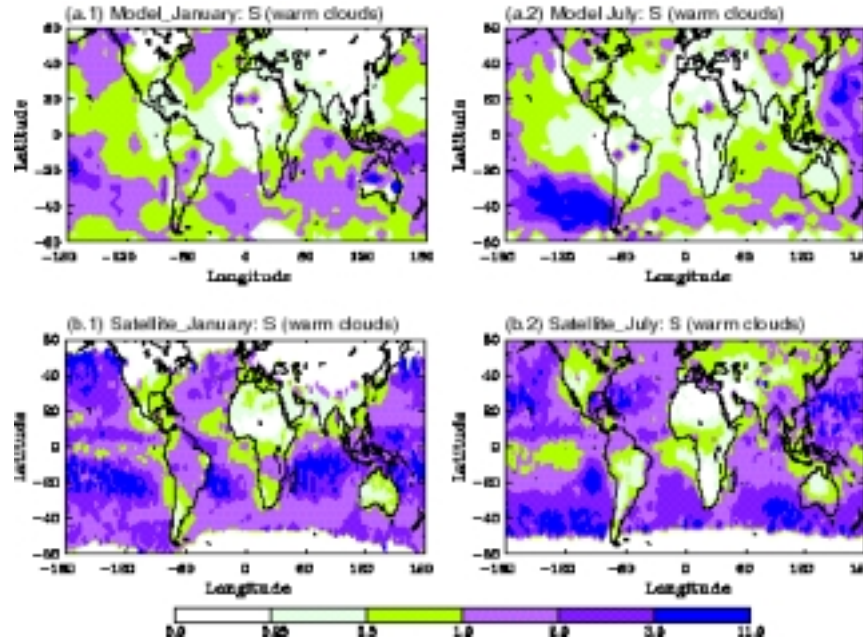


Figure. 4. Comparison of model calculated cloud susceptibility ( $\times 10^{-3} \text{ cm}^3$ ) for warm clouds with those inferred from satellite measurements.

(4) *First Indirect Forcing by Anthropogenic Aerosols*

Figure 5a presents the simulated first indirect forcing by anthropogenic carbonaceous aerosols for January and July. In general, the forcing in July is stronger than that in January, and yields a global average of  $-1.59 \text{ W m}^{-2}$  and  $-1.05 \text{ W m}^{-2}$ , respectively. The maximum value is about  $-7.7 \text{ W m}^{-2}$  along the west coast of Mexico in January and  $-8.6 \text{ W m}^{-2}$  along the east coast of Brazil in July. Anthropogenic carbonaceous aerosols together with natural particles are treated as an external mixture in the cloud drop parameterization and lead to an annual average forcing of  $-1.51 \text{ W m}^{-2}$ . This value is much lower than our previous study ( $-2.5$  to  $-4.5 \text{ W m}^{-2}$ , see Penner *et al.*, 1996) in which part of natural emissions were absent. Figure 5b shows the first indirect forcing by anthropogenic sulfate deposited onto pre-existing particles derived from natural emissions and anthropogenic carbonaceous sources. The maximum forcing is about  $-1.6 \text{ W m}^{-2}$  in January and  $-5.1 \text{ W m}^{-2}$  along the east coast of the United States. While the forcing pattern is similar to that calculated previously in Chuang *et al.* [1997], the forcing magnitude is considerably smaller and leads to a global annual average of  $-0.30 \text{ W m}^{-2}$ . Current calculations used the model-generated aqueous sulfate production rate. This rate is approximately 88% of the total sulfate source strength. These simulations may, therefore, be compared to the previously calculated forcing of  $-0.41 \text{ W m}^{-2}$  for a case of prescribed 85% sulfate production through the aqueous pathway [Chuang *et al.*, 1997]. The present values are smaller both because of the larger source strength for natural organic aerosols and because of the presence of sea salt and dust. Figure 6 indicates that the global average of the first indirect forcing by total anthropogenic aerosols is largest in April-June associated with tropical biomass burning of savanna and forested areas. Its magnitude varies seasonally from  $-1.2 \text{ W m}^{-2}$  in January to  $-2.4 \text{ W m}^{-2}$  in May and yields a global annual average of  $-1.85 \text{ W m}^{-2}$ . Because of the nonlinear relationship between cloud drop number and aerosol number concentrations, the total forcing does not equal the sum of the forcing from each individual source.

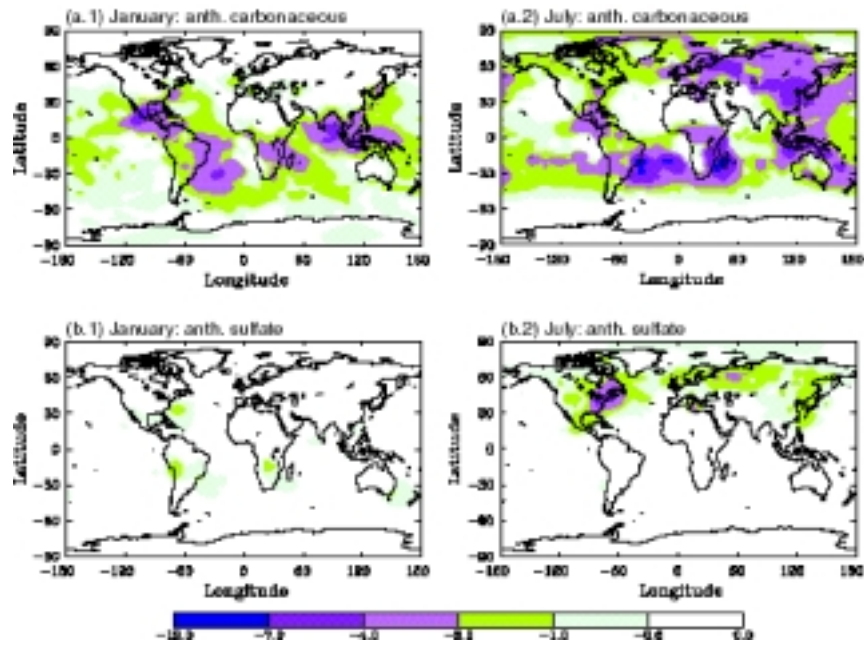


Figure 5. Simulated monthly average aerosol indirect forcing ( $W m^{-2}$ ) due to (a) externally mixed anthropogenic carbonaceous aerosols and (b) deposition of anthropogenic sulfate onto pre-existing particles (natural particles + anthropogenic carbonaceous particles).

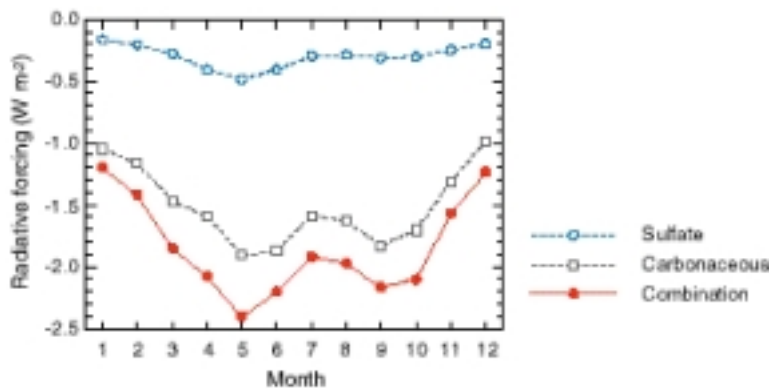


Figure 6. Seasonal variations of simulated global average of the first indirect forcing ( $W m^{-2}$ ).

## References

- Chuang, C. C., J. E. Penner, K. E. Taylor, A. S. Grossman, and J. J. Walton, 1997: An assessment of the radiative effects of anthropogenic sulfate, *J. Geophys. Res.*, **102**, 3761-3778.
- Cooke, W. F., C. Liou, H. Cachier, and J. Feichter, 1999: Construction of a 1 degree x 1 degree fossil fuel emission data set for carbonaceous aerosol and implementation and radiative impact in the ECHAM4 model, *J. Geophys. Res.*, **104**, 22,137-22,162.
- Kawamoto, K., T. Nakajima, and T. Y. Nakajima, 2001: A global determination of cloud microphysics with AVHRR remote sensing, *J. Clim.*, **14**, 2054-2068.

- Liou, S. S., J. E. Penner, C. C. Chuang, J. J. Walton, H. Eddleman, and H. Cachier, 1996: A global three-dimensional model study of carbonaceous aerosols, *J. Geophys. Res.*, **101**, 19,411-19,432.
- Penner, J. E., C. C. Chuang, and C. Liou, 1996: The contribution of carbonaceous aerosols to climate change, in *Proceedings of the 14th International Conference on Nucleation and Atmospheric Aerosols*, edited by M. Kulmala and P. E. Wagner, pp. 759-769, Elsevier Sci., New York.

## GACP BIBLIOGRAPHY

### (1) List of refereed papers and book chapters

- Penner, J. E., C. C. Chuang, and K. E. Grant, 1998: Climate forcing by carbonaceous and sulfate aerosols. *Clim. Dyn.*, **14**, 839-851.
- Lohmann, U., J. Feichter, C. C. Chuang, J. E. Penner, 1999: Prediction of the number of cloud droplets in a general circulation model by using different aerosol species. *Geophys. Res.*, **104**, 9169-9198.
- Lohmann, U., J. Feichter, C. C. Chuang, and J. E. Penner, 1999: Erratum. *J. Geophys. Res.*, **104**, 24,557-24,563.
- Grant, K. E., C. C. Chuang, A. S. Grossman, and J. E. Penner, 1999: Modeling the spectral optical properties of ammonium sulfate and biomass burning aerosols: Parameterization of relative humidity effects and model results. *Atmos. Environ.*, **33**, 2603-2620.
- Penner, J. E., Chuang, C. C., and K. Grant, 1999: Climate Change and Radiative Forcing by Anthropogenic Aerosols: A summary of current understanding. Submitted to La Jolla International School of Science, The Institute for Advanced Physics Studies, La Jolla, California, March 29-30, 1999.
- Chin, H.-N. S., D. J. Rodriguez, R. T. Cederwall, C. C. Chuang, J. J. Yio, Q. Fu, and M. A. Miller, 2000: A study on the retrieval of continental low-level stratiform clouds: Impact of sub-adiabatic character on the macroscopic microphysical properties and radiation budgets. *Mon. Wea. Rev.*, **128**, 2511-2527.
- Chuang, C. C., J. E. Penner, J. M. Prospero, K. E. Grant, G. H. Rau, and K. Kawamoto, 2001: Cloud susceptibility and the first aerosol indirect forcing: Sensitivity to black carbon and aerosol concentrations, submitted to *J. Geophys. Res.*
- Penner, J. E., S. Y. Zhang, M. Chin, C. C. Chuang, J. Feichter, Y. Feng, P. Ginoux, M. Herzog, A. Higurashi, D. Koch, C. Land, U. Lohmann, M. Mishchenko, T. Nakajima, G. Pitari, B. Soden, I. Tegen, and L. Stowe, 2001: A comparison of model- and satellite-derived optical depth and reflectivity, submitted to *J. Atmos. Sci.*
- Chuang, C. C., K. E. Grant, and J. E. Dignon, 2001: A scenario study of modeling the past and future direct aerosol forcing, to be submitted to *Atmos. Environ.*

### (2) List of non-refereed papers

- Lohmann, U., J. Feichter, C. C. Chuang, J. E. Penner, 1998: Using different aerosol species for activation of cloud droplet in a general circulation model. *Proceedings of the Conference on Cloud Physics and 14th Conference on Planned & Inadvertent Weather Modification*, August 17-21, 1998, Everett, Washington, 227-230.
- Chuang, C. C., and J. E. Penner, 1998: Use of transport modeling in integrated climatologies. Report for the 1st GACP Science Team Meeting, November 18-20, 1998, New York, New York. ([http://gacp.giss.nasa.gov/discussion/session\\_B.html](http://gacp.giss.nasa.gov/discussion/session_B.html))

- Chuang, C. C., 1999: Global modeling of tropospheric aerosols and their contribution to climate variation. First year report submitted to the NASA Global Aerosol Climatology Program, August, 1999.
- Chuang, C. C., J. E. Penner, and Y. Zhang, 2000: Simulations of aerosol indirect effect for IPCC emissions scenarios, *Proceedings of the 11th Symposium on Global Change Studies*, January 9-14, 2000, Long Beach, California, 320-323.
- Zhang, Y., J. E. Penner, C. C. Chuang, B. D. Santer, and K. Taylor, 2001: Changes in the vertical temperature structure associated with carbonaceous aerosols, *Proceedings of the 81 AMS Annual Meeting*.

(3) List of presentations at professional society meetings and conferences.

- Chuang, C. C., J. Dignon, and K. E. Grant, 1998: Climate forcing by the increasing anthropogenic sulfate aerosols in Asia, Presented at the Western Pacific Geophysics Meeting, July 21-24, 1998, Taipei, Taiwan.
- Chuang, C. C., J. Dignon, and K. E. Grant, 1998: Modeling temporal and regional variations of anthropogenic sulfate direct climate forcing during the past 40 years. Presented at the 5th Scientific Conference on the International Global Atmospheric Chemistry Project, August 19-25, Seattle, Washington.
- Chuang, C. C., 1998: Global modeling of tropospheric aerosols and their contribution to climate variation. Presented at the 1st Global Aerosol Climatology Project Science Team Meeting, November 18-20, 1998, New York, New York.
- Chuang, C. C., 1999: Modeling aerosol climatology at LLNL - Our capability, results, and perspective. Presented at the 1st LLNL/NCAR Meeting, January 7, 1999, Boulder, Colorado.
- Chuang, C. C., 1999: Aerosol distributions and indirect forcings for IPCC model scenarios. Presented at the IPCC WG1 Modeling Expert Meeting, July 15-16, 1999, Hamburg, Germany.
- Chuang, C. C., 1999: LLNL/Umich model capability and simulations for IPCC model scenarios. Presented at the 2nd Global Aerosol Climatology Project Science Team Meeting, September 29 - October 1, 1999, New York, New York.
- Chuang, C. C., 1999: IPCC sulfur comparisons. Presented at the 3rd LLNL/NCAR Meeting, October 28, 1999, Boulder, Colorado.
- Chuang, C. C., J. E. Penner, and K. E. Grant, 1999: Global Modeling of tropospheric aerosols and their contribution to climate variation. Presented at the AGU Fall Meeting, December 13-17, 1999, San Francisco, California.