FORM A: GACP ACCOMPLISHMENT REPORT

Name: Catherine C. Chuang

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

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

ABSTRACT

Atmospheric aerosols have significant effects on climate through their impact on the Earth's radiation budget. Part of our earlier studies have been focused on developing an understanding of global and regional sulfate aerosol abundances and investigating their direct and indirect climate effects with a fully coupled climate-aerosol model.

To address the radiative impact of other significant aerosol types on climate, we have developed emission inventories for carbonaceous and dust aerosols. We have also developed a unique annual emission inventory of anthropogenic SO_2 for the years 1950 through 1994. Moreover, we have modeled aerosol optical properties to account for changes in the refractive indices with relative humidity and dry aerosol composition and also developed parameterizations of cloud response to aerosol abundance to evaluate the importance of aerosol/cloud interactions on climate forcing. Based on our strength in this area, we propose to extend our previous effort to simulate the global distributions of all the important aerosol species and to assess their potential to mask part of the warming expected from greenhouse gas emissions.

GOALS

The goals of this proposal 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 climate; and (2) to identify and analyze the responsible processes in aerosol/cloud/climate interactions.

OBJECTIVES

The simulated aerosol concentrations and distributions will be validated with data available in in-situ campaigns and data retrieved from remote sensing instruments. We will examine the relative contribution of industrial and non-industrial anthropogenic sources as well as explore the difference in forcing pattern and magnitude by direct and indirect aerosol effects. In the later stages of this research we plan to contribute to integrated assessments of climate change by simulating the spatial pattern of response that would be expected from various future emission scenarios.

APPROACH

Our simulations require the use of a general circulation model (GCM) coupled to a global chemistry model. We have been systematically improving our ability to model anthropogenic aerosols and developed a method for treating the indirect forcing by anthropogenic aerosols. For simulation of climate response, the general circulation model is coupled to a 50-meter mixed-layer ocean and thermodynamic sea ice model with meridional oceanic heat flux prescribed. This model configuration is less susceptible to the troubling problem of "climate drift" found when atmospheric GCMs are coupled to full ocean general circulation models; it is also less expensive in terms of computer time. With our coupled model, we are able to predict the equilibrium response of climate to externally imposed changes, such as the concentration of trace gases or the emission rates of various aerosol precursors.

Recently, Lawrence Livermore National Laboratory has internally funded a project to implement an aerosol microphysics model into the LLNL's state-of-the-art parallel global chemistry model (IMPACT) to include the important aerosol dynamics as well as aerosol physical properties. This chemistry-aerosol model will be linked to the most updated version of the NCAR CCM. The new coupled model, available about a year from now, will replace the one we have been using in the past and at present. This new modeling capability will construct more realistic time-varying patterns of climate change for use in detection studies and place the estimate of aerosol climate effects on a much more quantitative foundation than is available at present.

TASKS COMPLETED

We have revised our climate/chemistry model to examine the forcing and response of the climate system to changes in 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 improved version was used to explore the climate forcings by both anthropogenic sulfate and carbonaceous aerosols. Furthermore, this new version was also used to initiate a study for the direct forcing assessment which 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. This parameterization was incorporated into our coupled model to evaluate the impact of absorbing particles on aerosol indirect forcing.

FUTURE PLANS:

Our project will proceed on three fronts. First, in order to identify the role of natural aerosols in the assessment of anthropogenic aerosol indirect forcing, we will examine the sensitivity of indirect forcing to aerosol characteristics and global concentration. Next, we will compare the simulated equilibrium pattern of climate response to the changes that have been observed over the past 20 years. Finally, in order to characterize future climate variation, we will perform simulations for different emission scenarios. We will investigate how a different spatial distribution of aerosol emissions (which is predicted to be more consistent with future fossil fuel consumption) might alter the pattern of climate response we simulate at stage two.

RESULTS

Our simulations indicate that the magnitude of the calculated direct aerosol forcing would be sensitive to the treatment for changes in refractive indices with relative humidity. It is found that the estimate of the direct forcing by anthropogenic sulfate aerosols can change from -0.81 Wm^{-2} to -0.55 Wm^{-2} if grid box average relative humidity is not allowed to increase above 90%.

The contribution of carbonaceous aerosols to direct climate forcing depends on how much organic carbon is associated with the black carbon. Our calculations show that the direct forcing associated with fossil fuel emissions of carbonaceous aerosols ranges from +0.16 to +0.20 Wm^{-2}, whereas the direct forcing of carbonaceous aerosols associated with biomass burning is calculated to range from -0.21 to -0.14 Wm^{-2}. However, these estimates have assumed that black carbon and sulfate remain externally mixed. As noted by Haywood and Shine (1995), an internal mixture of BC and sulfate would increase the amount of absorbed radiation, and could decrease the overall net forcing by 20 - 60%. Clearly further study of optical properties for internally mixed aerosols is needed to improve quantitatively our assessment of the aerosol direct forcing.

We also found that simulation without the BC absorbing effect on clouds could overestimate the *local* indirect forcing of anthropogenic carbonaceous aerosols by up to 20-25% over regions in Europe, Africa, and East China, although the change in the magnitude of global average is small (-1.44 Wm^{-2} with the BC effect versus -1.50 Wm^{-2} without the BC effect). Further investigation on the corresponding changes in cloud liquid water content and cloud lifetime is needed in order to better quantify the effects of aerosols on the radiation budget.

FORM B: GACP SIGNIFICANT HIGHLIGHTS

Name: Catherine C. Chuang

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

SIGNIFICANT HIGHLIGHTS

Geographical distributions of aerosol climate forcing simulated by our fully coupled climate/chemistry model are presented below. The global emissions of anthropogenic SO_2 and carbonaceous aerosols for use in our calculations are listed in Table 1.

(1) Simulated direct aerosol forcing (in Wm^{-2})

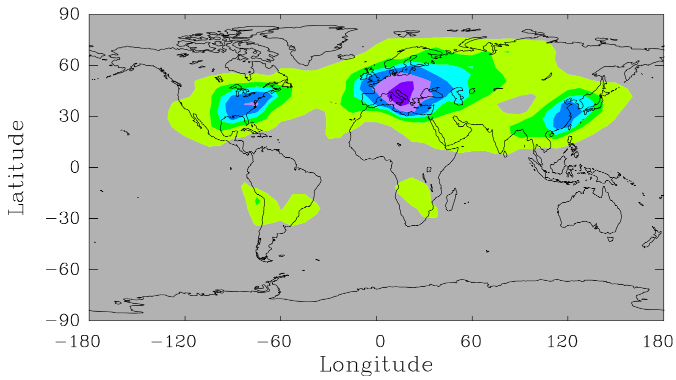

Fig. 1a. Anthropogenic sulfate aerosol forcing assuming a lognormal distribution with r_n = 0.05 μm and σ = 2. Global average = -0.81 Wm^{-2}.

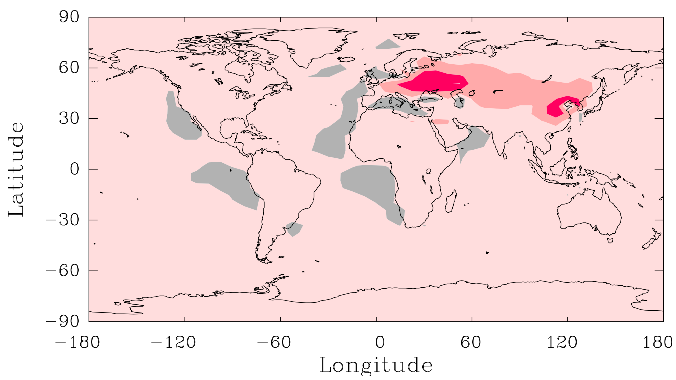

Fig. 1b. Carbonaceous aerosol forcing from fossil fuel emissions assuming a distribution measured for the Hardiman fire by Radke et al. (1988). Global average = 0.16 Wm^{-2}.

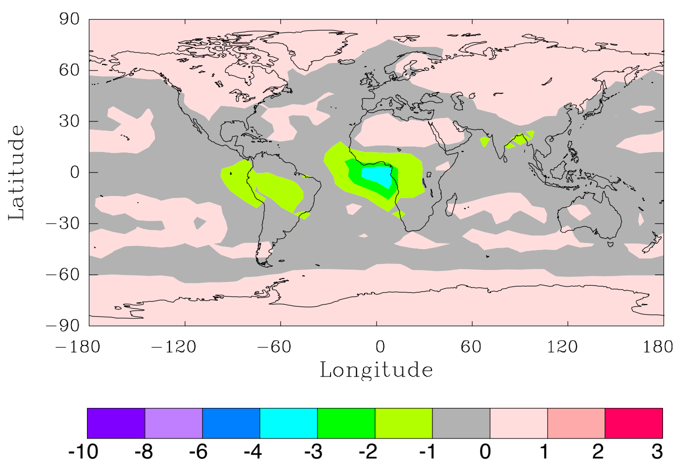

Fig. 1c. Carbonaceous aerosol forcing from biomass burning assuming the Anderson et al. (1996) size distribution. Global average = -0.21 Wm^{-2}.

(2) Simulated indirect aerosol forcing (in Wm^{-2}) associated with changes in cloud albedo

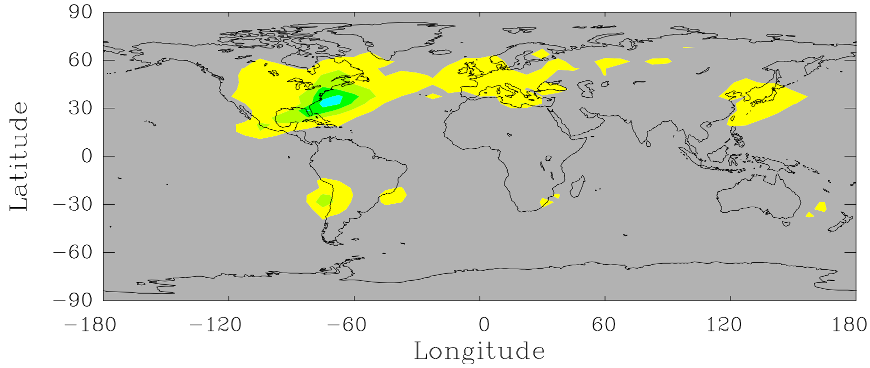

Fig. 2a. Anthropogenic sulfate forcing for internally mixed aerosols. Aerosol size distribution is assumed to vary with the deposition of anthropogenic sulfate either through condensation of H_2SO_4 or in-cloud oxidation of SO_2 on pre-existing particles which have a prescribed size distribution (Chuang et al., 1997). The total aerosol number concentration remains unchanged but particles would grow to larger size. The pre-existing particles are assumed to consist of particles from natural sources and anthropogenic carbonaceous aerosols. Global average = -0.22 Wm^{-2}.

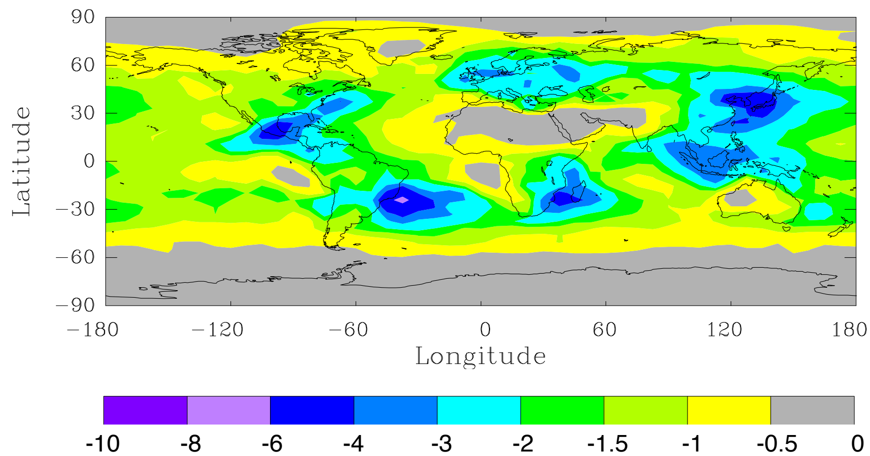

Fig. 2b. Anthropogenic carbonaceous aerosol forcing for externally mixed aerosols. It is assumed that the total aerosol number concentration is the sum of anthropogenic carbonaceous aerosols and the pre-existing particles derived from natural sources. Global average = -1.44 Wm^{-2}.

Table 1. Anthropogenic Sources of Sulfate and Carbonaceous Aerosols

	SO_2 (Tg S/yr)	Carbonaceous Aerosols (Tg/yr)	
		Organic Matter	Black Carbon
Fossil fuel and industry	75.7	30.6	6,6
Biomass burning	2.2	53.7	5.6

FORM C: FUTURE PLANS

Name: Catherine C. Chuang

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

In order to identify the role of natural aerosols in the assessment of indirect anthropogenic aerosol forcing associated with changes in cloud albedo, the first task we will carry forward for the second year is to examine the mechanisms that link aerosol sources to cloud drop number concentrations. Since the tropospheric lifetimes of aerosols and their precursor gases are short (on the order of only days or weeks), the mechanisms describing these links vary spatially and temporally. Moreover, the ranges of these aerosol loadings vary between the continents and oceans (and also within these regions), it is thus important to study the sensitivity of climate forcing to the aerosol characteristics and global concentration in order to identify the relative importance of natural particles derived from different sources.

Our second task for next year is to compare and validate our simulated aerosol concentrations and distributions with data available in in-situ campaigns such as the Atmosphere/Ocean Chemistry Experiment (AEROCE), Transport and Atmospheric Chemistry near the Equator-Atlantic (TRACE-A), Southern Hemisphere Aerosol Characterization Experiment (ACE-1), and Tropospheric Aerosol Radiative Forcing Observational Experiment (TARFOX) as well as data retrieved from remote sensing instruments such as the NOAA Advanced Very High Resolution Radiometer (AVHRR), Total Ozone Mapping Spectrometer (TOMS) and, if available, EOS detectors like Moderate Resolution Imaging Spectroradiometer (MODIS) and Multiangle Imaging Spectro-Radiometer (MISR). This work will be carried by close collaboration with other groups supported by GACP.

FORM D: GACP BIBLIOGRAPHY

Name: Catherine C. Chuang

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

BIBLIOGRAPHY:

a. List of publications (including books, book chapters, and refereed papers), using AMS bibliographic citation form.

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.*, in press.

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.

Chin, H.-N. S., D. J. Rodriquez, R. T. Cederwall, C. C. Chuang, J. J. Yio, Q. Fu, and M. A. Miller, 1999: A study on the retrieval of continental low-level stratiform clouds: Impact of sub-adiabatic character on the macroscopic microphysical properties and radiation budgets. Accepted for publication in *Mon. Wea. Rev.*

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.

Chuang, C. C., K. E. Grant, and J. E. Dignon, 1999: A scenario study of modeling the past and future direct sulfate forcing. To be submitted to *Atmos. Environ.*

Chuang, C. C., J. E. Penner, and G. E. Grant, 1999: Radiative forcing by effect of anthropogenic aerosols on cloud susceptibility: A sensitivity study to aerosol characteristics and global concentration. To be submitted to *J. Geophys. Res.*

b. List of printed technical reports and 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)

Chuang, C. C., K. E. Grant, and J. E. Penner, 1999: Modeling black carbon effect on radiative properties of clouds in a global climate model. *Proceedings of the 9th ARM Science Team Meeting*, March 22-26, 1999, San Antonio, Texas, in press.

c. List of oral presentations or posters at professional society meetings and conferences.

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., J. E. Penner, and K. E. Grant, 1999: Cloud/aerosol parameterizations: Application and improvement of GCMs - A joint effort between LLNL and Univ. Michigan, Presented at the 9th ARM Science Team Meeting, March 22-26, 1999, San Antonio, Texas.

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., J. E. Penner, and K. E. Grant, 1999: Effect of absorbing aerosols on indirect aerosol forcing. Presented at the Symposium on Atmospheric Chemistry - Climate Interaction, 22nd General Assembly of IUGG, July 18-30, 1999, Birmingham, United Kingdom.

Chuang, C. C., 1999: A new era of research in aerosol/cloud/climate interactions at LLNL. Lawrence Livermore National Laboratory LDRD ER Presentations to Oversight Committee, July 23, 1999, Livermore, California.