GACP 2nd YEAR PROGRESS REPORT

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

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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) Comparisons to in-situ measurements

We used a fully coupled climate/chemistry model together with emissions inventories to model the global aerosol concentrations [*Chuang et al.,* 2000]. The simulated surface aerosol concentrations were compared to measurements at a number of remote ocean sites to validate the adequacy of the model. Figure 1 presents the comparisons of model-predicted seasonal surface concentrations of non-seasalt sulfate, dust, and sea salt to measurements at American Samoa and Barbados. Most of the simulated concentrations are within one deviation of the mean of the measurements, but discrepancies do exist. For example, the predicted seasonality of non-seasalt sulfate at American Samoa and Barbados is reasonable but the simulated concentrations are always lower than the mean of the measurements. The model also consistently overestimates the sea salt concentrations at American Samoa from May to December. In addition, the model shows a significant seasonal variation at Barbados whereas the measured data do not show much seasonality.

Figure 2 shows the observed and simulated surface concentrations for both organic carbon and black carbon at a number of locations. 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. In addition, there are analytical difficulties in making valid measurements of the various carbon species. Despite these facts, most simulated values are within a factor of 2 of the measurements.

(2) Impact of anthropogenic aerosols on cloud susceptibility

A modified cloud drop parameterization from *Chuang et al*. [1997] was applied into our coupled model to better represent the number concentration of cloud drops nucleated on aerosols with different size distributions. We found that the increases of cloud drop number concentrations can be up to 200 cm^{-3} due to anthropogenic carbonaceous aerosols and up to 30 – 90 cm^{-3} due to change of aerosol size distribution associated with the deposition of anthropogenic sulfate on pre-existing particles. The presence of anthropogenic aerosols would reduce the cloud susceptibility by a value up to 40×10^{-3} cm^3 (see Figure 3). The presence of industrial aerosols has significantly reduced cloud susceptibility in the northern hemisphere in January, and biomass aerosols have reduced susceptibility in the southern hemisphere in July. Our calculated monthly average cloud

susceptibilities are consistent to those retrieved from the AVHRR (Advanced Very High Resolution Radiometer) measurements in a single cloud event. The retrieved magnitude of susceptibility is about 0.23×10^{-4} cm^3 in stratus off the west coast of southern Africa to about 20×10^{-3} cm^3 in thin stratus off the California coast [*Platnick and Twomey,* 1994].

(3) Aerosol indirect forcing through the Twomey effect

The pattern of aerosol indirect forcing is determined by the magnitude of cloud susceptibility, cloud frequency as well as the incoming solar radiation. We found that biomass aerosols acting as CCN would cool the climate system by about –1.16 W m^{-2}, while carbonaceous aerosols from fossil fuel combustion acting alone may cool the system by about –0.52 W m^{-2} (see Table 1). However, because of the nonlinear nature of cloud drop nucleation, the independently computed indirect forcings by carbonaceous aerosols derived from different anthropogenic sources do not add linearly. The total indirect forcing by carbonaceous aerosols is estimated to be about –1.51 W m^{-2}. Variations in the natural organic aerosol emissions from 8.37 to 41.85 Tg yr^{-1} lead to variations in this global average forcing from -1.67 to -1.27 W m^{-2} (see Table 2). Applying a better representation of natural particle emissions together with the model-generated aqueous sulfate production rate, we refined our previous estimate of the indirect forcing by sulfate aerosols to be of the order –0.30 W m^{-2}. While the pattern of forcing from carbonaceous aerosols is approximately equal in each hemisphere (-1.64 vs. -1.38 W m^{-2} in the northern and southern hemispheres, respectively), that from anthropogenic sulfate remains primarily in the northern hemisphere. In addition, indirect sulfate forcing varied from –0.31 to –0.26 W m^{-2} when variations of up to a factor of 5 in the natural organic aerosol emissions were considered.

Sensitivity studies have been performed to quantify the uncertainty attributed to various chemical and physical properties of aerosols. The absorption of radiation by black carbon associated with clouds and its impact on the indirect forcing has been examined. We found that if absorption by BC in clouds is not included, indirect forcing by carbonaceous aerosols may be overestimated by 15 – 25% in regions where BC emissions are pronounced. Our sensitivity studies also revealed that the magnitude of the simulated aerosol indirect forcing was highly sensitive to the existing level of natural aerosol abundance. Without accounting for existing dust and sea salt particles, the indirect forcing by anthropogenic aerosols could be overestimated by at least a factor of 2. Therefore, further understanding of the interactions between anthropogenic components and natural particles together with a more thorough investigation on aerosol/cloud interactions is needed in order to better quantify the indirect effects of aerosols on the radiation budget.

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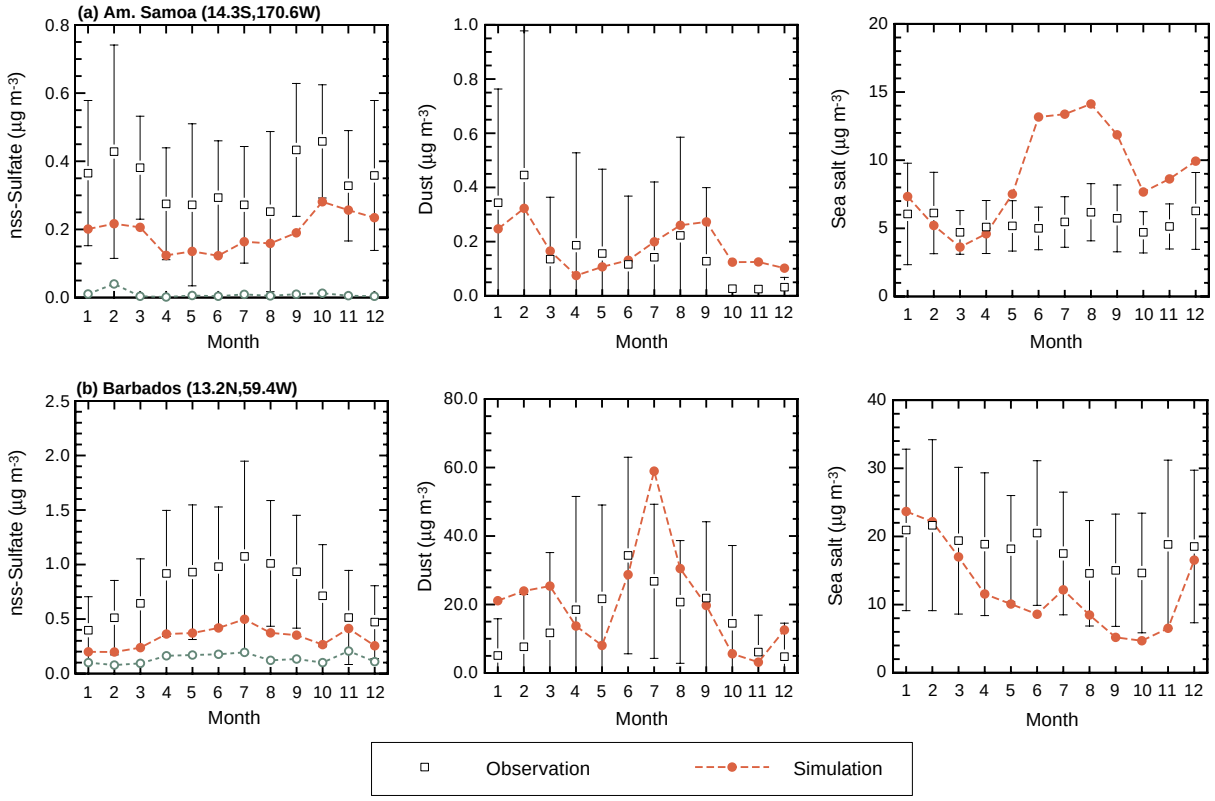


Figures 1. Modeled seasonal surface concentrations of nss-sulfate, dust, and sea salt versus measurements at American Samoa and Barbados. Error bars are one standard deviation above and below the mean of the measurements as compiled by Savoie and *Prospero* [private communication, 2000]. Also shown are the simulated concentrations of anthropogenic sulfate (open circles).

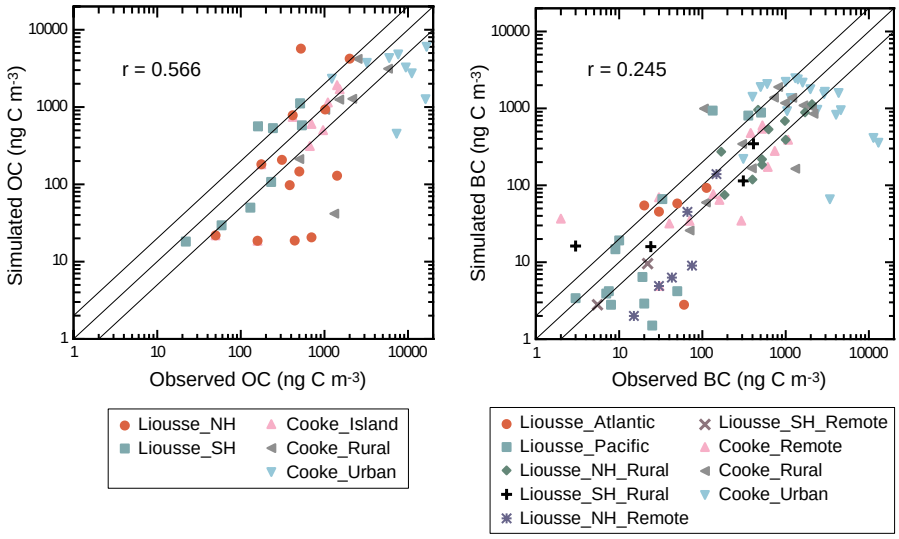
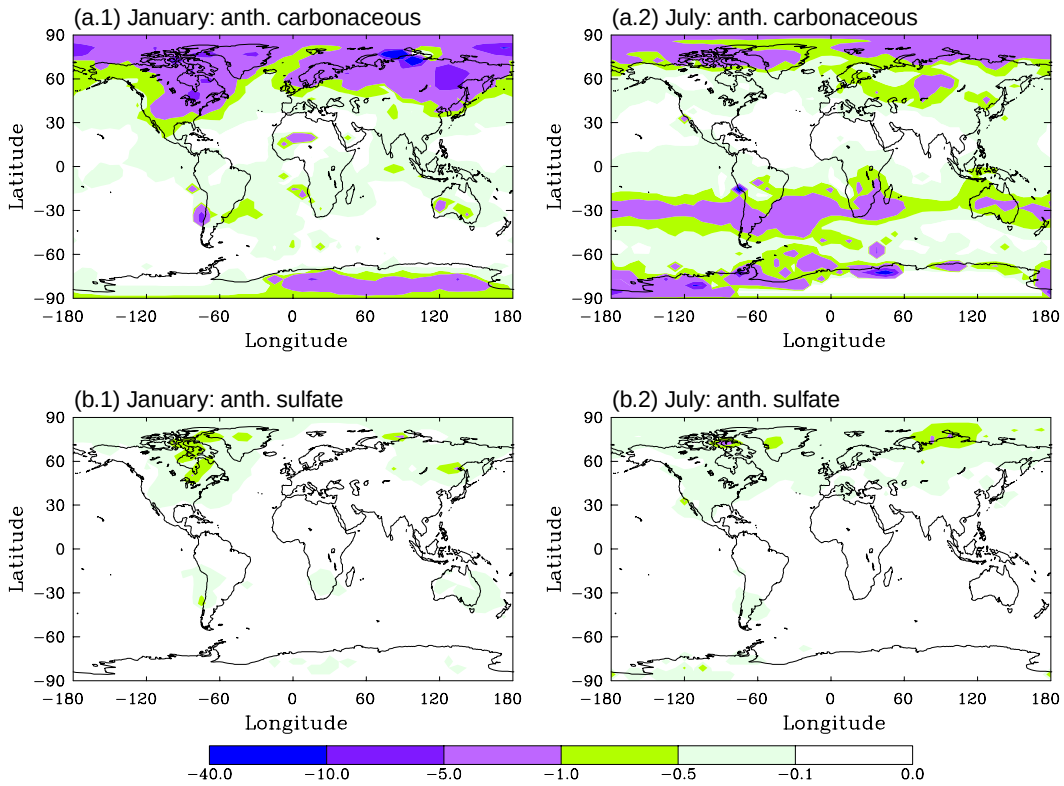


Figure 2. Observed and simulated surface concentrations of organic carbon and black carbon at a number of locations. Observations refer to those summarized by *Liousse et al.* [1996] and *Cooke et al.* [1999]. Correlation between the measured and modeled values is presented by coefficient r. Two thin lines represent the range above and below the observed values by a factor of 2.



Figures 3. Decreases of cloud susceptibility ($\times 10^{-3}$ cm^3) by anthropogenic carbonaceous aerosols (a.1 and a.2) and by anthropogenic sulfate (b.1 and b.2).

Table 1. Annual Average Indirect Aerosol Radiative Forcing (W m^{-2})

	Carbonaceous (nOM = 16.74 Tg yr^{-1})			Sulfate	Combined
	Fossil Fuel	Biomass	Total		
Global	-0.52	-1.16	-1.51	-0.30	-1.85
NH	-0.92	-0.98	-1.64	-0.45	-2.12
Land	-1.02	-0.69	-1.47	-0.43	-1.85
Ocean	-0.85	-1.17	-1.75	-0.46	-2.29
SH	-0.11	-1.33	-1.38	-0.15	-1.57
Land	-0.10	-1.32	-1.35	-0.20	-1.56
Ocean	-0.12	-1.33	-1.39	-0.14	-1.57

Table 2 Sensitivity of the Calculated Indirect Forcing (W m^{-2}) to Global Aerosol Concentrations

	nOM				nOM = 16.6 Tg yr^{-1}					
	8.37 Tg yr^{-1}		41.85 Tg yr^{-1}		No Dust		No Sea Salt		No Dust&Sea Salt	
	SO	OM/BC	SO	OM/BC	SO	OM/BC	SO	OM/BC	SO	OM/BC
Global	-0.31	-1.67	-0.26	-1.27	-0.39	-2.82	-0.36	-2.07	-0.51	-4.06
NH	-0.46	-1.82	-0.39	-1.38	-0.61	-3.68	-0.50	-1.90	-0.76	-4.76
Land	-0.47	-1.65	-0.36	-1.14	-0.60	-3.48	-0.46	-1.48	-0.66	-3.83
Ocean	-0.46	-1.93	-0.41	-1.53	-0.61	-3.80	-0.53	-2.17	-0.83	-5.35
SH	-0.16	-1.53	-0.13	-1.17	-0.16	-1.96	-0.23	-2.24	-0.26	-3.37
Land	-0.21	-1.62	-0.15	-0.99	-0.21	-2.07	-0.24	-1.54	-0.27	-2.56
Ocean	-0.14	-1.50	-0.13	-1.21	-0.15	-1.93	-0.23	-2.40	-0.26	-3.55

GACP 3rd YEAR STATEMENT OF WORK

Assessments of the radiative impact by atmospheric aerosols rely on a precise description of aerosol optical properties and an accurate representation of temporal and spatial variations of aerosol distributions. Parts of our earlier studies have been aimed at developing an understanding of global aerosol abundance as well as developing parameterizations for use in global climate models to treat the spectral optical properties of sulfate and carbonaceous aerosols. The first task for next year is to extend our treatment to dust and sea salt in order to compute their contribution to the total aerosol optical thickness in a global scale. We will compare the simulated total aerosol optical depths and extinction profiles to those retrieved from satellite measurements. We will validate whether our aerosol emissions inventories are correct and whether our treatments of transport and transformation are reasonable. We are interested in identifying the degree and conditions under which model and the observed aerosols demonstrate significant biases or departures in variability. In regions with significant aerosol extinction, we will characterize the scale of spatial covariance and the effects of such variability on radiative forcing.

In order to characterize the future climate variations, our second task is to perform multi-year simulations along with the IPCC newly developed anthropogenic emissions scenarios to estimate the present and future projections of aerosol forcing up to 2100. We will not only calculate the climate forcing by both direct and indirect effects of aerosols but also examine the climate feedback associated with the presence of aerosols. In addition, we will investigate the net radiative fluxes by the changes of natural emissions associated with climate change. This work will provide us a more quantitative range for aerosol climate effects as compared to those from greenhouse gases.

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