

Aerosol Simulations by LLNL IMPACT and Comparisons with Field Measurements

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Introduction

The first step needed to assess aerosol climate effects is to provide the global concentrations and vertical profiles of different aerosol components. These distributions cannot be easily obtained from satellite retrievals or field measurements but are key to estimating the magnitude of aerosol forcing. A new version of Lawrence Livermore National Laboratory (LLNL) Integrated Massively Parallel Atmospheric Chemical Transport (IMPACT) model with a shortwave radiation module was specially designed to develop the aerosol climatology and to explore the impact of aerosols on climate.

Model

IMPACT, developed at the LLNL, is a three-dimensional (3D) global Eulerian chemistry/aerosol model with a fully interactive chemical mechanism (~100 prognostic species) (Penner et al. 1998; Liu and Penner 2002; Rotman et al. 2001). This model includes a comprehensive representation of chemical and physical processes in both the troposphere and stratosphere. IMPACT accounts for changes in aerosol and chemical species from advective and convective transport, vertical diffusion, dry deposition (species and surface-type dependent), wet scavenging (scavenging in subgrid wet convective updrafts, as well as rainout and washout in precipitating columns), gravitational settling, and photochemistry. A computationally efficient version of IMPACT (version T3A) with a shortwave radiation module was specially designed for aerosol-related studies. This new version contains a compact chemical mechanism for the prediction of sulfate and also predicts the distributions of organic and black carbon, dust, and sea salt. By applying the monthly averages of O₃, OH, HO₂, and NO₃ from IMPACT simulations with interactive chemistry, sulfate aerosols are formed through both gas-phase (with OH) and aqueous-phase (with H₂O₂ and O₃) oxidation from SO₂ and DMS (dimethylsulfide). A correction factor was applied to the monthly average of HO₂ to account for the non-linear effect of diurnal variation of HO₂ on the production of H₂O₂ (HO₂ + HO₂ → H₂O₂ + O₂) and the predicted sulfate concentration. A look-up table for this correction factor was derived from the LLNL two-dimensional (2D) box model (Kinnison and Connell 1996) as a function of latitude, altitude, and time. Other aerosol types are either emitted as particles by nature or formed quickly from their gas phase precursors and hence are injected into the global model in the particulate form. IMPACT T3A is faster than the interactive chemistry version by a factor of 10. We have analyzed the simulated sulfur cycle to ensure that this new version is not only efficient but also in good agreement with the interactive chemistry version.

The spatial resolution in IMPACT is dictated by the input meteorological fields obtained from either a general circulation model (GCM) or assimilated data, such as that from the Data Assimilation Office (DAO) at National Aeronautics and Space Administration (NASA) Goddard. Using meteorology from a GCM allows us to examine the historical and future climate effects of aerosol/cloud interactions. Using assimilated data allows us to examine predictions associated with a particular time period and meteorological situation. The DAO assimilated data that we have used cover the period November 1996 through May 1998 with 6-hour averaged fields at 2 latitude \times 2.5 longitude horizontal resolution and 46 levels in the vertical. IMPACT simulations using this meteorology have provided direct comparisons of trace species to observations from the 1997 NASA SONEX (SSAS Ozone and Nitrogen Oxide Experiment) project (Chuang et al. 2002) as well as simulations of the Pinatubo eruption (Liu and Penner 2002).

Results shown in Figure 1 are the annually averaged concentrations of different aerosol components at the surface using the meteorology from the National Center for Atmospheric Research MACCM3 and emissions data for 1980, while Figure 2 shows the annual column burden of individual aerosol components from different sources. The majority of anthropogenic SO₂ is from fossil fuel and industry in the Northern Hemisphere with a maximum over Europe followed by the eastern United States and China. Contrary to the anthropogenic sources, the natural sources are strongly season dependent and distributed roughly evenly between the two hemispheres. Carbonaceous aerosols, composed of organic matter and black carbon (BC), are mainly emitted from biomass burning and fossil fuel combustion. As shown in Figure 2, biomass burning sources are dominant in south America, Africa, and southeast Asia while fossil fuel sources are mainly located in the northern hemisphere. Our model predicts a low BC concentration, 0.1 - 0.5 $\mu\text{g m}^{-3}$, (see Figure 1[c]) over the Indian subcontinent, while BC concentrations measured by aircraft during the intensive field phase (February - March 1999) of the INDOEX range from 0.6 to 6.3 $\mu\text{g m}^{-3}$ (Mayol-Bracero et al. 2000). This discrepancy is caused by the low BC emissions over the Indian subcontinent in our 1980 emissions database. Figure 1(d) presents the simulated surface concentrations of sea salt with the maximum located in a region between 40°S-60°S. It is noted that the total source strength of sea salt is more than twice as large in the southern hemisphere compared to the northern hemisphere. Figure 1(e) shows the simulated global distribution of surface dust concentrations. It is not surprising that the maximum dust concentration is located in the Sahara desert region in Africa. Simulations show that these African dust particles as measured are transported over the North Atlantic and Caribbean (Prospero 1999).

Comparison to Surface Measurements

We compared model results from IMPACT T3A to surface measurements at a number of remote ocean sites (Prospero, private communication, 2000) to validate the accuracy of the new version (Figure 3). Most measuring sites are located at coastal sites on the climatological windward shore of islands or continental coastlines. The aerosol data are presented at monthly means of at least several years of data and in some cases almost 20 years of data.

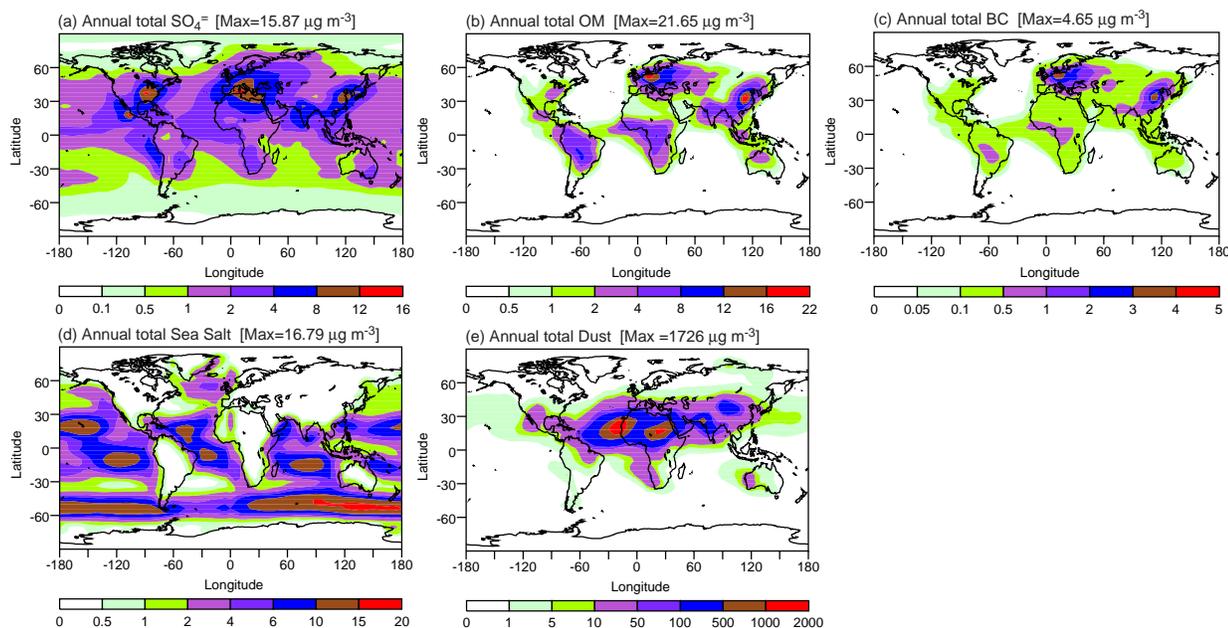


Figure 1. Predicted surface concentrations for (a) sulfate, (b) organic matter, (c) black carbon, (d) sea salt, and (e) dust using IMPACT T3A with meteorological fields from MACCM3.

The model reproduces the major features of the observed sulfate aerosols reasonably well, although concentrations are overestimated at some locations. Simulated concentrations of sea salt in general are lower than the observations. This discrepancy is attributed to the over estimation of gravitational settling velocity for large sea salt particles. Simulated dust concentrations are very low at high latitudes in the southern hemisphere. This low concentration may be caused by the vertical diffusion and convection that are not efficient enough to bring the large dust particles to higher levels for long-range transport. Finally, the model overestimates winter dust concentrations at the northern Pacific locations. This behavior is associated with the Asian dust source strength that is a factor of 10 larger in winter than in spring.

Conclusions

To better represent the surface measured data, we are examining the treatments of diffusion and removal processes in the model. We have compared the simulated surface concentrations of aerosols with our previous results from GRANTOUR (a 3D Lagrangian global chemistry/aerosol model). Consistent agreement was found between these two models, though magnitudes of aerosol concentrations may differ. We are in the process of calculating the aerosol optical thickness of individual as well as total aerosols to compare to the Advanced Very High Resolution Radiometer retrieval data. This is a further validation of the accuracy of the model. A shortwave radiation module has also been implemented into IMPACT. Calculations of aerosol forcing (direct and the first indirect) will be performed in the near future.

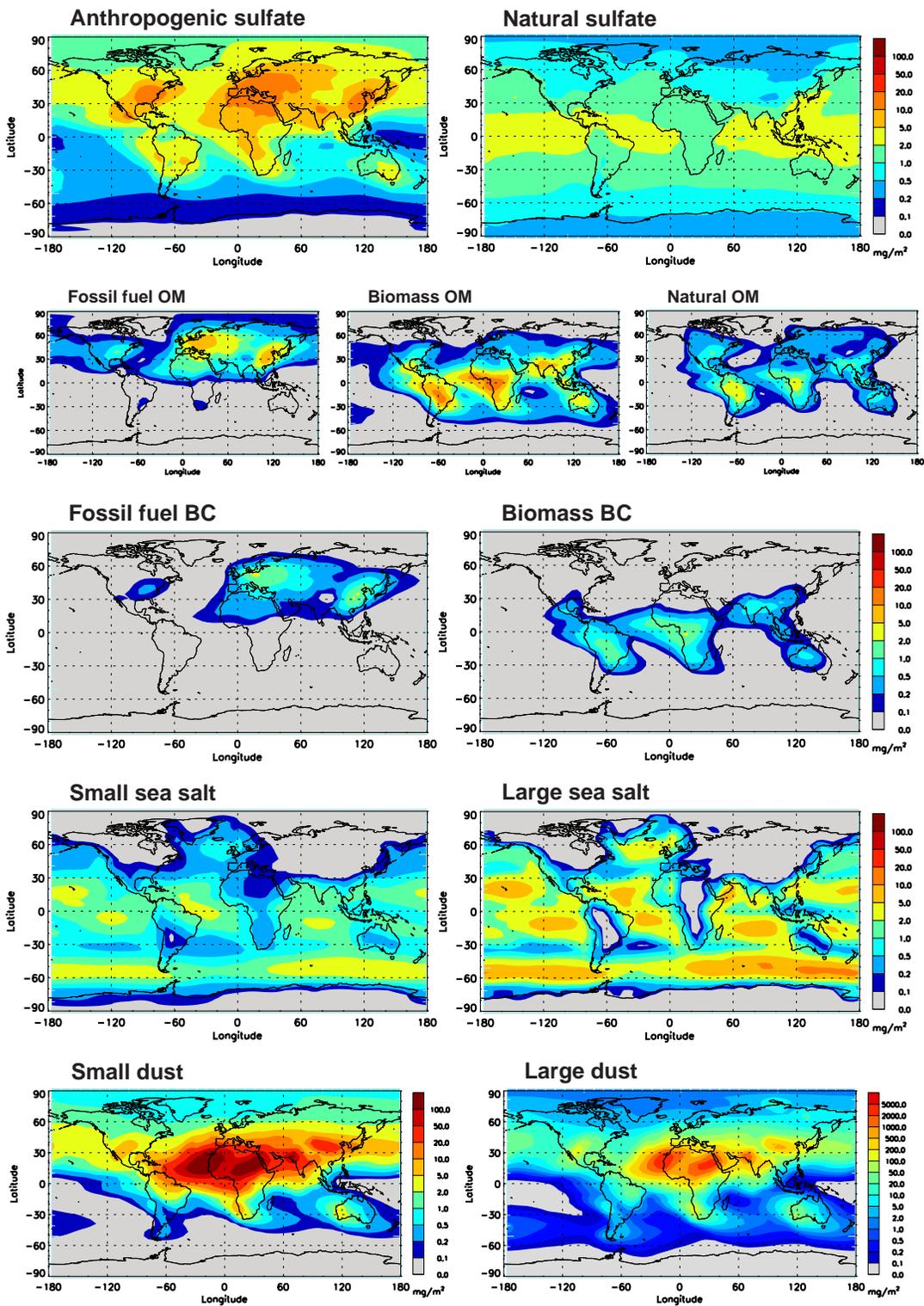
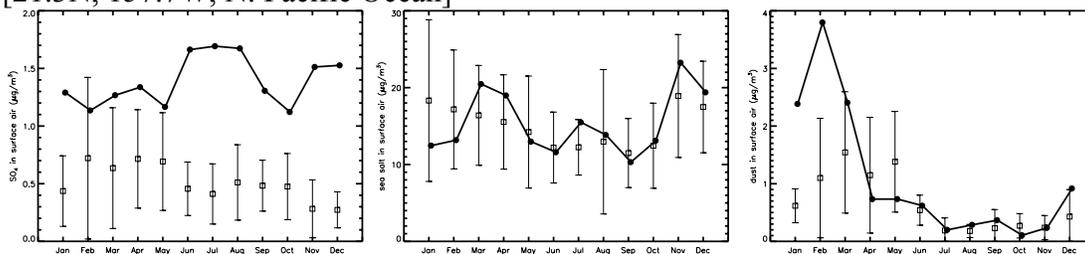
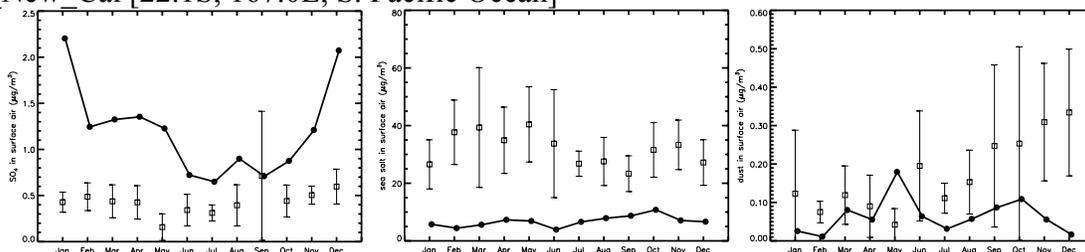


Figure 2. Predicted annual column burden (mg/m^2) of each aerosol component from difference sources.

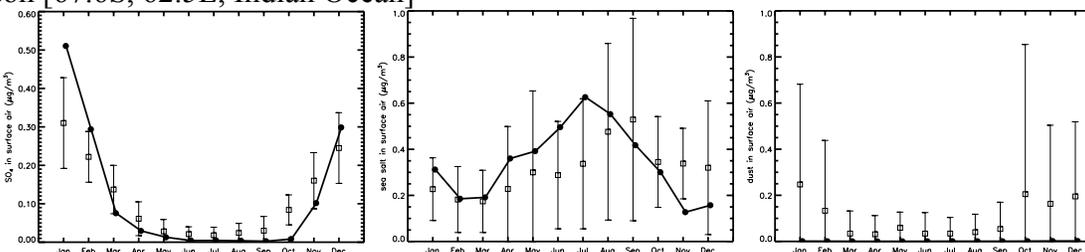
Oahu [21.3N, 157.7W, N. Pacific Ocean]



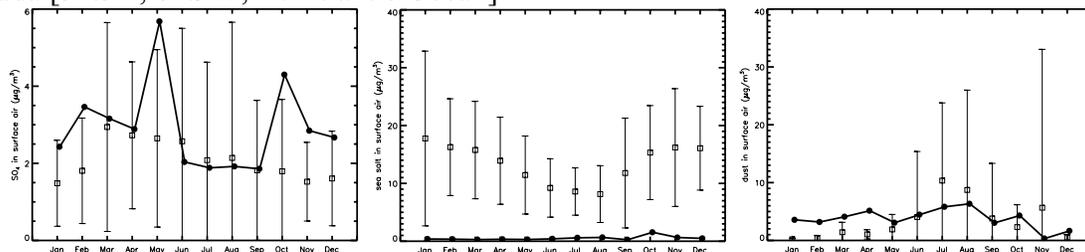
Yate New Cal [22.1S, 167.0E, S. Pacific Ocean]



Mawson [67.6S, 62.5E, Indian Ocean]



Bermuda [32.3N, 64.9W, N. Atlantic Ocean]



Palmer Station [64.8S, 64.0W, S. Atlantic Ocean]

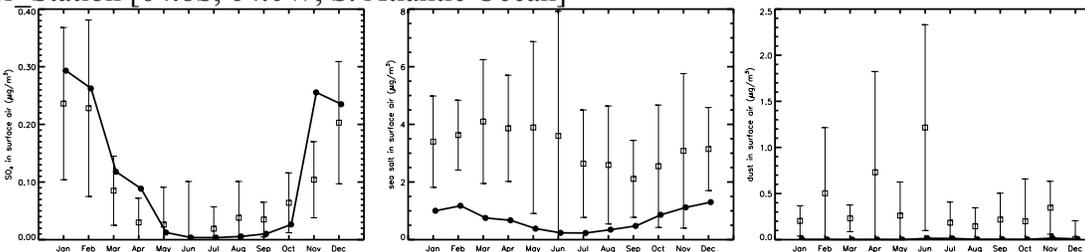


Figure 3. Modeled seasonal surface concentrations of nss-sulfate, sea salt, and dust versus measurements at a series of Pacific and Atlantic locations. Error bars are one standard deviation above and below the mean of the measurements as compiled by Savoie and Prospero (private communication, 2000).

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