Regional Modeling of Particulate Chemistry and its Effects on Cloud-Aerosol Interactions over the Southeastern Pacific Ocean

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1. Overview

The persistent stratocumulus cloud deck off the Chilean and Peruvian coasts in combination with abundant biogenic and anthropogenic sources of aerosol precursors make the southeastern Pacific Ocean a natural laboratory for studying cloud-aerosol interactions. Both cloud and aerosol properties vary significantly across the region, but the interactions between them are poorly understood. The effects of natural (e.g., volcanic) and anthropogenic (e.g. copper smelters) land sources on aerosol composition in the marine boundary layer are uncertain because of the altitude of these sources and complex circulation patterns. Pockets of open cells (POCs) that frequently form in the stratocumulus cloud deck are thought to form primarily in pristine maritime air masses, where low CCN concentration promotes rapid formation of drizzle, which further reduces CCN concentration. Yet, POCs also form in polluted air masses transported from the coast. Understanding regional transport and transformation of particulate and gaseous matter is therefore important for defining anthropogenic influence on cloud properties and radiation balance in the region.

VOCALS (VAMOS Ocean-Cloud-Atmosphere-Land Study) is an international program that coordinates scientific activities that will improve our understanding of the coupled ocean-atmosphere-land system over the southeastern Pacific Ocean. As part of VOCALS, a field campaign to be conducted in November 2008 will obtain a combination of in-situ and remotely sensed aerosol and cloud measurements. Aircraft and ship missions will focus on understanding the processes that control precipitation, including the role of atmospheric aerosols, their transport from the land to the ocean, and their depletion by clouds.

The primary scientific question we plan to address is: What are the effects of aerosol chemistry on the evolution of CCN and stratocumulus clouds downwind of large anthropogenic point sources along the Pacific coast of Chile?

In preparation for the VOCALS field campaign, we have run WRF for October 2006 to examine how well it simulates marine stratus, identify the horizontal transport pathways and vertical mixing of natural and anthropogenic aerosol precursors, and quantify the effects of simulated aerosol chemistry on the evolution of CCN and stratocumulus clouds. Our model simulations were also provided to the Pre-VOCALS model assessment that is critically assessing the ability of several regional and global models to simulate synoptically-varying clouds, meteorology, ocean circulation, and aerosols over the southeastern Pacific Ocean [*Wood*, 2008].

2. Cloud-Aerosol Interactions in WRF

Trace-gas chemistry and aerosols are solved "online" with the meteorology in WRF (Grell et al. 2005), enabling feedbacks among aerosols, radiation, cloud, and chemistry to be simulated. Details of the specific feedback processes we have implemented in WRF using the CBM-Z photochemical mechanism and the MOSAIC aerosol model [*Zaveri et al.,* 2008] are described in *Fast et al.* [2006], *Gustafson et al.* [2007], and *Chapman et al.* [2008]. The cloud-aerosol interactions are currently only coupled to the Lin et al. microphysics scheme in WRF. For this study, we employ versions of CBM-Z and MOSAIC in version 2.2 of WRF that also treat trace gas and aerosol processes associated with oceanic dimethylsulfide (DMS) emissions.

3. Model Configuration and Emissions

Two modeling domains were employed: an outer domain 5220 km wide (Δx = 45 km) that encompasses the southeastern Pacific Ocean and portions of Chile, Peru, Argentina, Ecuador, and Boliva and an inner domain that is 2340 km wide (Δx = 15 km) and encompasses the 2008 field campaign sampling area. 44 vertical levels were employed. 29 vertical levels were placed within 2 km of sea-level to resolve the marine boundary layer and shallow stratocumulus clouds.

Global 1 x 1 degree emission inventories from the Emission Database for Global Atmospheric Research, version 3.2 (EDGAR3.2) [*Olivier and Berdowski*, 2001], the Precursors of Ozone and their Effects in the Troposphere (POET) project, and the Aerosol Intercomparison (AeroCom) project [*Dentener et al.*, 2006] formed the basis of the hourly emission input files. Because global inventories do not include diurnal factors (and for many species are annual estimates only), emissions of a given species were assumed to be the same for each hour of each day. SO_2 emission values were supplemented with the more detailed estimates for five power plants, one copper smelter, and one volcano provided by Dr. Laura Klenner (Universidad de Chile). Biomass burning estimates were neglected. Other emissions were computed "on-line" including:

- Dimethylsulfide (DMS) emissions were generated on-line using a global inventory of oceanic DMS concentrations [*Kettle and Andreae*, 2000], regridded to the WRF-chem domains, and model wind speeds and temperatures via the air-sea mass transfer parameterization of *Nightingale et al.* [2000]
- Sea-salt emission rate parameterized as a function of model wind speed [*Gong et al.*, 1997]
- Dust emissions, computed via the parameterizations of *Shaw et al.* [2008] that depend on simulated friction velocity, vegetation cover, soil type, and soil moisture

Three additional passive scalars were added to the WRF-chem simulations to define transport pathways of various sources: 1) SO_2 from smelter and power plants, 2) SO_2 from volcanoes, and 3) SO_2 emitted from anthropogenic surface sources.

The simulation period was 00 UTC October 1 – 00 UTC November 1, 2006. The initial and boundary conditions for the meteorological parameters were based on GFS analyses at 1° grid spacing and at 12-h time intervals. Initial and boundary conditions for trace gas and particulate species were based on the global MOZART model [*Horowitz et al.,* 2003] output at T42 resolution (~240 km) and at 6-h time intervals provided by Louisa Emmons (NCAR).

Three simulations were performed:

1) A "no aerosol" simulation that is equivalent to the standard version of WRF
2) A "prescribed aerosol" simulation in which a uniform CCN is specified
3) An "interactive aerosol" simulation that employs trace gas and aerosol chemistry and fully interactive aerosol-cloud feedbacks

4. Results

An example of the predicted cloud distribution over the inner nested domain (Δx = 15 km) at 12 UTC October 15 from simulation 3 is shown in Fig. 1. Most of the cloud optical depth can be attributed to shallow stratocumulus clouds at the top of the marine boundary layer, except for the convective clouds over the Andes Mountains in Peru. At this time, a stratus deck is present along the northern coast of Chile. While the sky is clear to the south, most of the ocean is covered with stratocumulus with varying thickness. There are also pockets of clear skies in the off-shore stratocumulus deck; however, the 15-km grid spacing cannot fully-resolve all the spatial variations observed by satellite.

The predicted cloud fields have been evaluated using data derived from the TRMM satellite. One metric was the number of cloudy grid cells over the entire outer domain. Satellite measurements were apportioned to the WRF grid cells and only those cells with valid data were compared. The results from simulation 1 with no cloud-aerosol interactions are shown in the top panel of Fig. 2.

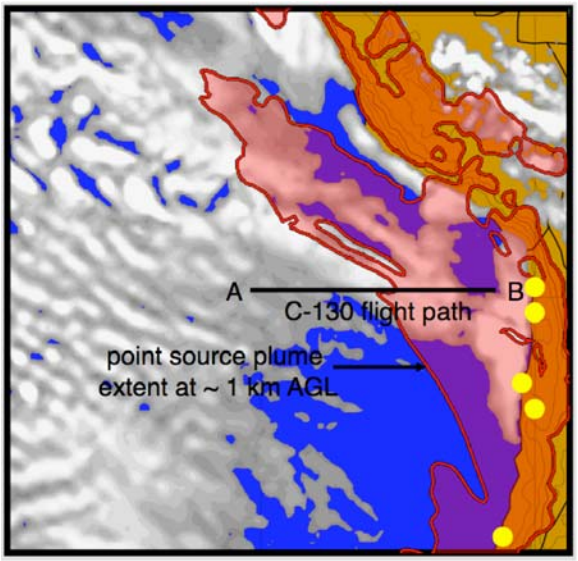

Fig. 1. Cloud optical depth (gray shading) and extent of SO_2 tracer plume from point sources at ~ 1 km AGL (red shading) at 12 UTC October 15, 2006 over the inner nested domain.

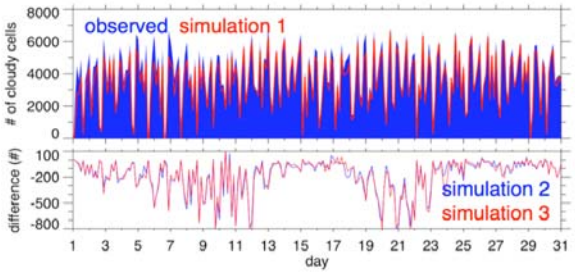

Fig. 2. Observed and simulated (meteorology only) number of cloudy cells over the outer model domain (top) and difference between simulations with and without aerosol-cloud interactions (bottom).

The model was able to capture the diurnal variation in cloudiness over the domain, with highest values in the early morning. The simulated peak values are somewhat lower than observed, suggesting the model does not predict enough stratocumulus clouds over the domain.

Also shown in Fig. 1 is the extent of the SO_2 tracer plume from point sources at ~ 1 km AGL. The southerly and southeasterly flow transported SO_2 over the ocean and roughly parallel to the coast of Peru. The highest concentrations are 200-300 kilometers from the coast. Vertical cross sections indicate that SO_2, other trace gases, and particulates are entrained into portions of the stratocumulus desk (not shown). The SO_2 tracers suggest that the power plant and copper smelter sources are the largest contributors, with their concentrations 10 – 100 times higher than other anthropogenic sources in the region. While the volcanic sources are located at 2 – 4 km above sea-level, the model suggests that a portion of these plumes are entrained into the coastal air mass and transported over the ocean.

The "A – B" line in Fig.1 denotes one proposed NCAR C-130 aircraft path that includes transects below, within, and above the stratocumulus deck. The figure illustrates that the aircraft will sample "dirty" air close to the coast and more "pristine" air masses away from the coast. DOE's G-1 aircraft, with its shorter flight duration, will sample mostly within a few hundred km of the coast.

The presence of aerosols and aerosol precursors in the marine stratus (Fig. 1) suggest that cloud-aerosol interactions may be important. The impact of cloud-aerosol interactions on cloudiness over the outer domain is shown in the bottom panel of Fig. 2. Both simulations 2 and 3 show reductions in the number of grid cells with clouds, with the greatest reductions during two periods: 9-12 October and 19-23 October. Interestingly, the results for the simulation with a simple prescribed value of CCN and the full-chemistry simulation were similar. It is important to note that these are aggregate results over the entire domain and large differences between simulations 2 and 3 were found for specific locations (e.g. along the coast) and times (not shown).

Our presentation will include additional analysis of the three simulations as well as some comparisons of WRF with other model predictions.

5. Future Work

Since the cloud-aerosol interactions are coupled only with the Lin et al. microphysics scheme, we will be coupling the cloud-aerosol interaction modules to other microphysics schemes (Thompson and Morrison schemes) that may better simulate characteristics of marine stratocumulus clouds. Since quantifying aerosol precursor sources is critical in understanding the importance of cloud-aerosol interactions, we are currently working with Chilean scientists to develop higher resolution anthropogenic emissions and better characterize point sources in the region.

After the field campaign, we will perform retrospective simulations for the entire VOCALS sampling period to evaluate how well the modules implemented in WRF predict the impact of clouds on aerosol properties and the impact of aerosols on cloud properties (spatial extent, optical depth, droplet number, precipitation). The extensive measurements made by aircraft, ship, surface, and satellite measurements will be used in our evaluation. We anticipate that the retrospective simulations will employ smaller grid spacings to better resolve the observed variations in stratocumulus.

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References

Chapman, E.G., W.I. Gustafson Jr., R.C. Easter, J.C. Barnard, S.J. Ghan, M.S. Pekour, and J.D. Fast, 2008: Coupling aerosols-cloud-radiative processes

in the WRF-chem model: Investigating the radiative impact of large point sources. Submitted to *Atmos. Chem. Phys.*

Dentener, F., S. Kinne, T. Bond. O. Boucher et al., 2006: Emissions of Primary Aerosol and Precursor Gases in the Years 2000 and 1750 Prescribed Data-sets for AeroCom. *Atmos. Chem. Phys.*, 6, 4321-4344.

Gong, S.L., L.A. Barrie, and J.-P. Blanchet, 1997: Modeling sea-salt aerosols in the atmosphere: 1. Model development. *J. Geophys. Res.*, 102, 3805-3818.

Fast, J.D., W.I. Gustafson, R.C. Easter, R.A. Zaveri, J.C. Barnard, E.G. Chapman, G.A. Grell, and S.E. Peckham, 2006: Evaluation of ozone, particulates, and aerosol direct radiative forcing in the vicinity of Houston using a fully-coupled meteorology-chemistry-aerosol model. *J. Geophys. Res.*, 111, doi:10.1029/2005 JD006721.

Grell, G.A., S.E. Peckham, R. Schmitz, S.A. McKeen, G. Frost, W.C. Skamarock, and B. Eder, 2005: Fully coupled "online" chemistry within the WRF model. *Atmos. Environ.*, 39, 6957-6975.

Gustafson Jr., W.I., E.G. Chapman, S.J. Ghan, and J.D. Fast, 2007: Impact on modeled cloud characteristics due to simplified treatment of uniform cloud condensation nuclei during NEAQS 2004. *Geophys. Res. Lett.,* 34, L19809.

Kettle, A.J. and M.O. Andreae, 2000: Flux of dimethylsulfide from the oceans: A comparison of data sets and flux models. *J. Geophys. Res.*, 105, 26793-26808.

Nightingale, P.D., G. Malin, C.S. Law, A.J. Watson, P.S. Liss, M.I. Liddicoat, J. Boutin, and R.C. Usptill-Goddard, 2000: In situ evaluation of air-sea gas exchange parameterizations using novel conservative and volatile tracers, *Global Biogeochem. Cycles*, 14, 373-387.

Olivier, J.G.J. and J.J.M. Berdowski, 2001: Global emissions sources and sinks. In: Berdowski, J., Guicherit, R. and B.J. Heij (eds.) "The Climate System", pp. 33-78. A.A. Balkema Publishers/Swets & Zeitlinger Publishers, Lisse, The Netherlands. ISBN 90 5809 255 0.

Shaw, W.J., K.J. Allwine, B.G. Fritz, F.C. Rutz, J.P. Rishel, and E.G. Chapman, 2008: An evaluation of the wind erosion module in DUSTRAN. *Atmos. Environ.*, 42, 1907-1921, doi:10.1016/j.atmosenv.2007.11.022.

Wood, R., 2008: VOCALS model assessment: http://www.atmos.washington.edu/~robwood/PreVOCA/index.htm

Zaveri R.A., R.C. Easter, J.D. Fast, and L.K. Peters, 2008: Model for simulating aerosol interactions and chemistry (MOSAIC). *J. Geophys. Res.*, doi:10.1029/2007JD008792.