

# 1 Atmospheric Composition

#### **Strategic Research Questions**

- 3.1 What are the climate-relevant chemical, microphysical, and optical properties, and spatial and temporal distributions, of human-caused and naturally occurring aerosols?
- 3.2 What are the atmospheric sources and sinks of the greenhouse gases other than CO<sub>2</sub> and the implications for the Earth's energy balance?
- 3.3 What are the effects of regional pollution on the global atmosphere and the effects of global climate and chemical change on regional air quality and atmospheric chemical inputs to ecosystems?
- 3.4 What are the characteristics of the recovery of the stratospheric ozone layer in response to declining abundances of ozone-depleting gases and increasing abundances of greenhouse gases?
- 3.5 What are the couplings and feedback mechanisms among climate change, air pollution, and ozone layer depletion, and their relationship to the health of humans and ecosystems?

See Chapter 3 of the *Strategic Plan for the U.S. Climate Change Science Program* for detailed discussion of these research questions.

The composition of the atmosphere on global and regional scales influences climate, air quality, the stratospheric ozone layer, and weather. The interactions have impacts on human health and the vitality of ecosystems and hence have high relevance to society. CCSP research on atmospheric composition focuses primarily on how human activities and natural processes affect atmospheric composition, and how these changes in turn relate to societal issues. The issues embrace multiple disciplines, cross many spatial scales, and are highly interrelated. As a consequence, the CCSP research is a highly coordinated endeavor that involves observational studies, laboratory investigations, and diagnostic and modeling analyses to provide the timely, accurate, and useful scientific information needed by decisionmakers in the United States and abroad.

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Much progress has been made to date in understanding the role of atmospheric composition in Earth's climate. Efforts have focused on the areas of largest uncertainty in understanding how atmospheric constituents other than carbon dioxide affect the forcing of climate. Atmospheric fine particles ("aerosols") can have either warming or cooling effects, depending on many factors. CCSP research has made progress in defining those factors, and has recently taken steps to address the next levels of complexity in the issue by looking at the interactions of aerosols with clouds. For FY 2008, CCSP's Atmospheric Composition research will advance further to investigate how water vapor, an ever-present climate gas, affects climate. Because water vapor is linked to trace gases, aerosols, and clouds, CCSP research will be addressing some of the most highly interrelated, multidimensional aspects of climate forcing.

#### HIGHLIGHTS OF RECENT RESEARCH

The following paragraphs provide selected highlights of recent research supported by CCSP-participating agencies.

Protecting Earth's Ozone Layer Also Helped Slow Climate Change. 1 A 1987 international agreement to reduce ozone-depleting chemicals has also slowed global warming by years, according to a new study by CCSP scientists and their colleagues. The double effect occurred because compounds that destroy the atmosphere's ozone layer also act as greenhouse gases. The ozone layer shields the Earth from harmful ultraviolet radiation. To protect this layer, nations around the world signed the Montreal Protocol in 1987 to control the production and use of ozone-depleting substances. While protecting the ozone layer, the Montreal Protocol and its Amendments have also cut in half the amount of greenhouse warming caused by ozone-destroying chemicals that would have occurred by 2010 had these substances continued to build unabated in Earth's atmosphere. The amount of warming that was avoided is equivalent to 7 to 12 years of rise in carbon dioxide (CO<sub>2</sub>) concentrations in the atmosphere during the 2000 to 2010 time frame. Earlier studies showed that continued growth in ozonedepleting substances would lead to significant warming of Earth's climate. The new analysis quantifies the near-term climate benefits of controlling these substances (see Figure 1).





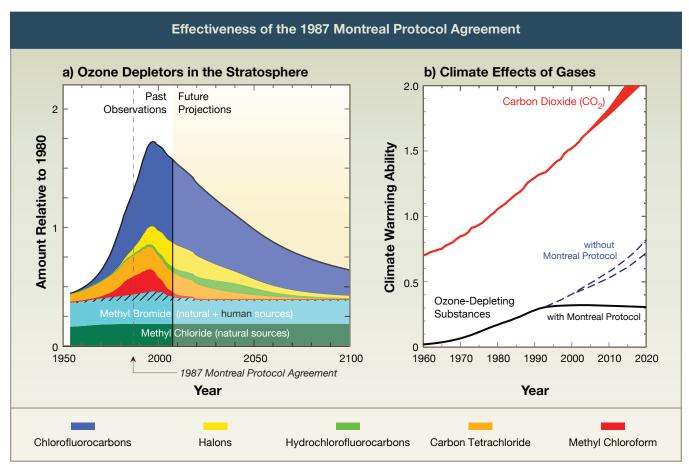


Figure 1: Effectiveness of the 1987 Montreal Protocol Agreement. Past, present, and projected future abundances of ozone-depleting substances in the stratosphere show the effectiveness of the 1987 Montreal Protocol agreement that reduced worldwide use of these substances (panel a). Because ozone-depleting substances are also greenhouse gases, the Montreal Protocol and its amendments gave an early start to slowing climate warming (panel b). The left axis (Climate Warming Ability) gives the climate forcing in units of watts per meter squared (Wm<sup>-2</sup>). Credit: G.J.M. Velders, Netherlands Environmental Assessment Agency; S.O. Andersen, USEPA; J.S. Daniel, NOAA; D.W. Fahey, NOAA; and M. McFarland, DuPont Fluoroproducts.

Satellite Studies of Water Vapor and Ozone-Depleting Gas Transport.<sup>2</sup> Analyses of Aura satellite observations of water vapor and carbon monoxide at high altitudes, and their comparison with model calculations, show that thunderstorms over Tibet provide a pathway for water vapor and chemicals to travel from the lower

atmosphere into the stratosphere. Since water vapor has a strong influence on climate, learning how it reaches the stratosphere can help improve climate prediction models. Similarly, understanding the pathways that ozone-depleting chemicals can take to reach the stratosphere is essential for understanding future threats to the ozone layer.

#### C H A N G I N G P L A N E T O U R

Improved Estimates of the Recovery of the Antarctic Ozone Hole.<sup>3</sup> CCSP research has improved our understanding of atmospheric motions that transport ozone-depleting substances from the lower atmosphere to the Antarctic stratosphere, showing that this transport is slower than previously estimated. As a result, the revised estimate for recovery of Antarctic ozone will occur about 15 years later than previously thought, in approximately 2065 instead of 2050. Such research during the ozone layer's recovery phase is crucial for policymakers.

Completion of International Assessment of the Ozone Layer. 4 CCSP researchers played key roles in the completion of the international state-of-understanding assessment of the ozone layer, which was provided in FY 2007 to the over 190 nations (including the United States) that are Parties to the United Nations Montreal Protocol on Substances that Deplete the Ozone Layer. The Scientific Assessment of Ozone Depletion: 2006 summarizes current understanding regarding the extent of ozone depletion globally and at the poles, describes the current abundances of ozone-depleting gases in the

atmosphere, assesses the future expectations for the recovery of the ozone layer and its SCIENTIFIC ASSESSMENT OF SCIENTIFIC ASSESSMENT OF Ozone Depletion: 2006 **OZONE DEPLETION: 2006** Twenty Questions and Answers out the Ozone Layer: 2006 Update **Executive Summary** (0) (1) **(1)** 

relation to possible future climate change, and updates the past, current, and expected future status of ultraviolet radiation at

the Earth's surface. CCSP researchers were prominent in the leadership, preparation, and review of the assessment—a 2-year endeavor that involved over 300 scientists from 31 countries around the globe. Global decisionmakers will consider the information in the over 500 pages of the report as they discuss possible future actions to protect the stratospheric ozone layer.

Field and Laboratory Investigations on Atmospheric Composition and Climate. 5,6,7 A combination of field experiments for the Mexico City area, the northeastern United States, and other regions, together with laboratory studies, have better defined aerosol formation processes, their properties, and their abundances. The studies have shown a higher than expected formation of organic aerosols within the atmosphere, which could potentially have a cooling effect. The research has also demonstrated the influence of aging and composition on aerosol properties, and the ubiquity of

absorbing (warming) aerosols and black carbon in the atmosphere. The information will enable more accurate calculation of aerosol influences on climate through their absorption and scattering of light; results that will ultimately lead to more accurate model estimates of the climatic role of aerosols.

Research Indicates Importance of Anthropogenic Secondary Organic Aerosol.<sup>5</sup> Organic aerosol particles produced within the atmosphere, called "secondary organic aerosol" (SOA), are important to climate because they interact with sunlight and affect the energy balance of Earth's atmosphere. About 90% of secondary organic aerosol is currently believed to arise from the oxidation of natural volatile organic compounds of biological origin. Volatile organic compounds produced by human activity have therefore not been included in most modeling studies that assess the relevance of SOA to climate forcing. However, a recent study examining aerosol production in Mexico City indicates the presence of SOA production pathways not currently accounted for, and suggests that the human-caused sources of SOA are much more important than had been thought. Findings from this study (see Figure 2) show that amounts of SOA produced for any reacted amount of anthropogenic volatile organic compounds are as

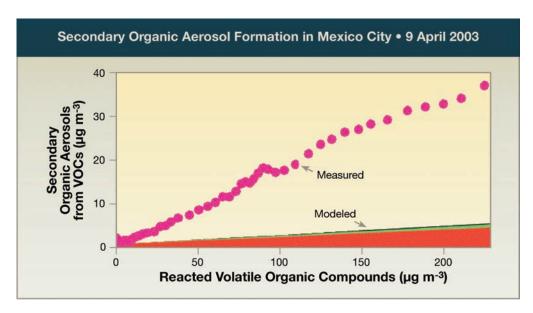


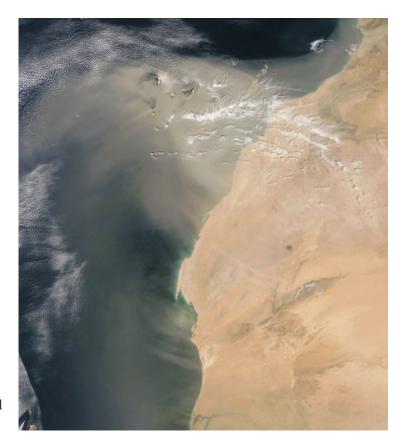
Figure 2: Secondary Organic Aerosol (SOA) Formation in Mexico City. Current state-of-the-art models under-predict the rapid formation of large amounts of SOA. Here, data are shown from Mexico City on 9 April 2003. Solid dots indicate observations and shaded areas indicate the predicted SOA-mass concentration attributed to classes of anthropogenic volatile organic compounds, such as aromatics (red), alkenes (green), and alkanes (black). Credit: R. Volkamer, University of California, San Diego; J.L. Jimenez, University of Colorado; F. San Martini, National Academy of Sciences; K. Dzepina, University of Colorado; Q. Zhang, SUNY Albany; D. Salcedo, Universidad Autónoma del Estado de Morelos, Cuernavaca, Mexico; L.T. Molina, University of California, San Diego; D.R. Worsnop, Aerodyne Research; and M.J. Molina, University of California, San Diego (redrawn from Geophysical Research Letters with permission from the American Geophysical Union).

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much as eight times greater than predicted by current models. The research will enable modelers to more accurately represent the atmospheric processes related to SOA, ultimately leading to improved climate projections.

First Analyses from the Gulf of Mexico Climate Study. <sup>8</sup> Several Federal agencies, with significant university, private, and non-profit sector participation, completed a major field mission, the Gulf of Mexico Atmospheric Composition and Climate Study, in early FY 2007 and started analyzing the wealth of data that resulted from the mission. This intensive field study took place in August through October 2006 in the Texas/northwestern Gulf of Mexico region and was focused on providing a better understanding of the sources and atmospheric processes responsible for the formation and distribution of ozone and aerosols in the atmosphere and the influence that these species have on the radiative forcing of climate regionally and globally, as well as their impact on air quality, human health, and regional haze. Rapid synthesis reports on the data were produced within the first few weeks after completion of the mission, an unprecedented turnaround time for communication of the results of a field mission. Early findings are helping to improve the simulation of the radiative forcing of climate change by lower atmosphere ozone and aerosols.

Models Evaluated Using Saharan Dust Storm Data.<sup>9</sup> A large Saharan dust storm raged across the North African desert in March 2006, the largest storm in the last several years. The event was captured by ground-based instruments of the Atmospheric Radiation Measurement (ARM) Mobile Facility (AMF) that were deployed in Niamey, Niger, during 2006, as well as by instruments flown onboard the Meteosat-8 geostationary satellite platform, and instruments onboard the Terra and Aqua polar-orbiting satellite platforms. The combination of AMF and satellite observations provides the first wellsampled direct evaluations of the effects of the dust storm on solar and thermal radiation across the atmosphere, allowing researchers to test their understanding of how dust affects the radiant energy budget of the atmospheric column. This information is a key component in computer models that simulate both regional and global weather and climate.



#### Highlights of Recent Research and Plans for FY 2008

Field and Modeling Studies of Aerosols and Clouds. 10,11,12 A significant but inadequately understood area of climate research involves the effects of aerosol on cloud formation, cloud properties, and cloud lifetimes. An analysis of results from an interagency field experiment, the Cloud Indirect Forcing Experiment, indicates that an increase in aerosol produces higher concentrations of small drops in certain types of maritime clouds. Aircraft and satellite observations of the changes to the maritime clouds show that these microphysical effects result in brighter clouds that have a cooling effect by reflecting more of the incident sunlight back to space. In other CCSP studies, modeling of the effect of carbonaceous (soot-like) aerosol showed a reduction in cloudiness with increasing aerosol amount, as a result of aerosol absorption modifying the heating of the surface and the atmosphere. Finally, another modeling study showed no evidence for aerosol increasing the lifetime of individual cumulus clouds as is usually hypothesized. CCSP researchers also completed a FY 2007 field campaign over the U.S. Southern Great Plains to study the interactions of atmospheric aerosols and fair weather cumulus clouds downstream of a midsize urban area (Oklahoma City), the Cumulus Humilis Aerosol Processing Study (CHAPS). Observations from this campaign will aid in development and evaluation of climate model parameterizations of cloud-aerosol processes. CHAPS involved coordination of experiments between the CCSP Atmospheric Composition and Global Water Cycle Interagency Working Groups.

Phytoplankton Emissions and Cloud Properties over the Southern Ocean. <sup>13</sup> Researchers have long thought that emissions from marine phytoplankton influence aerosols and clouds, but evidence for how this natural process occurs has been scarce. With satellite remote-sensing data and a cloud parcel model, CCSP researchers have shown that over a large area of the Southern Ocean, phytoplankton blooms are correlated in space and time with increases in cloud droplet concentrations and decreases in effective cloud droplet radius. The modeling study showed that the changes in cloud properties could be attributed to the formation of organic aerosol particles arising from the oxidation of a hydrocarbon (isoprene) that is emitted by phytoplankton. This effect seems to act in concert with sulfur emissions from phytoplankton, which have previously received much greater attention.



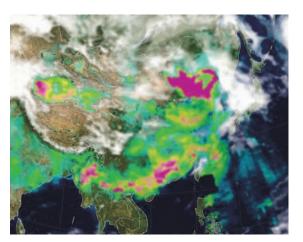
#### HIGHLIGHTS OF PLANS FOR FY 2008

CCSP will continue to gather and analyze information through measurement, modeling, and assessment studies to enhance understanding of atmospheric composition and of the processes affecting atmospheric chemistry. Key research plans for FY 2008 follow.

Tropical Composition, Clouds, and Climate Coupling. CCSP researchers will begin their analysis of data from a FY 2007 field mission in Costa Rica to study how climate is linked to atmospheric composition and clouds in the tropical summer convective wet season. This successful Tropical Composition Cloud and Climate Coupling field mission involved three major aircraft (DC-8, ER-2, WB-57F) making over 20 research flights, using some 60 instruments, involving balloon launches from three locations, and engaging over 250 participants. Analyses will incorporate data from aircraft flights and ground measurements, as well as Aura satellite observations, to address scientific questions related to how clouds, aerosols, and trace gases influence radiative heating in the very active tropical atmosphere.

These activities will address Questions 3.1 and 3.2 of the CCSP Strategic Plan.

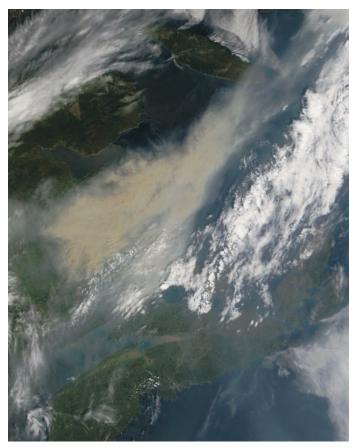
International Polar Year Research on Arctic Aerosols and their Connections to Clouds, Radiation, and Ice Melting. The long-range transport of anthropogenic pollution from North America, Europe, and western Asia creates the aerosols associated with the so-called Arctic haze, a phenomenon that recurs every winter and spring. The direct and indirect climate impacts of the aerosols can be quite different in the Arctic compared to elsewhere, because high surface reflections from snow and ice mean that even weakly absorbing aerosol layers can heat the Earth/atmosphere system in the Arctic. As part of International Polar Year (IPY) research, CCSP scientists will conduct field missions to investigate Arctic aerosol/climate connections in this unique environment. Spring and summer measurements from satellites, aircraft, and the surface will be



made in collaboration with the larger IPY study POLARCAT (Polar Study using Aircraft, Remote Sensing, Surface Measurements, and Models of Climate, Chemistry Aerosols, and Transport). Springtime observations will be made to assess the long-range transport of anthropogenic pollution to the Arctic and its contribution to Arctic Haze, ozone chemistry, and the possible connections between Arctic aerosols



### Highlights of Recent Research and Plans for FY 2008



and the melting of polar ice. Summertime observations will be made to assess fire emissions from the boreal forests. Analyses of these measurements will ultimately improve the ability of current models to simulate the influence of anthropogenic pollution and boreal fires on the Arctic atmosphere and climate. In 2008, new aerosol measurements made on the North Slope of Alaska will be used to evaluate model simulations of clouds and aerosol influences in the Arctic.

This activity will address Questions 3.1 and 3.3 of the CCSP Strategic Plan.

#### Completion of CCSP Synthesis and Assessment Product

2.3. CCSP researchers will finalize the second phase of CCSP Synthesis and Assessment Product 2.3, Aerosol Properties and their Impacts on Climate. The first phase of development of this product was to produce major scientific reviews on the following three topics: dependence of radiative forcing by tropospheric aerosols on aerosol composition in the north Atlantic, Pacific, and Indian Ocean regions; measurement-based understanding of aerosol radiative

forcing from remote-sensing observations; and model intercomparison to quantify uncertainties associated with indirect aerosol forcing. The second-phase product will draw upon the scientific information gathered by the development of the Intergovernmental Panel on Climate Change Fourth Assessment Report and the National Research Council review, *Radiative Forcing of Climate Change*. Authors will draw from these community-wide assessments of climate change (and the aerosol-climate topic inclusively) in writing this synthesis and assessment product.

This activity will address Questions 3.1 and 3.2 of the CCSP Strategic Plan.

VOCALS Cloud/Aerosols Field Study. Extensive and persistent layers of stratus clouds occur off the subtropical west coasts of Africa and of North and South America. These cloud decks have a significant impact on Earth's radiation budget. Aerosols, arising from natural processes and from human activity, have important influences on the brightness and persistence of these clouds. The Variability of the American Monsoon System (VAMOS) Ocean-Cloud-Atmosphere-Land Study (VOCALS) is planning a field campaign that will study the stratus deck off the Pacific coast of Chile and Peru, using in situ and remote aircraft observations, along with satellite and ship-based measurements. Natural and human sources of particles will be observed, as well as the

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roles these particles play in the determining the brightness and lifetimes of stratus clouds. The VOCALS field campaign is planned for October 2008; preliminary work to simulate the southeast Pacific stratus deck will begin earlier that year.

This activity will address Questions 3.1, 3.2, and 3.3 of the CCSP Strategic Plan.

The Ice in Clouds Experiment. The Ice in Clouds Experiment (ICE) will take place in November 2007. The goal of this study is to improve understanding of ice nucleation in the atmosphere. This knowledge will improve the modeling of ice cloud formation, precipitation, and climate effects. The specific objective of ICE is to show that under given conditions, direct measurements of the thermodynamic and kinetic environments of clouds (temperature, humidity, wind) and specific measurable



characteristics of the aerosol, including chemical composition, can be used to predict the number of tiny ice particles that are initially seeded by existing atmospheric particles. ICE will use airborne measurements of clouds along with coordinating ground measurements in mountainous locations such as the Front Range of Colorado and Wyoming. Close collaboration between theory, field, lab, and modeling studies will be emphasized.





#### Highlights of Recent Research and Plans for FY 2008



Completion of CCSP Synthesis and Assessment Product 2.4. In FY 2008, CCSP researchers will finalize CCSP Synthesis and Assessment Product 2.4, Trends in Emissions of Ozone-Depleting Substances, Ozone Layer Recovery, and Implications for Ultraviolet Radiation Exposure. This report will focus on updating trends in stratospheric ozone, ozone-depleting gases, and ultraviolet exposure, and on improving model evaluations of the sensitivity of the ozone layer to changes in tropospheric composition and climate, along with the implications for

the United States. This information is key to ensuring that international agreements to phase out production of ozone-depleting substances are having the expected outcome: recovery of the protective ozone layer.

This activity will address Question 3.4 of the CCSP Strategic Plan.

## ATMOSPHERIC COMPOSITION CHAPTER REFERENCES

- 1) **Velders**, G.J.M., S.O. Andersen, J.S. Daniel, D.W. Fahey, and M. McFarland, 2007: The importance of the Montreal Protocol in protecting climate. *Proceedings of the National Academy of Sciences*, **104**, 4814-4819, doi:10.1073/pnas.0610328104.
- 2) Fu, R., Y. Hu, J.S. Wright, J.H. Jiang, R.E. Dickinson, M. Chen, M. Filipiak, W.G. Read, J.W. Waters, and D.L. Wu, 2006: Short circuit of water vapor and polluted air to the global stratosphere by convective transport over the Tibetan Plateau. *Proceedings of the National Academy of Sciences*, 103, 5664-5669.
- 3) Newman, P.A., E.R. Nash, S.R. Kawa, S.A. Montzka, and S.M. Schauffler, 2006: When will the Antarctic ozone hole recover? *Geophysical Research Letters*, 33, doi:10.1029.2005GL025232.
- 4) WMO, 2007: Scientific Assessment of Ozone Depletion: 2006. Global Ozone Research and Monitoring Project—Report No. 50. World Meteorological Organization, Geneva, Switzerland, 572 pp.
- Volkamer, R., J.L. Jimenez, F. San Martini, K. Dzepina, Q. Zhang, D. Salcedo, L.T. Molina, D.R. Worsnop, and M.J. Molina, 2006: Secondary organic aerosol formation from anthropogenic air pollution: Rapid and higher than expected. *Geophysical Research Letters*, 33, L17811, doi:10.1029/2006GL026899.
- 6) Quinn, P.K., T.S. Bates, D. Coffman, T.B. Onasch, D. Worsnop, P.D. Goldan, W.C. Kuster, T. Baynard, J.A. de Gouw, J.M. Roberts, B. Lerner, and A. Stohl, 2006: Impact of sources and aging on submicrometer aerosol properties in the marine boundary layer across the Gulf of Maine. *Journal of Geophysical Research*, 111, D23S36, doi: 10.1029/2006JD007582.
- 7) Gao, R.S., J.P. Schwartz, K.K. Kelly, D.W. Fahey, L.A. Watts, T.L. Thompson, J.R. Spackman, J.G. Slowik, E.S. Cross, J.-H. Han, P. Davidovits, T.B. Onasch, and D.R. Worsnop, 2007: A novel method for estimating light-scattering properties of soot aerosols using a modified single-particle soot photometer. Aerosol Science and Technology, 41, 125-135.
- 8) See <esrl.noaa.gov/csd/2006>.
- Slingo, A., T.P. Ackerman, R.P. Allan, E.I. Kassianov, S.A. McFarlane, G.J. Robinson, J.C. Barnard, M.A. Miller, J.E. Harries, J.E. Russell, and S. Dewitte, 2006: Observations of the impact of a major Saharan dust storm on the Earth's radiation budget. *Geophysical Research Letters*, 33, L24817, doi:10.1029/2006GL027869.
- Jiang, H. and G. Feingold, 2006: Effect of aerosol on warm convective clouds: Aerosol-cloudsurface flux feedbacks in a new coupled large eddy model. *Journal of Geophysical Research*, 111, doi:10.1029/2005JD006138.
- 11) Jiang, H., A. Xue, G. Teller, G. Feingold, and Z. Levin, 2006: Aerosol effects on lifetime of shallow cumulus. *Geophysical Research Letters*, 33, doi:10.1029/2006GL026024.
- 12) Wilcox, E.M., G. Roberts, and V. Ramanathan, 2006: Influence of aerosols on the shortwave cloud radiative forcing from North Pacific oceanic clouds: Results from the Cloud Indirect Forcing Experiment (CIFEX). Geophysical Research Letters, 33, L21804, doi:10.1029/2006GL027150.
- 13) **Meskhidze**, N. and A. Nenes, 2006: Phytoplankton and cloudiness in the Southern Ocean. *Science*, **314**, 5804, doi:10.1126/science.1131779.

