The Second Intercontinenal Transport and Climatic Effects of Air Pollutants (ICAP) Workshop

October 21-22, 2004

The Friday Center, Chapel Hill, North Carolina

The second ICAP workshop was hosted by the Carolina Environmental Program of the University of North Carolina at Chapel Hill in the Friday Center on Thursday and Friday, October 21 and 22, 2004. More than 80 national and international scientists attended the two-day workshop. Presentations were delivered during five sessions over the two days.

Program Perspectives

The ICAP program's priorities target efforts to understand climate interactions through the integration of climate and international/regional air pollution issues, as well as the impacts of climate and air pollution feedbacks. This U.S. EPA program involves work with other countries through international collaborations. Aspects of international transport of air pollutants are analyzed based on sound science that is subjected to continual evaluation and refinement. Efforts are coordinated to enable strong communication and to formulate activities used to develop international policy.

Global Modeling and Intercontinental Transport

Studies of intercontinental transport of ozone reveal important findings. Anthropogenic NO_x and non-methane volatile organic compound (NMVOC) emissions outside North America enhance surface ozone over the U.S. by 2-6 ppbv on average. Policy-relevant background ozone levels over North America are 20 to 35 ppbv lower than is currently assumed for risk assessment studies (40 ppbv); this results in underestimation of ozone increments from U.S. air pollution. Anthropogenic methane emissions increase surface ozone by another 4-6 ppbv by increasing the ozone background. GCM studies using halocarbon tracer show that there is a clear transport of the plume from the U.S. to northern Europe. Sources from North America contribute to higher ozone episodes over Europe, and affect both seasonal variations and shorter-term temporal trends.

How does pollutant transport differ between North America and eastern Asia, and what are the impacts of oxidation on climate? To what extent does European pollution affect central and eastern Asia, and how does this compare with the effect from North America? The European impacts on Central Asia average 10-30 ppbv for CO, and 1-4 ppbv for O_3. In Japan the North American influence on mid-tropospheric ozone seems to be greater than the European influence.

Global sensitivity modeling studies show that (1) the largest total ozone reductions can occur for regions with high NO_x emissions, and (2) global O_3 changes are most sensitive to NO_x reductions from tropical low-NO_x-emitting regions (SE Asia, South America, India, and Australia).

Air Quality and Climate

Global modeling simulations also suggest a warming due to fossil fuel (ff) and biomass burning (bb) emissions of black carbon (BC) of +0.25 to +0.3 K in the 5-to-10-year average, with a range of +0.15 K to +0.5 K. Maximum warming and cooling due to greenhouse gases (GHGs) and aerosols exceed those from GHGs alone. Aerosols provide a further perturbation of the effects of GHGs to enhance extreme cool and warm climate conditions. Modeled aerosol particles and gas-phase precursors appear to decrease precipitation in mountainous regions and increase precipitation beyond the mountains, to cool surface temperature, to slightly increase atmospheric temperature, to reduce solar and ultraviolet (UV) radiation, and to increase infrared radiation (IR) to the surface in California.

Future emissions of carbonaceous aerosols were projected based on a model developed by Bond and Streets. The model is driven by Intergovernmental Panel on Climate Change (IPCC) regional forecasts of energy, fuel use, and economic activity. The source of carbonaceous aerosols is unburned carbon emitted due to inefficient combustion of fuel, and contains black carbon, which is composed of aerosol particles less than 1 μm in diameter emitted mostly as elemental carbon, as well as particulate organic carbon (OC), in which carbon is bonded to other atoms. The study took into account changes in energy use and fuel type by sector and by region of the world. Improvements in particle control technology included a shift from low-level to high-level technologies over all sectors, as well as improvements in technology within a fuel use sector, e.g., the automobile industry. The results seem to indicate a potential for stabilizing emissions levels of aerosols over the next several decades, partly due to the phase-out of inefficient technologies in the developing world, resulting in reduction of primary aerosol emissions. However, the increase in use of automobiles will increase emissions, somewhat offsetting the reductions due to improved technologies. OC and BC declined under all scenarios for both anthropogenic and biomass burning sources

Modeling studies by Goddard Institute for Space Studies (GISS) show that the impact of BC is sensitive to its vertical profile. When BC is put in the near-surface layer (959 mb), the reduction of low cloud cover over land is smaller than that over the ocean. When BC is put at a higher level (850 mb), the reduction of low cloud cover occurs mostly over land and the coastal region near source, while most of the increase in low cloud cover occurs over the ocean.

Mercury, Emissions Inventories, and Regional Modeling

The importance of wildfires for Hg emissions is being explored. There is high uncertainty in Hg emissions because O_3 and OH can turn Hg^0 to gas-phase mercury. Vegetation acts as a sink for Hg. However, it can also re-emit Hg. The question is how long does it take between deposition and emission.

For biomass burning, the speciation from China differs from the speciation from the West. For mercury, the deposition is more important than the source regions in delineating areas with high concentrations. There are two peaks in the U.S. mercury pattern: in California, and over the Northeastern U.S. The highest anthropogenic emission category of mercury in the U.S. is attributed to power plants (41.5%), followed by mobile sources (24.8%). However, there is evidence that Hg^0 and Hg^{II} are reduced in power plant plumes. Most models do not take this reduction into consideration. From a modeling standpoint, the grid models over predict mercury deposition compared to plume dispersion models. The use of plume-in-grid models in regional models may alleviate some of these problems.

The nesting approach using initial and boundary conditions from a global model for a regional model is much better than the traditionally used approach of fixed boundary conditions for regional modeling. It should be noted that in general the simulations within the global and regional domains are based on two models with different chemical mechanisms. Interpretation of the results should therefore be treated with caution. The same VOCs will mean different things according to the mechanism used. For example, in the case of the GEOS-CHEM and CMAQ models, the GEOS-CHEM chemical mechanism treats VOCs differently than does the CB4 mechanism. Using boundary conditions from GEOS-CHEM, CMAQ shows more variable concentrations at different layers compared to the fixed-boundary-conditions case. Within CMAQ, the SAPRC mechanism predicts higher mercury concentrations than the CB4 mechanism.

In addition to the differences in chemical mechanisms used in global and regional models, the dynamic consistency between the two models could be an important issue in model performance. For example, winds and other meteorological parameters differ when using GISS and MM5 as meteorological drivers to a chemistry-transport model.

Mercury emissions from vegetation are another source of very high uncertainty in global and regional transport models. The global emissions estimate of mercury from vegetative sources varies between 1400 and 3400 tons/yr. Mercury emissions from natural sources are especially important in summer. The major sources are related to emissions from wetlands, maples, pines, and oaks. Anthropogenic mercury emissions dominate in winter. Emission corrections for meteorological parameters (solar radiation, temperature, etc.) are a key factor in accurate estimates of mercury emissions. Temporally resolved, gridded mercury emissions are needed for chemistry-transport models. Emission speciation is important for local deposition, and is dominated by reactive gaseous mercury and particulate mercury from anthropogenic sources. Thus the emission from natural sources is not likely to significantly affect Hg deposition.

Observational Studies: Field Measurements and Satellite Data

Measurement studies confirm the inter-annual variability of intercontinental transport of pollutants. Springtime is the main season for transport; however, summer also shows some interesting and important features. The level of particulate matter (PM) background concentrations seems to have shown no change during the past few years. GEOS-CHEM simulations show non-zero concentrations of CO being transported to the west coast of the U.S. Good correlations between CO and Hg concentrations are shown over Asia, and the Hg/CO ratio provides an important index to characterize the transport of pollutants from Asia. The study shows that the transport is dominated not only by industrial sources but also by dust sources during specific years. For example, the dust storm in April 2001 (April 8, 12, and 14) included both dust and CO. The subsidence affected the boundary layer height getting into the U.S., which made a substantial contribution to $PM_{2.5}$ in the West and East. The year 2003 seems to have been unusually high for biomass emissions in Russia, due to an estimated 9 million hectares burned in Siberian wildfires. The Navy real-time model showed exceedances of NAAQS in several locations in the U.S. on June 6, 2003. The background ozone levels showed a strong correlation with the amount of area burned in Siberia. The trend in background ozone, which is strongest in spring and summer, needs to be more accurately represented.

Episodic transport was also shown not to be the best metric for studying transport from the Pacific. Data from IMPROVE show quite a strong signal of dust transported over the Pacific that is very different from that due to local dust emissions. Organic material shows a high correlation with dust. The Asian component represents 50% to 80% of the regional background of fine PM in the western U.S. The Asian background concentration value is highly variable.

Saharan dust from Asia touches the U.S. in July. This is considered the largest single source of fine particle emissions in the world. Analysis of fine dust (a combination of silicon, aluminum, iron, magnesium, etc.) during 1992-2003 shows episodic maxima that can be 2-3 times larger than the average. Chemical composition is a good measure for distinguishing between local and Saharan dust; the aluminum/silicon ratio can be used as an index in this regard.

Regional and Global Modeling Studies (Asia and Europe)

A model inter-comparison study evaluated the performance of eight models compared to measurements at selected locations in Japan and Taiwan. The study showed large variability among model results even when using the same meteorology and emissions. The inter-annual variability of the transport is evident from the analysis. The effect of the inter-annual meteorological variability on sulfur deposition can be larger than the effect of changes in emissions. Sources of uncertainty in the model predictions include uncertainties in activity patterns and different emissions technologies for CO and BC.

Comparing modeled and observed ratios, such as ΔBC/ΔCO, is a good tool for analysis. Using observations at various altitudes, the Hg/CO ratio did not seem to reveal a simple relationship. Another dimension of intercontinental transport can be analyzed in terms of economic impacts through examining the cost in dollars of each country. Hong Kong and China have the most influence (33% of the global export merchandise), followed by the U.S. (23%). In China, the contribution of export merchandise to industrial emissions ranges from 8% to 91%. The contribution of export to ambient concentrations of NO_x is 18% to 24%, of SO_2 is 21% to 27%, and of VOC is 5% to 7%.

The meteorological patterns associated with short-term climate variability play an important role in characterizing intercontinental transport. The phase of the North Atlantic Oscillation (NAO) controls transport strength and speed, with the positive phase being associated with strong transport across the Atlantic. The variability of the springtime Total Ozone Residual (TOR) based on satellite measurements over the Atlantic is linked to the transport patterns modulated by the NAO. The relationship between the Arctic Oscillation and TOR is even stronger. Similar relationships have been found between pollution over India and the El Niño Southern Oscillation (ENSO) index and the strength of the monsoon flow. In general, population and ozone pollution are strongly correlated in India and China. Asian dust outbreaks are strongly correlated with the Northeast Asian monsoon and inversely related to ENSO.

A regional modeling study of a dust episode over China showed that the difference in grid resolution, while having less impact on the average dust emission rate and on the total budgets, may show differences in timing and spatial distribution. In the source regions the vertical distribution of mass budget can differ by 10%. The differences are much larger near continental outflow regions (a factor of 3 above 4 km).

The lower atmosphere of the Tibetan Plateau has important atmospheric chemistry processes. Atmospheric pollutants transported through the plateau have strong seasonal variation. Stratospheric intrusion events are evident based on Mt. Everest surface observations. The Katabatic wind brings ozone-rich air masses to the surface. The high altitude and albedo result in high UV radiation and high OH concentrations (could be as high as 10^7 molecules cm^{-3}), which result in a high oxidative capacity of the atmosphere and impact the lifetime of pollutants, especially CH_4.

PM_{10} is the dominant air pollutant in China, based on frequency of occurrence. TRACE-P and local emissions inventories show some differences in the emissions of PM_{10}, with TRACE-P underestimating urban area emissions compared to local inventories overall. Local inventories were provided only for urban areas (i.e., no rural emissions were available for comparison).

Workshop Discussions

- *What is the role of current and future anthropogenic emissions outside North America and the potential effects on U.S. domestic air quality?*

- *What is the role of anthropogenic emissions from the U.S. and other developed countries and the potential effects on air quality in other regions?*

- *What is the role of pollutant emissions (e.g., ozone, PM precursors, methane, BC) in contributing to regional climate change?*

- *What other direct and indirect effects of pollutants can contribute to regional climate change?*

The above four bullets are policy-relevant questions that are thought to characterize intercontinental transport and the air quality impacts on regional and global climate.

- *What further improvements are needed in emissions inventories?*

Such improvements should be thought of in terms of scale (regional and hemispheric) and also in terms of processes (vehicular, industrial, shipping, biofuels, biomass burning, natural [wildfires, vegetation/soils, lightning], and unconventional sources). We still need better integration of natural and manmade emissions. We need to improve emission factors, especially for CO, PM, and hydrocarbons (HC). We need to improve and extend inventories for Persistent Organic Pollutants (POPs), Hg, and other species. The same is true for spatial allocation proxies, especially for industries.

- *What is the role of "inverse modeling" as a tool for using satellite observations and ambient air quality measurements to enhance emission inventories?*

If done correctly, this can be an important tool to enhance emission inventories and consequently improve modeling simulations.

- *What are the uncertainties in intercontinental transport modeling?*

Many uncertainties still exist when modeling intercontinental transport, such as the processes of venting from continents and scavenging during this venting.

- *What are the best approaches to evaluating intercontinental transport models?*

Measurements from satellites seem to be a valuable source for evaluating models, along with data based on field measurement studies. For example, satellite data from MODIS show the inter-annual variability of smoke events, which could be useful in model evaluation.

- *What are the key improvements needed in model simulation of aerosol and chemical processes?*

Aerosol and cloud microphysics and radiative interactions are research areas that need more scientific attention. Heterogeneous chemistry and hydrocarbon chemistry are also among the important topics for improving model simulations.

- *What are the physical and numerical procedures needed to address climate forcing by air pollutants?*

Many numerical and physical approaches may need to be tested. For example, two-way nesting between a global and a regional model has not been used extensively. Also, most of the modeling simulations use the offline meteorology-chemistry concept. A coupled meteorology-chemistry model may be adequate to examine the impacts of pollution on climate. In addition, we need to develop a concise modeling intercomparison protocol—basically, how do global and regional models compare and what are the similarities and differences.

- *How do we balance improvements in air quality and emissions on one side with cost and economic consequences on the other side?*

The derivative approach shown in Terry Keating's slides could be very useful. Where shall we put our investment? Mercury and POPs may be more important than ozone and PM, since mercury is global. We need to do the science correctly and design experiments to account for feedback between model results and observations. Mercury is a perfect example that shows a strong connection between model results and observations.

- *What is the importance of dynamical and chemical consistencies when nesting a regional air quality model with a global chemistry model?*

Maybe we can have a Northern Hemispheric version of CMAQ. However, the computational costs may be high, and having the tropics at the boundary would ignore many processes across that region. It will also depend on the problem at hand. For example, do we need a hemispheric or global model to simulate ozone? The time scale is another issue; global or hemispheric simulation may be important when running seasonal or annual simulations. What if we are examining control strategies over a small domain or urban area for a certain episode? It is possible that if we put the boundary conditions far enough into a relatively clean area that we can get a reasonable assessment of the control strategies using a regional model.

- *How do we integrate observations into modeling simulations to improve model performance?*

Four-dimensional data assimilation (FDDA) has to be done correctly in order to benefit from including measurements and observations in models. We have to move quickly on this issue. It is important, however, that we be able to analyze processes that contribute to the evolution and transport of pollutants, without having the measured fields overwhelm the dynamical attribution of the modeling concept. While assimilation itself is not a trivial problem due to the secondary chemical reactions of many chemical species, it is possible as an early approach to use patterns (temporal trends, for example) to develop consensus between model outputs and observations. We need to develop tools to extract information from data that is relevant to model integration.