A51A-07 0930h

Aircraft Observations of the Tampa Urban Plume during BRACE: Transport, Photochemical, and Depositional Processes

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Staff from NOAA's Air Resources Laboratory conducted airborne measurements of trace gases and aerosols in the Bay Region Atmospheric Chemistry Experiment (BRACE) using the NOAA Twin Otter. The Twin Otter flew more than 90 hours in 21 flights in and around the Tampa metropolitan region in May, 2002, at altitudes of 60-3000 m MSL. Flights were conducted over rural and suburban areas, over the centers of Tampa and St. Petersburg, and over Tampa Bay and the Gulf of Mexico. The overall objective of the aircraft measurements in BRACE was to study the emission, transport, and photochemical transformations of nitrogen and other ozone precursors in the Tampa area. Continuous instrumentation was used to measure NO, NOX, NOY, HNO3, CO, SO2, O3, CH2O, and H2O2. A semi-continuous GC technique with luminol detection was used to measure PAN. Filter packs were used to make integrated measurements of nitric acid and inorganic aerosols in both fine and bulk aerosol size fractions. Stainless steel grab cans were filled during flight for post-flight analysis of NMHCs by GC/FID/MS. The urban plume was sampled under a variety of meteorological regimes, as it was advected by the prevailing winds over the Florida peninsula (with continuing input of natural and anthropogenic precursors along the advection path) and, in other cases, over the Gulf of Mexico, where additional chemical inputs were negligible and the plume was relatively unaffected by turbulent deposition processes. Case studies will be used to compare and contrast the photochemical processes in the plume under these different regimes. The observed relationships and variations of trace gas concentrations will be used to determine the efficiency of ozone production, as well as instances of NOX or VOC limitation. Sampling the plume at varying downwind distances, over both land and water, allows the determination of overall rates of photochemical ozone production, NOX and SOX oxidation, and estimates of depositional losses of trace species.

A51A-08 0945h

Studies of Tampa Bay Region Power Plant Plumes during the Bay Region Atmospheric Chemistry Experiment (BRACE)

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The NOAA Air Resources Laboratory made aircraft measurements of chemical and meteorological parameters during 21 flights of the NOAA Twin Otter as part of the Bay Region Atmospheric Chemistry Experiment (BRACE). BRACE was conducted in May 2002. The aircraft flew horizontal transects upwind and downwind of the urban area on 13 of these flights to characterize the urban and power plant plumes. Vertical profiles from 60 to 3000 m MSL were made on most flights. Profiles were made over the Gulf of Mexico, Tampa Bay, and various land sites. On many flights, transects were located immediately downwind of the urban region and power plants and at successive distances farther downwind to characterize the horizontal distribution and chemical processing of the plumes as they aged. At each distance, data was collected during multiple passes at different altitudes to characterize the vertical structure. Many of the downwind passes were flown over the Gulf where sources are limited and the plumes can be observed in relative isolation. The contribution of the power plant plumes are analyzed to determine changes in the vertical and horizontal distribution of the plumes; horizontal fluxes of NOx, NOy, and ozone; production of ozone; deposition rates; and changes on successive days of regional background and concentration maxima caused by the power plant emissions.

A51B MCC: 3018 Friday 0800h

Use of Inverse Modeling for Constraining Global Budgets of Atmospheric Trace Gases I *(joint with B)*

Presiding: **A M Michalak,** NOAA Climate Modeling and Diagnostics Laboratory**; W Peters,** NOAA Climate Modeling and Diagnostics Laboratory**; P P Tans,** NOAA Climate Modeling and Diagnostics Laboratory

A51B-01 0800h INVITED

Regional Carbon Flux Estimation by Inversion of Mesoscale Tracer Transport

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Estimation of carbon fluxes over large regions by tracer transport inversion at global scales is now well established in carbon cycle science, and several studies are now underway in the Europe, the US, and in Brazil to apply similar methods to regional studies over continental areas. Continuous measurements of CO_2 have the potential to better constrain regional CO_2 fluxes through temporal variations associated with synoptic and mesoscale meteorology, and this class of mesoscale inverse problem is expected to be very important in the upcoming North American Carbon Program. We are conducting a pilot study of mesoscale CO_2 inversions using a network of seven continuous analyzers in the region of a tall TV tower (WLEF) in northern Wisconsin. Here we report preliminary results of inverse modeling studies using the new data and experiments using synthetic data for the year 2000. The mesoscale inverse problem is complicated by the need to treat initial conditions, lateral boundary fluxes, and the interaction between mesoscale transport and heterogeneous surface fluxes. Some studies have used zoomed or nested grids in global models to resolve near-field transport, whereas others have specified lateral boundary fluxes from global models. We have taken the latter approach, and simulate tracer transport over a domain much larger than the area of the observations to keep lateral boundaries at a distance. We have simulated the entire year 2000 over a 100 km grid covering most of North America using the CSU Regional Atmospheric Modeling System (RAMS), with two nested grids (dx=20 km and dx=4 km). Meteorological boundary conditions are specified from the NOAA/NCEP reanalysis, with CO_2 mixing ratios at lateral boundaries from the mean of the global TransCom models after flux adjustment to match remote flask observations. We simulate the influence of upstream fluxes on hourly measurements of CO_2 mixing ratio at each tower using a Lagrangian Particle Dispersion Model that calculates thousands of back trajectories for each observation. Linear combinations of spatial and temporal variations of upstream surface fluxes were then scaled to provide optimum agreement with observations constrained by reasonable diurnal and synoptic variations. Our results highlight the importance of vertical resolution, subgrid-scale turbulence, and cloud-scale transports in determining upstream sources and sinks. Sources within one day's advective travel time of the receptor have a disproportionate influence on the estimated flux, which suggests that spacing of observational networks should be made with this metric in mind. Continuous measurements were found to exert significantly stronger constraint on upstream surface fluxes than subweekly vertical profiles.

URL: http://biocycle.atmos.colostate.edu

A51B-02 0815h

Top-down Constraints on North American Carbon Fluxes using CO2:CO Correlations from Aircraft Data

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We employ measurements of CO2, CO and CO2:CO correlations in conjunction with 3-D simulations from a global chemical transport model (GEOS-CHEM) to better constrain specific sources contributing to North American CO2 exchange. The CO2:CO emission ratio varies with efficiency of combustion and provides a characteristic signature of source region and source type, and in particular, of combustion versus biospheric influence. Our analysis of CO2:CO correlations in Asian outflow demonstrated the utility of these measurements in constraining the regional biospheric source of CO2 [Suntharalingam et al. 2003]. Here we employ observed correlations from aircraft campaigns (NOAA and NASA-GTE) over and around the North American continent in conjunction with CO2 and CO measurements from NOAA/CMDL surface sites. We use the GEOS-CHEM model, driven by our best emissions estimates, to identify influences of separate source regions on the CO2:CO measurements. Comparison of modeled and observed concentrations and correlations then enables evaluation of the a priori emissions distributions.

A51B-03 0830h

A First Attempt to use Variability at Synoptical Time Scales in Constraining Trace gas Emissions

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Global scale inversions of e.g. CO_2 fluxes from atmospheric observations normally employ simulated and observed mixing ratios, averaged over a monthly or yearly period. Reasons for this approach may be computational costs, the use of climatological meteorological fields, a limited number of observations, and a limited performance or resolution of the transport model. On or close to the continents it is expected, however, that concentration variations on the shorter time scales contain information about the emission distribution of the observed trace gas. The newly developed Adjoint Version of Tracer Model, version 5 (TM5) has been used to obtain tracer emissions based on 6-hourly averaged observations . TM5 is a global model that allows a two way nested zoom over specific regions. In the model version that is used here, a horizontal resolution of 1x1 degree is employed over Europe. The region of interest is the Mediterranean sea. Measurements taken there during the Minos campaign have been analyzed by using back-plumes' calculated with the adjoint TM5.

A51B-04 0845h

Emission Verification of Greenhouse Gases on the Sub-continental Scale Using Tall Tower Observations and Inverse Trajectory Modeling

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Inert (on the time scale of several days) greenhouse gases like CH_4, N_2O and SF_6 are almost ideal gases to model in trajectory models, as (photo)chemistry and uptake processes can be neglected. The forward and backward (inverse) calculations of atmospheric transport presented here have been performed by using a simple trajectory model, utilizing 3D 96 hour backward trajectory data based on ECMWF analysed windfields.

The trajectories were calculated using the FLEXTRA model v3.3 (*Stohl et al., 1999*). The most important factor determining the atmospheric concentrations besides the greenhouse gas emission strengths obviously is the atmospheric mixing layer height. This mixing height is determined in the model by following a critical Richardson number scheme. We used a long time series of 3 years of continuous high precision vertical concentration gradient measurements of CH_4 and N_2O at the tall tower of Cabauw in the center of the Netherlands to derive estimates of the West European spatial distribution of emissions of these gases and the corresponding trends. The forward and inverse calculations and the high resolution vertical concentration gradient data provide maps of both the local and more remote distribution of the sources. The inverse calculation uses the SVD matrix inversion technique to determine the best fitting solution to the overdetermined system that follows from the combination of the source receptor matrix derived from the trajectory model and the observed concentration data. The resulting best estimate for the emissions of CH_4 are for most parts of Western Europe in agreement with the current inventory data. The inherent uncertainty of the transport model and measurements and how these uncertainties translate into the uncertainty of the inverse calculated emissions is estimated by using a Monte Carlo error analysis technique. Other error estimations are determined from using ensemble trajectories with horizontal and vertical deviations. Several problems related to solving overdetermined problems still remain and options to overcome these will be discussed. **References**
Stohl, A., L. Haimberger, M.P. Scheele, and H. Wernli (1999): An intercomparison of results from three trajectory models. Meteorol. Applications 8, 127-135.

A51B-05 0900h INVITED

How Well Can We Constrain Regional CO2 Budgets Using Inverse Models?

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The atmospheric distribution of CO2 is the 4D response to surface CO2 sources and sinks (or forcing), and the (incomplete) mixing by the atmospheric circulation. Commonly used inversion techniques divide the total forcing spatially and temporally into regions and periods with unknown forcing strengths, and seek the optimal set of forcing strengths that best produces the observed response, in the context of an atmospheric circulation/ transport model. Because of the rapidity of CO2 mixing in the atmosphere, the sparsity of atmospheric observations, the heteorogeneity of the land surface, and high-frequency variability of surface fluxes, current inversion methods have yielded source strengths only for large spatial regions. In this paper, we review what we have learned about carbon sources and sink through inverse modeling, and explore how we could use satellite observations and ground-based ecological observations in inverse models for improving the resolution of CO2 sources and sinks.

A51B-06 0915h

Inverse Modeling of Carbon Monoxide Surface Sources Using MOPITT Satellite Data

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Carbon monoxide CO is a key component of the troposphere. It is the principal sink of hydroxyl radicals OH in the free troposphere (CO global mean lifetime is about 2 months) and thus it controls indirectly the lifetime of many other species, such as methane CH4. In the presence of nitrogen oxides, NOx ($> 10 - 15$ pptv), and sunlight, CO is a precursor of tropospheric ozone O3. The processes leading to the emission of CO are fairly well established. CO is a byproduct of fossil fuel use and incomplete biomass combustion. The incomplete oxidation of hydrocarbons also produces substantial amounts of CO. Uncertainties attached to CO sources are still high and, as a result, the comparison of model results and observations can show large discrepancies. These discrepancies are used to optimize the monthly sources of CO over large regions for the period April 2000 to March 2001.

A51B-07 0930h

Inverse Modeling of CO_2 Sources and Sinks Using Satellite Data: A Synthetic Inter-comparison of Measurement Techniques and Their Performance as a Function of Space and Time

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Several measurement concepts have been proposed for measuring CO_2 from space. Currently, the AQUA and ENVISAT satellites are in orbit and the possibility to retrieve CO_2 from their instruments (AIRS and SCIAMACHY) is being investigated. Measurements from satellites in combination with inverse modeling techniques might lead to significantly improved CO2 flux estimates. We conducted synthetic inverse modeling experiments to investigate the expected performance of several measurement techniques including those used in AIRS, SCIAMACHY and the planned OCO mission. Our inverse modeling set-up solves for fluxes on a global domain and at a relatively high resolution (8x10 degree). This allows us to study the performance of the instruments as a function of temporal and spatial scale. Our results show characteristic strengths and weaknesses of the techniques that were tested, emphasizing the importance of a high measurement sensitivity to CO_2 near the surface and global coverage. An analysis will be presented of the scales that are resolved by the satellite measurements and how they compare with the current surface networks. We will address the issue of instrument precision requirements, and suggest realistic scientific aims of instruments that are in flight at present. Finally, we'll give recommendation for future research and discuss potentially promising new instrument concepts.

A51B-08 0945h

Continental-scale partitioning of fire emissions during the 1997-2001 El Niño / La Niña period

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During the 1997-1998 El Niño event, the terrestrial biosphere experienced drought conditions that triggered widespread increases in fire activity. We combined satellite-based estimates of the timing and spatial distribution of fires, biogeochemical modeling, and an inverse analysis of atmospheric CO anomalies to evaluate the contribution of fire emissions from different continents to trace gas variability during this period. While anomalously high fire emissions from Southeast Asia accounted for 60% of the global fire emissions anomaly during the El Niño period, our analysis identified that significant, and previously underestimated, contributions from Central America (20%) northern boreal regions (10%), and South America (south of the equator; 10%) were also critically important in terms of explaining observed atmospheric trace gas anomalies. Globally, total carbon emissions from fires were 2 Pg C/yr higher in 1998 than in 2000, and accounted for 2/3 of the CO2 growth rate anomaly during late 1997 and early 1998.

A51C MCC: 3020 Friday 0800h

Deep Convective Transport *(joint with SA, AE)*

Presiding: **E Ray,** NOAA Aeronomy Laboratory**; J W Elkins,** NOAA Climate Monitoring and Diagnostics Laboratory

A51C-01 0800h

Evidence of Deep Convective Transport of Reactive Constituents to the Tropical Tropopause Layer Using Convective Influence Calculations

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Isentropic convective influence calculations are performed to identify upstream convective systems impacting air sampled just below the subtropical and tropical tropopause on a flight of the NASA WB-57F from Houston, Texas south to 5°N during the September 1999 airborne mission Atmospheric Chemistry of Combustion Effects Near the Tropopause (ACCENT). Interpretation of ensembles of in situ tracer measurements, in particular NO_y, O_3, and CH_3I, have indicated that much of the air sampled on this flight had been recently injected by deep convection, although from non-pristine marine boundary layers. In addition there were more limited stretches of the flight track with evidence of biomass burning and industrial sources. Consistent with this finding, convective influence calculations indicate that the great majority of the parcels along the flight track had been affected by deep convective events at some time in the seven days preceding the flight; the majority of these were oceanic systems. Under the anticyclonic upper tropospheric flow pattern prevailing north of ∼11°N, the marine convective influence came from systems in the Gulf of Mexico, around Cuba and over the waters east of the Bahamas. Influence from continental systems was sporadic except for one extended portion of the track with air that the calculations indicated had been injected by land-based systems over Guatemala within the previous 12-24 hours. South of ∼11°N, a group of systems east of the Lesser Antilles were major contributors of marine influence. However, as the aircraft approached its turnaround at 5°N, the calculations also show influence from land-based systems over Colombia, consistent with the tracer measurements. One important feature of the results is that the convective influence over the course of the 2600-km flight track is derived from a relatively small number of systems. This is a direct result of the minimal dispersion of the trajectories, particularly those emanating from the marine systems. This can be understand as a consequence of the weak shears in this anticyclonic, summer hemisphere circulation and suggests that, in the tropical troposphere layer at least, the tracer profiles of individual air masses are likely to persist far longer than in the dynamical rough and tumble near the jet streams and other more strongly sheared or vertically mixed environments.

A51C-02 0820h

Observations of deep convective transport of reactive constituents to the tropical and mid-latitude tropopause region

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