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From August 13 to September 25, the Southern African Regional Science Initiative's (SAFARI 2000) dry-season airborne campaign coordinated ground-based measurement teams, multiple research aircraft, and satellite overpasses across nine African nations. Among many others, unique coordinated observations were made of the evolution of massive, thick haze layers produced by biomass burning, industrial emissions, marine and biogenic sources.

The NASA Ames Airborne Tracking 14-channel Sunphotometer (AATS-14) was operated successfully aboard the University of Washington CV-580 during 24 data flights. The AATS-14 instrument measures the transmission of the direct solar beam at 14 discrete wavelengths (354-1558 nm) from which we derive spectral aerosol optical depths (AOD), columnar water vapor (CWV) and columnar ozone. Flying at different altitudes over a fixed location allows derivation of layer AOD and CWV. Data taken during feasible vertical profiles allows derivation of aerosol extinction and water vapor density.

In the talk, we show comparisons with ground-based AERONET sun/sky photometer results, with ground based MPL-Net lidar data, and with measurements from a lidar (CPL) aboard the high-flying ER-2 aircraft. We will use measurements from the Ames Solar Spectral Flux Radiometer to derive estimates of solar spectral forcing as a function of aerosol thickness. Validations of MODIS, MISR and TOMS satellite aerosol and water vapor retrievals will also be presented.

A42B-11 1620h

Airborne FTIR Measurements Obtained Aboard the UW CV-580 During SAFARI-2000

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The University of Montana/Forest Service Airborne FTIR (AFTIR) was installed on the University of Washington Convair-580 for 19 flights between 14 August and 14 September 2000. We quantified the major trace gases in minutes-old biomass burning smoke (namely CO₂, CO, CH₄, C₂H₄, C₂H₂, CH₃OH, CH₃COOH, HCOOH, NH₃, NO, NO₂, and HCN) from 9 fires in both arid and humid, wooded savannas where most global biomass burning occurs. Several of these measurements coincided with TERRA/ER2 overpasses. The AFTIR fire measurements confirmed the importance of oxygenated organic compounds in tropical smoke and provided an emission factor for HCN (a potential biomass burning tracer) that is 20 times higher than previously thought. AFTIR also documented some rapid post-emission chemical transformations in smoke. We measured actual formation rates for ozone and acetic acid in smoke downwind from two savanna fires. The ratio of excess ozone to excess CO reached 9% after a few hours of photochemical processing. The similar ratio for acetic acid increased from 1.4 to approximately 5% over the same time period. We observed cloud scavenging of methanol, ammonia, and acetic acid from smoke and a simultaneous, cloud-related source of formaldehyde in the plumes from two other savanna fires. To our knowledge, the SAFARI-2000 flights provided the first comprehensive characterization of savanna fire smoke samples with explicitly known smoke ages and post-emission processing scenarios. We also measured vertical profiles for CO₂, CO, CH₄, and H₂O under TERRA/ER2 at 5 locations in the southern African gyre, one location in the continental outflow over the Atlantic, and one location in the inflow adjacent to the Indian Ocean. During a 3-aircraft intercomparison we observed trace gas enhancement in the free troposphere due to deep cumulus convection. Finally, we measured high NO_x emission factors for ships off Namibia. Taken together, our FTIR-based measurements of the emissions from ships, savanna fires, the

production and use of biofuels, and the related structure of the African, dry-season troposphere could contribute substantially to global atmospheric chemistry models.

A42B-12 1635h

Multi-Platform Haze Layer Characterization for the Eastern Coastal Region of Southern Africa during SAFARI 2000

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As part of the Southern African Regional Science Initiative (SAFARI 2000) Third Intensive Campaign, two South African Weather Service research aircraft flew missions with the objective to capture trace gas and aerosol characteristics for a variety of pyrogenic, biogenic, and industrial emissions. Coastal research flights were conducted off the southern African coastline covering an area from approximately Richards Bay, South Africa to Maputo, Mozambique. Vertical profiles were conducted in the vicinity of Inhaca Island, Mozambique, which served as a SAFARI 2000 and AERONET validation site. These profiles consisting of aircraft measurements of O₃, SO₂, CO, NO, aerosol concentrations, and size distributions indicate the presence of distinct elevated layers of trace gases and aerosols. The profiles are presented in conjunction with synoptic meteorological data to understand the controls that atmospheric circulation patterns exert on haze layers with respect to horizontal extent, transport and vertical structure. Comparisons of in-situ data obtained by aircraft instrumentation with ground-based AERONET data and with remotely-sensed aerosol products are used to identify possible biases in remotely-sensed retrieval methods.

A42C MC: 133 Thursday 1330h

Bjerknes Lecture

A42C-01 1335h

Testing Cloud Parameterizations Used in Climate Models Against Observations and High-Resolution Cloud Models

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Cloud feedbacks are the single greatest unknown limiting the credibility of simulations of anthropogenic climate change. Current research is aimed at evaluating the physical realism of the cloud parameterizations used in climate models. These evaluations are of course fundamentally based on comparison with observations, including both field data and satellite data. In addition, results from high-resolution cloud models are being combined with observations to provide more stringent tests of cloud parameterizations. This paper will summarize recent work in this area, with emphasis on the evaluation of models through the use of observations obtained through the U.S. Department of Energy's ARM ("Atmospheric Radiation Measurements") Program. We will also discuss recent suggestions by W. Grabowski and others that the high-resolution cloud models can themselves be used as "super parameterizations" within climate models.

A42D MC: 133 Thursday 1445h

Tropospheric Chemistry and Constituents I

Presiding: A Volz-Thomas,

Forschungszentrum Juelich, Institut fuer Chemie und Dynamik der Geosphaere II; J Lamarque, NCAR

A42D-01 1445h

Development and Evaluation of Uniform- and Stretched-grid Versions of the University of Maryland Chemical Transport Model

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We have developed the 3-D global University of Maryland Chemical Transport Model (UMD-CTM) with a stretched-grid feature which allows transport and chemistry to be computed with mesoscale resolution in a region of interest. The model contains the stretched-grid transport algorithm of Allen et al. (2000) and the SMVGEAR II chemical solver, as well as convective transport, eddy diffusion, emission, dry deposition, wet scavenging, and stratospheric influx schemes. The model was first assembled with a uniform grid, run for a complete year, and evaluated with a variety of surface, airborne, and sonde observations. For most species no systematic biases were found in the model results, allowing us to proceed with the stretched-grid version and with applications to investigate the roles of various factors controlling tropospheric chemistry. Park et al. will present results of the stretched-grid UMD-CTM for a series of convective episodes over the central U. S.

A42D-02 1500h

Deep Convection and Its Chemical Consequences over the Central U. S. in a Stretched-grid Chemical Transport Model

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The stretched-grid version of the University of Maryland Chemical Transport Model (UMD-CTM) has been run for the June 1985 period of the Kansas/Oklahoma PRESTORM deep convection field program. The model is driven by assimilated meteorological data from the stretched-grid version of the GEOS-3 DAS. Horizontal resolution is uniformly 0.5 deg. over the central U. S., stretching to a maximum grid size of about 4 deg. on the opposite side of the globe. We examine particular case study convective events in terms of how well the convection is represented in the model and through comparisons of model output with measured trace gas species such as CO, NO, and ozone. Comparison of the model output with mean profiles of airborne measurements for three flow regimes was also performed. The magnitude of the enhancement of photochemical ozone production in the upper troposphere over the central U. S. following major convective events is examined.

A42D-03 1515h

Coupling Atmospheric Chemistry with a Convective Boundary Layer Model

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A convective boundary layer model that couples atmospheric chemistry with a convective boundary layer model was developed for use as a theoretical tool for detailed investigations of the relationship between atmospheric chemistry and boundary layer processes. Also the model will be used for the development of new ad-joint and inverse methods. The system is low-order with 7 variables (3 meteorological and 4 chemical). The model simulates the development of the layer height, the horizontally averaged potential layer temperature, the temperature jump at the upper layer boundary and it includes a chemical mechanism that consists of a scheme of consecutive reactions. The mechanism approximates the chemical scheme for the formation of ammonium nitrate. The model has been applied to simulate field data from the Gulf of Mexico. Ship-based observations of sea surface temperature and atmospheric temperature were made over a period of 18 hours from New Orleans through the Gulf during the wintertime. This period is also in

A42D-04 1530h

Modeling Halogen Chemistry in the Troposphere With a Global 3D Model

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The impacts of halogens (Cl, Br, I) in the marine boundary layer received considerable attention recently as they are important for the budget of other reactive species (e.g. Ozone, OH, DMS). Reactive halogens originate either from sea salt aerosol (Cl, Br) or are directly emitted as organic substances from the oceans (I). So far, model studies were restricted to box or one-dimensional models.

Here we present first results using the global three-dimensional chemistry transport model MATCH. We included a reduced set of gas phase reactions and a parameterization of heterogeneous chemistry. The concentration as well as the vertical profile of sea salt particles are diagnosed from the wind speed using simple parameterizations. First results of the importance of halogen chemistry in the troposphere on a global scale will be presented.

A42D-05 1545h

Regional Modelling of Photooxidants: Strengths, Limits and Future Perspectives

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Three-dimensional numerical atmosphere-chemistry models focussing on specific regions of interest, e.g. Europe or North America are valuable tools to study trace species distributions in the atmosphere, to prepare and interpret field measurement campaigns and to determine the contribution of individual processes to total concentration changes.

However, studies with limited area models have not accounted so far for the influence and the variability of transoceanic transported pollution. There is increasing evidence now that intercontinental transport of photooxidants and precursors takes place in northern mid-latitudes, particularly in the free troposphere. But the

impact of such processes on the levels of pollution of the regions downwind is still poorly known.

A today possibility to consider the large scale distribution of trace species and their temporal variability in limited area models is a global-regional nesting approach as it is also applied for weather forecast simulations. An application of such a nesting approach from the global to the meso-gamma model scale (focus on Europe, Germany, Berlin) will be presented.

As substantial progress in satellite observations of the tropospheric chemical composition including vertically resolved information can be expected in the coming years, assimilation of these data to, for example, forecast chemical weather evolution is one of the future challenges and tasks.

A42D-06 1600h

Identification of CO plumes through assimilation of MOPITT data

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In this paper, we display the use of data assimilation to isolate a specific plume of pollutants originating from the surface. For that purpose, we perform the assimilation (using a suboptimal Kalman filter) in a global chemistry/transport model of a specific chemical species over the whole globe and over the whole globe minus the region of interest. The difference of these two simulations is shown to follow the path of the plume until the assimilation outside the region of interest dampens the signal. As an application we identify CO plumes originating from the forest fires in Montana during the summer of 2000.

A42D-07 1615h

Evaluation of Nitrogen Species Using MOZART During the TOPSE Experiment

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In the troposphere, Nitrogen oxides (NOx) is closely related to ozone chemistry. Nitrogen oxides is also intricately linked to the hydroxyl radical OH, another key atmospheric oxidizing species. The reaction between NO2 and OH leads to the formation of relatively stable nitric acid (HNO3), which can be removed from the atmosphere by precipitation and hence provides an important source of nitrogen for the biosphere.

The distributions of nitrogen species are analyzed during the TOPSE (Tropospheric O3 Production about the Spring Equinox) experiment. The experiment took place from February to May, 2000 in a series of 7 round trip missions from Colorado to northern latitudes. Using the NCAR C-130 flying laboratory, the TOPSE experiment covered a latitude range from 40N to near the North Pole, and from 100 ft. to 25,000 ft. in altitude. The experiment measured O3, NOx, sulfate aerosol and other concentrations which provides a unique characterization of the temporal and spatial distribution of these species to evaluate the effect of several physical and chemical processes on nitrogen species in this region. These data are compared with the calculations of a global chemical/transport model (Model for Ozone And Related chemical Tracers (MOZART)). The model is a comprehensive tropospheric chemical/transport model, calculating the global distribution of 56 gas-phase chemical species. The nitrogen species in the model include NOx, HNO3, N2O5, HNO4, NO3, and PAN. In this study a series of model runs is performed to study the sensitivity of some major factors (including heterogeneous reaction on sulfate aerosols, washout of nitrate in rain drops, stratospheric intrusion, lightning production of NO, and gas to aqueous phase conversion in cloud drops) to the NOx and HNO3 concentrations at high latitudes in North America during winter and early spring. The results show that the heterogeneous reaction on sulfate aerosols and wet deposition of HNO3 have significant effects on NOx and HNO3 concentrations in the lower troposphere, while the stratospheric intrusion affects strongly on HNO3 concentration in the upper troposphere during TOPSE.

A42D-08 1630h

First Results of NOy Measurements Made in MOZAIC Aboard Commercial Aircraft

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The European MOZAIC Program comprises measurements of ozone and water vapor aboard five Airbus A340 aircraft operated by four European airlines. Since the beginning of the project in 1994, more than 150,000 hours of in-flight data were collected. In the second phase of MOZAIC, a small, light-weight and fully automatic NOy instrument was developed at FZ-Jlich and certified by Lufthansa Technik. The NOy instrument, which is calibrated in-situ with zero air, NO and NO2, was installed aboard an aircraft of Lufthansa in Jan 2001 and is producing data since April 2001 (more than 100 successful flights so far). The instrument is exchanged and serviced at monthly intervals. The detection limit is 50 ppt at an integration time of 4s (principal time resolution 0.1 s).

The NOy data obtained from flights between Europe, North America, Asia and Africa are discussed in conjunction with ozone and water vapor with respect to the influence of stratospheric air, pollution plumes from the continental boundary layer and aircraft emissions.

A42D-09 1645h

The Global Distribution and Trends of Atmospheric Nitrous Oxide

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Nitrous oxide is increasing slowly in the atmosphere. It is a potent greenhouse gas, perhaps some 200-300 times more effective, molecule for molecule, than CO2, and it also regulates stratospheric ozone. Yet very little is known about the magnitudes of the sources and sinks of nitrous oxide and why it is increasing. During the last 15 years or so NO2 concentrations have been increasing at about 0.6 ppbv/y or about 0.2 percent/y.

Since pre-industrial times the concentration has risen by about 27 ppbv or 9.4 percent. Based on this change, it is estimated that there must be about 7 Tg/yr of new emissions which are probably anthropogenic. We will examine the present knowledge of the atmospheric latitudinal concentration distribution and trends. Ice core data from various studies are analyzed to show that there are significant systematic errors in both the absolute concentrations and the estimated ages of the ice from which the air is taken for analysis. Moreover, ice core data are imprecise, requiring averaging procedures to make them comparable with modern measurements. The present data are adjusted to arrive at a composite time series that extends to back to about 1000 years. The results show that pre-industrial concentrations were about 287 ppbv and that increases started in the first half of the 20th century (concentrations in 1996 were about 314 ppbv).

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