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Convective clouds provide an efficient mechanism for transporting aerosols to the upper troposphere. Although observational data in the upper troposphere are still limited, the few measurements available all indicate the existence of high concentrations of small particles, possibly due to the vertical transport related to deep convection. In addition, with sufficiently low temperature, high relative humidity, and relatively high concentrations of aerosol precursors; the outflow regions of convective clouds are likely areas for new aerosols to form, adding even more particles to the upper troposphere. In order to simulate convective cloud transport along with cloud processing of aerosols we have developed a 3-D cloud-resolving model with an interactive explicit aerosol module. A baseline simulation suggests good agreement in the upper troposphere between modeled and observed results including concentrations of aerosols in different size ranges, mole fractions of key chemical species, and concentrations of ice particles. A set of 34 sensitivity simulations has been carried out to investigate the sensitivity of modeled results to the treatment of various aerosol physical and chemical processes in the model. The size distribution of aerosols is proved to be an important factor in determining the aerosols' fate within the convective cloud. Nucleation mode aerosols ($0 < d < 5.84$ nm) are quickly transferred to the larger modes as they grow through coagulation and condensation of H₂SO₄. Accumulation mode aerosols ($d > 31.0$ nm) are almost completely removed by nucleation and impact scavenging. However, a substantial part (up to 10% of the boundary layer concentration) of the Aitken mode aerosol population (5.84 nm $< d < 31.0$ nm) reaches the top of the cloud and the free troposphere. The sensitivity simulations performed indicate that in order to sustain a vigorous storm cloud, the supply of CCN must be continuous over a considerably long time period of the simulation. Hence, the treatment of the growth of particles is in general more important than the initial aerosol concentration itself.

A32A-06 1145h

Infrared Study of Water Adsorption on Na-Montmorillonite

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It is estimated that 350 Tg/yr of mineral aerosol particles are available for long range transport. A large fraction of transported mineral aerosol is composed of clays, such as smectite and kaolinite group clays. Because of its unique ability to expand upon addition of water, we have chosen to study the adsorption of water to Na-montmorillonite, a member of the smectite group, under conditions representative of the earth's troposphere. In addition to our studies of these particles as surfaces for heterogeneous chemistry, these particles are also being investigated in other laboratories for their potential as cloud condensation nuclei and for their interference in infrared satellite measurements of temperature. Na-montmorillonite has also been studied as a possible component of the surface of Mars. We have used a Knudsen cell equipped with transmission Fourier transform infrared (FT-IR) spectroscopy to quantify water adsorbed to Na-montmorillonite as a function of temperature and relative humidity. We find at 200 K and 220 K, the water content of the clay increases from 42 mg H₂O/g clay at 15% relative humidity to 111 mg H₂O/g clay at 58% relative humidity with respect to water. These results are in excellent agreement with room temperature, gravimetric measurements. This suggests that although water adsorption to Na-montmorillonite depends strongly on relative humidity there is not a strong temperature dependence. Comparisons to the existing Martian studies, potential relevance to satellite measurements and the implications of our results to tropospheric chemistry will be discussed.

A33A CC: 220 C-E Wednesday 1330h

Tropical Water Vapor: New Understanding and New Challenges III Posters (joint with H, OS, GC)

Presiding: I Folkins, Dalhousie University; S Sherwood, Yale University

A33A-01 1330h POSTER

Water Vapor and Cirrus in the Tropical Tropopause Layer Observed by UARS

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The Microwave Limb Sounder (MLS), an instrument on the Upper Atmosphere Research Satellite (UARS), is sensitive to water vapor in the tropical tropopause layer. We exploit a new dataset from MLS which has enhanced vertical resolution and gives new insights into the vertical structure of water vapor between 215 and 56 hPa. The Cryogenic Limb Array Etalon Spectrometer, another of the instruments on UARS is sensitive to sub-visible cirrus formation in the region of the tropical tropopause. Cirrus clouds play an important role in the dehydration of this region and can contribute to the destruction of ozone. Combining these datasets, we investigate the seasonal and intraseasonal variability of water vapor and cirrus and the implications for stratosphere-troposphere exchange.

A33A-02 1330h POSTER

Asymmetry and Non-linearity in Upper Tropospheric Humidity Variability

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Changes in upper tropospheric humidity (UTH) are important in understanding the climatic response to increased anthropogenic forcing. This paper explores the asymmetry and non-linearity in the response of subtropical UTH to tropical intraseasonal forcing. HIRS (High Resolution Infrared Sounder) UTH data with pentad time resolution, spanning the period January 1979 to December 1998, were used and compared with ERA-40 (European Centre for Medium-Range Weather Forecasts 40 year Reanalysis) UTH and dynamical fields. During northern winter, subtropical UTH anomalies, varying on intraseasonal timescales, propagate eastwards from northern Africa to the central Pacific. These anomalies are associated with the Rossby response to tropical intraseasonal heating in a baroclinic atmosphere. There is a discontinuity in the propagation of moist anomalies over the Tibetan Plateau, whilst dry anomalies show reduced propagation in the lee. This is consistent with the expected evolution of cyclones and anticyclones incident on high topography. Correlations of five-day mean UTH show no significant linear correlation between moist anomalies over Indonesia and subtropical anomalies to the north. This appears to be explained by the limitation of dry anomalies in the region of low ambient vorticity south of the East Asian Jet Stream.

A33A-03 1330h POSTER

Evaporative Moistening Above and Below the Melting Level

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Convective Downdrafts that occur above the 5 km melting level are physically different from those that occur below the melting level. Above 5 km, evaporative moistening tends to be self limiting. Because the evaporation of falling ice quickly brings the tropical atmosphere to saturation locally, the overall rate of evaporative moistening becomes dynamically controlled by the rate at which deep convective regions can import dry air from non convective regions and export humid air to the subtropics. Below the 5 km melting level, the relative humidity of a neutrally buoyant downdraft tends to stay the same or decrease (from its point of origin). This implies that, although an increase in the

downdraft mass flux below the melting level still requires a convergent inflow of air, there is no requirement that this air be dry. This air can therefore arise from a localized mid-level circulation associated with cumulus congestus clouds.

A33A-04 1330h POSTER

Insights into tropical water vapor using stable isotopes

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Stable isotopes of water (H₂-18O and HDO) provide an integrated history of condensation and evaporation processes in the atmosphere due to large temperature dependent fractionation. In the upper tropical troposphere, these isotopes can be used to help us understand the sources and sinks of tropical water vapor and condensate. This work presents an analytic model of water and its isotopes, and compares it to observations from the CRYSTAL-FACE experiment in July 2002. The results indicate that the upper tropical troposphere contains a large amount of water lofted as ice, and highlights the role of convection in maintaining the water vapor distribution in the upper troposphere. The role of convection in hydrating and dehydrating the tropical upper troposphere is discussed. The implications for stratospheric humidity and for water vapor and high cloud feedbacks on climate are also discussed.

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A33A-05 1330h POSTER

Sensitivity improvements in measurement of water vapor isotopic composition in the upper troposphere and lower stratosphere

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There has been much interest in the scientific community in using the isotopic composition of water vapor in the uppermost troposphere and lower stratosphere as a tool to understand the transport and control of water vapor in these regions. The effectiveness of that tool is however a function of the capabilities of the instruments available to measure it. The only model scenarios that can be differentiated are those whose isotopic differences exceed the resolution of the measurements, both in terms of precision and of spatial resolution. We describe here a new in-situ instrument designed specifically for measurement of water vapor isotopes that provides an order of magnitude improvement in sensitivity over previously available technology. The Harvard ICOS (Integrated Cavity Output Spectroscopy) Isotope Instrument is an aircraft-borne absorption spectrometer, approximately 1 meter in length but with an effective absorption pathlength of 4 km, a factor of 40 longer than those in traditional Herriott cell spectrometers. The resulting improved measurement sensitivity allows scientifically useful measurements on a 1 s timescale, providing 200 m horizontal resolution from typical high-altitude aircraft. Estimated total measurement precision is 2%, equivalent to 10 per mil in typical tropopause-region conditions. This precision is required for addressing issues such as the seasonal cycle of water vapor isotopic composition in the lower stratosphere, which could range from virtually absent to as much as 80 per mil, depending on the mechanisms dehydrating air entering the stratosphere. We present model results for different scenarios and demonstrate that the instrument capabilities are sufficient to provide useful discrimination between them. The instrument is scheduled for first flight in autumn of 2004.

A33A-06 1330h POSTER

Upper Tropospheric Water Vapor from AIRS Measurements: Comparison to ARM Sondes

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Water vapor concentrations in the free troposphere acts as a powerful greenhouse gas, yet it remains poorly characterized. The Atmospheric Infrared Sounder (AIRS) on the EOS Aqua satellite has been making measurements since September of 2002. The retrieval products from AIRS include water vapor and temperature profiles with high vertical resolution, mapping over 85% of the globe twice daily. This talk is focused on upper tropospheric humidity as measured by AIRS. We will show a global view of the water vapor fields from 200 to 400 mb, as well as an intercomparison of sonde measurements and AIRS retrievals of UTH from the two ARM sites, the Tropical Western Pacific and the Southern Great Plains.

A33A-07 1330h POSTER

The Effect of Dust Aerosols on Marine Ice Clouds: A Study of MODIS Level-3 Daily Retrieval

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The effect of dust aerosols on marine ice cloud particle size was studied using the Moderate Resolution Imaging Spectroradiometer (MODIS) Level-3 retrieval data. Daily averaged optical thickness of aerosol and daily statistics of ice cloud particles' effective radii were analyzed for May to September 2002 over two regions, a region in the tropical east Atlantic offshore of North Africa, and a region in the northern Indian Ocean in between the Indian continent and the Persian peninsula. It was found that, for clouds that are deep (cloud-top temperature <230K) and optically thick, increasing aerosol optical depth is associated with smaller cloud effective radii. This implies that, through deep convection, dust aerosols blown off the continents during summertime are lofted to the upper troposphere and reduce the effective radii of clouds. The sensitivity of the cloud effective radius to the aerosol loading is quite different between the regions, and reasons for this will be discussed.

A33A-08 1330h POSTER

Tracing the origins of tropical water vapor in a general circulation model

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Understanding the effects of clouds upon tropical water vapor requires information regarding the geographic distribution of the clouds influencing water vapor in a particular target volume. This perspective also helps to identify certain aspects of the circulation important to water vapor. An idealized tracer approach is used to determine the geographic origins of water vapor in a general circulation model, and selected results are compared to prevailing theoretical paradigms of vapor transport. As expected, tropical water vapor enters the free troposphere primarily in convectively active regions, and the vertical structure of a vapor age tracer tracks the descent of air masses as they drift downwind into clear-sky areas. However, the zonal orientation of time-mean flow patterns allows subtropical rather than deep-tropical latitudes to be the primary contributors to subtropical water vapor, and as a result the convective sources are seasonally variable and often continental in location. Stratospheric vapor source distributions indicate that the large-scale ascent which lifts moisture through the model tropopause occurs over a broader range of longitudes than would be consistent with time-mean thermodynamic conditions, highlighting the importance of waves in the desiccation of stratospheric air.

A33A-09 1330h POSTER

A Seasonal Analysis of the Influence of Convection on Tropical and Subtropical UTH using TRMM Precipitation Radar

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Climate change due to the infrared water vapor feedback depends critically on the upper tropospheric humidity (UTH) distribution in the tropics and subtropics, yet the relative impacts of possible sources on this distribution remains uncertain. In this study, we flag tropical vertical water vapor profiles from the Halogen Occultation Experiment (HALOE) and Stratospheric Aerosol and Gas Experiment II (SAGE II) according to water vapor mixing ratio near 200 mb, and integrate a five day isentropic back trajectory from each profile using European Centre for Medium-Range Weather Forecasting (ECMWF) ERA-40 winds. These trajectories are then matched spatially and temporally with Tropical Rainfall Measurement Mission (TRMM) Precipitation Radar (PR) 2A25 volumetric radar reflectivities and binned according to whether the parcel in question has undergone convection that penetrates deeper than 15 km, convection that does not penetrate beyond 15 km, or no convection at all during the previous five days. Subtropical HALOE and SAGE II vertical water vapor profiles are similarly analyzed to determine the distribution of profiles originating in convective outflow from the tropics, subtropical convection from below, and transport from the extratropics. Seasonal variations in both source distributions are investigated across two summers (JJA 1998 and 1999) and two winters (DJF 1998-99 and 1999-2000). We briefly explore the potential benefits of applying the forthcoming EOS-Aura Microwave Limb Sounder measurements of water vapor to this study.

A33B CC: 520 D Wednesday 1330h

Tropospheric Chemistry (Climate)
(joint with B, GC)

Presiding: R V Martin, Dalhousie University; J Thornton, University of Toronto

A33B-01 1330h

Increased Northern Hemispheric Tropospheric CO Burden in 2002 And 2003 Detected From the Ground and From a Satellite

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Carbon monoxide total column amounts in the atmosphere have been measured between January 2002 and December 2003 in the High Northern Hemisphere (30°-90° N, HNH) using infrared spectrometers of high and moderate resolutions. They were compared to the mixing ratios measured in the surface layer and to the total column amounts measured by the Terra/MOPITT instrument. All the data reveal increased CO abundances in comparison with other years. Maximum anomalies (deviations from the "normal" monthly means, averaged over 2000-2001 or over 1996-2001) were observed in October 2002 and August 2003. Nonetheless, these enhancements were twice as little comparing to the record high CO anomaly in October 1998. Most likely, CO emissions from the strong boreal forest fires in Russia and in Canada induced increasing CO burdens.

A33B-02 1345h INVITED

Challenges in the Incorporation of National Emissions into Global Emission Inventories: The United States as an Example.

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Accurate global emission inventories and their trends over decades are critical requirements to accurately model the chemistry of the present and past global troposphere. To a large extent, global inventories are developed by compiling individual national inventories. However, the methodologies employed to develop the national inventories are not always compatible with direct incorporation to the global scale, and the national inventories may have substantial inaccuracies themselves. Consequently global inventories reflect these problems. For example the EDGAR global emission inventory indicates that the CO emissions of the U.S. have increased modestly from 1980 to 1995, while current U.S. emission tabulations report substantial decreases over this period. Further, ambient measurements indicate that U.S. emissions may have decreased even more rapidly than given in the national tabulations. The goal of this presentation is to review U.S. emissions of ozone precursors in the context of global emissions, and to identify possible areas of concern for the accuracy of global models.

A33B-03 1415h

Comparison of the The Radiative forcing of Tropospheric Ozone with Models

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There has been little experimental verification of the radiative forcing from tropospheric ozone. This paper reports on the progress which has been made towards validating the predictions of the climate forcing associated with tropospheric ozone. Measurements have been taken over the last three years with a new technique which was developed to measure the greenhouse radiative fluxes from greenhouse gases beneath clouds. These measurements are valuable since there are large spatial and temporal variations in some gases which make it difficult to quantify their climate forcing. As a result of the poor state of knowledge of the radiative forcing associated with tropospheric ozone, its reduction has been omitted in the Kyoto protocol for the reduction of greenhouse gases as are other prime constituents of smog such as nitric acid or PAN. In our technique, measurements of the surface radiative forcing from the gases below the cloud are taken against the cold black body background of the cloudy sky. Radiative fluxes from ozone, carbon monoxide, nitrous oxide, nitric acid and aerosols have been measured. The process also yields remote sensing measurements of the average boundary layer ozone concentrations. Measurements of the tropospheric ozone surface forcing made for a number of summer days during the past three years have shown that the average surface forcing is about 0.23 W/m². This translates to a radiative forcing of about 0.3 W/m²; it is interesting to note that this value is consistent with the global value of 0.3