Density functional theory (DFT) and ab initio calculations have been employed to investigate the OH-toluene adduct isomers. The geometries and energetics of the four isomers of the OH-toluene adduct radicals as well as their corresponding transition states are presented. The DFT and ab initio theories applicable to the OH-toluene reaction system are evaluated. We also present calculations of the rate constants and isomeric branching ratios of the formation of the OH-toluene adduct isomers.

A51F-0141 0830h POSTER

Mechanism and OH Yield of Ozonolysis of Isoprene

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Although ozonolysis of isoprene represents an important nighttime source of tropospheric hydroxyl radicals, its chemical mechanism remains highly uncertain. The O3-isoprene reaction proceeds through several intermediates, including chemically activated primary ozonides and carbonyl oxides which are critical in determining the final product distribution. We have studied the the formation and unimolecular reactions of primary ozonides and carbonyl oxides arising from the O3-initiated reactions of isoprene using density functional theory (DFT) and ab initio molecular orbital calculations. The structures and energies of these intermediates as well as the transition states and products of their unimolecular reactions were determined. The ab initio energetics were used to determine the reactions and activation enthalpies of formation and cleavage of primary ozonides and unimolecular reactions of carbonyl oxides. We also investigate the reaction pathways of these intermediates using statistical-dynamical master equation and transition state theory. OH yields from both prompt and thermal decomposition of carbonyl oxides are determined.

A51F-0142 0830h POSTER

Two High-Resolution, Quantitative, Infrared Spectral Libraries for Atmospheric Chemistry

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The Pacific Northwest National Laboratory (PNNL) and the National Institute of Standards and Technology (NIST) are independently creating quantitative, 0.10 cm^{-1} resolution, infrared spectral libraries of vapor phase compounds. Both libraries contain many species of use to the gas-phase spectroscopist, including for atmospheric chemistry. The NIST library will consist of approximately 100 vapor phase spectra primarily associated with volatile hazardous air pollutants (HAPs) and suspected greenhouse gases, whereas the PNNL library will consist of approximately 400 vapor phase spectra associated with DOEs remediation mission.

Data are being recorded from 600 to 6500 cm^{-1} to cover not only the classical fingerprint region, but much of the near-infrared as well. The wavelength axis is calibrated against published standards. To prepare the samples, the two laboratories use significantly different sample preparation and handling techniques: NIST uses gravimetric dilution and a continuous flowing sample while PNNL uses partial pressure dilution and a static sample. The data are validated against one another and agreement on the ordinate axis is generally found to be within the statistical uncertainties (2σ) of the Beers law fit and less than 3 % of the total integrated band areas for the 4 chemicals used in this comparison. The nature of the two databases and the rigorous nature used to acquire the data will be briefly discussed.

A51F-0143 0830h POSTER

Detailed Validation of MOPITT Instrument Radiances Using In-situ Profile Data

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Operational retrievals of carbon monoxide profiles using data from the MOPITT (Measurements of Pollution in the Troposphere) instrument are based on a total of 12 calibrated satellite radiances in two spectral bands. To a large extent, the quantitative agreement of the satellite radiances with values calculated by the MOPITT operational forward radiative transfer model determines the ultimate accuracy of the MOPITT CO retrieval results. For example, a radiance bias as small as 1% in one particular signal may produce a retrieval bias in the CO profile larger than 10%.

Validation of both MOPITT Level 1 (calibrated satellite radiances) and Level 2 (CO profile and total column retrievals) products is primarily based on comparisons of MOPITT results with in-situ profiles acquired as part of a program conducted by NOAA's Climate Monitoring and Diagnostics Laboratory. Regular sampling flights have been made at five globally-distributed sites. The goal of the work presented here is to identify and quantify MOPITT radiance biases using the CMDL in-situ data in a manner that accounts for all significant sources of error. These potential error sources include local surface characteristics (e.g. surface emissivity), proximity to CO emission sources, in-situ vertical sampling characteristics, and MOPITT cloud-clearing performance. By minimizing errors associated with each of these effects, we show that the goal of quantifying MOPITT radiance biases to within 1% is realistic.

A51F-0144 0830h POSTER

Recent Global Measurements of Atmospheric COS and Historic Trends Inferred from Firn Air at the South Pole

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Available measurements of carbonyl sulfide provide a picture of atmospheric distributions and seasonal variations that are somewhat inconsistent with our current understanding of COS sources and sinks [see Kjellstrom, *J. Atmos. Chem.*, 1998]. Over the past 1.5 years, we have made measurements of COS at 10 ground-based sampling stations to address some of these concerns. In addition, we have measured COS in firn air from the South Pole in an attempt to reconstruct an atmospheric history of this gas dating back to the early 1900s. The recent data show large seasonal variations at most sites in both hemispheres; summer mixing ratios at Arctic and continental US sites are 20-30% lower than observed in spring. In the northern hemisphere (NH), the timing of the seasonality lends support to strong summertime losses of COS owing to uptake by vegetation and/or soils. The only regular seasonality noted previously for COS in the NH was by total column absorption measurements at mid-latitudes that indicated slightly *higher* ($\sim$3%) mean tropospheric mixing ratios in summer. In the southern hemisphere (SH), smaller ($\sim$10% peak-to-peak) variations are observed at Tasmania and show a maximum during austral summer, perhaps owing to enhanced oceanic flux during that season. No seasonality is apparent at American Samoa (14 S), where larger sample-to-sample variations are observed. Our data suggest annual mean surface mixing ratios for COS that are slightly higher in the SH (by $\sim$2%), but the NH/SH ratio varies by about $\pm$6% depending upon the season.

Data from air trapped in the snow pack (firn) at South Pole suggest that COS mixing ratios in the early 1990s were $\sim$20% lower than observed today. The mixing ratio increases appear to have occurred predominantly in the early part of that century.

A51F-0145 0830h POSTER

Collection and Analysis of Firn Air from the South Pole, 2001

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In January 2001, we collected an archive of 20th century air from the firn (snowpack) at the South Pole. Samples were collected into separate pairs of 3L glass flasks for measurements of O2/N2 (Bowdoin/Princeton) and carbon cycle gases (CMDL); individual 3L stainless steel and glass flasks for measurements of halocarbons, N2O, SF6, and COS; large (33L) stainless steel canisters for maintaining an archive of air for future analyses; and a few canisters each for measurement of 14CH4 (NIWA/CSIRO) and very low-level analyses of SF6 (SIO). Although it was hoped to obtain air dating back to the turn of the century, the analyses suggest that the earliest date was 1925 for CO2 and the mid- to late teens for heavier gases such as methyl bromide or methyl chloride.

This talk will compare the analyses of halocarbons in these recently collected samples to those of air in flasks sampled at the South Pole in 1995. We also will present some results for compounds not measured in the 1995 South Pole samples owing to a paucity of air. Measurements made of the same gases in the firn air at both ends of this six-year interval, along with real-time atmospheric measurements of the same gases, are useful in evaluating assumptions about diffusion in the firn and may allow for the direct calculation of diffusion coefficients at low temperatures. This, in turn, would improve age estimates for firn air samples. New measurements will add to our existing histories established for the 20th century from analyses of firn air samples collected in both Greenland and Antarctica.

URL: http://www.cmdl.noaa.gov

A51G MC: 123 Friday 0830h

Diagnosing Systematic Errors in Numerical Models of the Climate System I

Presiding: **G L Potter,** Lawrence Livermore National Laboratory**; J J Hack,** National Center for Atmospheric Research

A51G-01 0830h INVITED

Addressing the Information Gap between Global Climate Models and Observations

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The output from a large number of model simulations is growing rapidly. This includes model data from Global Climate Models (GCMs), newer "World Models"(with more sophisticated treatment of terrestrial systems, the cryosphere, biogeochemistry, atmospheric chemistry, and/or oceanographic components), reanalysis model simulations, and numerical weather models. The data from these simulations if not now, will soon exceed the amount of archived observational data. Yet,

the scientific value of the model output data is transient and decreases over time, in contrast to the scientific value of the observational data, which persists indefinitely, and in many instances increases over time. For these reasons it is imperative that data generated from models be evaluated and used as quickly as possible. After overcoming data access issues, which can be formidable at times, there are a number of scientific issues that can confound analyses focused on evaluating the adequacy of models through comparison with observations and vice-versa.

Model output is ideal for analysis in many ways. Usually there are no missing data, there is an abundance of variables available for analysis, the data are usually available in a systematic manner over both space and time (often in grid-cell format), and time-dependent biases in climate model simulations are usually small relative to forced changes. The output from weather models or re-analysis model simulations can however, be affected by time-dependent biases in the observations and other biases as a result of changes in the quantity of observations used to initialize the model(s).

Observational data, on the other hand, are collected irregularly in space, are subject to periods of missing data, and often are affected by time-dependent biases. Moreover, there are a dearth of variables that can be analyzed, and the calculation of key derived key variables often have large uncertainties. Nonetheless, observations are the basis for evaluating the adequacy of a model, yet it is clear that models can be used to improve the observational data base.

There are a number of areas where the observational data can be best assessed by making direct use of the output from weather and climate models. Examples include:

(1) the use of climate model output statistics from weather models run in quasi-real time to help identify time-dependent biases in observations, (2) the application of climate model output to assess the affect of missing data, and (3) for observing network design and operation climate, model output can be used to guide the necessary spatial and temporal sampling frequency necessary to adequately monitor climate.

We argue that if analyses such as these were routinely applied to observational data, then climate data bases would be much more effective in evaluating the efficacy of climate models.

A51G-02 0850h INVITED

Successes and Problems with Climate Models: Present and Future

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Examination of the history of climate modeling shows that we are moving toward more objective and quantitative methods of measuring how modeling is improving. Side by side comparison of simulated and observed quantities has been the usual method for determining if the models are performing the correct simulations. We are now moving toward more statistically sound comparisons of the models, although we have to exercise caution because of the limitations of the observations. We do not observe many parts of the earth's climate system very well so the knowledge of what the climate is has a substantial uncertainty, especially in parts of the southern hemisphere atmosphere. The same limitations apply even more so to the oceans, and regions covered by sea ice. We are greatly aided by measurements based upon satellite methods and there is still a strong need for highly calibrated in situ monitoring measurements of the climate system. Even with the limitations concerning data coverage and accuracy, the climate models show some systematic biases. The Atmospheric Model Intercomparison Project (AMIP) and Coupled Model Intercomparison Project (CMIP) have been crucial in showing the common aspects of model bias, however, it is difficult to relate specific biases to the model differences. There is still controversy about whether to use flux adjustments as a method of removing bias in climate models. Models that do not use flux adjustment often use "tuning" to lower the amount of bias. This talk will show what has been learned about model biases in the Parallel Climate Model (PCM) and the Community Climate System Model (CCSM).

A51G-03 0910h INVITED

Are Climate Models Sensitive to the Microphysics of Clouds?

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Parameterization transplant experiments with general circulation models (GCMs) demonstrate that details of cloud-radiation interactions can have large potential effects on the simulated climate. However, in order to determine which aspects of sub-grid physics are important and which parameterizations are most realistic, detailed comparisons with observations are essential. One useful diagnostic tool for making such comparisons is the single-column model (SCM), which consists of one isolated column of an atmospheric GCM. When an SCM is forced with observed horizontal advection terms, the parameterizations within the SCM produce time-dependent vertical profiles of fields which can be compared directly with measurements. In the case of cloud microphysical schemes, these fields include cloud altitude, cloud amount, liquid and ice content, particle size spectra, and radiative fluxes at the surface and the top of the atmosphere. Comparisons with data from the Atmospheric Radiation Measurement (ARM) Program show conclusively that prognostic cloud algorithms with detailed microphysics are far more realistic than simpler diagnostic approaches. These results also demonstrate the critical need for more and better in situ observations of cloud microphysical variables.

A51G-04 0930h

Simulating Reversibility and Assessing Systematic Sources of Errors in Climate Models

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A challenge common to weather, climate and seasonal numerical prediction is the need to simulate accurately long range transport and reversible isentropic processes in combination with appropriate determination of sources/sinks of energy and entropy. A means to study a model's accuracy in simulating internal hydrologic processes is to determine its capability to simulate the appropriate conservation of potential and equivalent potential temperature as surrogates of dry and moist entropy under reversible moist adiabatic processes in which clouds form, evaporate and precipitate. Within the experimental design to examine the differences that develop between the equivalent potential temperature as simulated by the governing equations and its proxy simulated by a transport equation, the continuum equations demand the difference vanish at all discrete model information points throughout the simulation.

In an extension to an earlier study, zonal-vertical cross sections of the differences, relative frequency distributions of the differences and the vertical structure of systematic differences are examined. In the situation where all biases vanish, measures of numerical accuracy are readily related to the classical triangular distribution of the differences of two random variates. A final consideration is to place the random and systematic components of differences within a probability perspective in which the normal distribution is utilized to assess whether the magnitude of the average difference exceeds that expected to develop from the presence of the random component.

A51G-05 0946h

Testing for the linearity of responses to multiple anthropogenic climate forcings

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To test whether climate forcings are additive, we compare climate model simulations in which anthropogenic forcings are applied individually and in combination. Tests are performed with different values for climate system properties (climate sensitivity and rate of heat uptake by the deep ocean) as well as for different strengths of the net aerosol forcing, thereby testing for the dependence of linearity on these properties. The MIT 2D Land-Ocean Climate Model used in this study consists of a zonally averaged statistical-dynamical atmospheric model coupled to a mixed-layer Q-flux ocean model, with heat anomalies diffused into the deep ocean. Following our previous studies, the anthropogenic forcings are the changes in concentrations of greenhouse gases (1860-1995), sulfate aerosol (1860-1995), and stratospheric and tropospheric ozone (1979-1995). The sulfate aerosol forcing is applied as a surface albedo change. For an aerosol forcing of -1.0 W/m^2 and an effective ocean diffusivity of 2.5 cm^2/s, the nonlinearity of the response of global-mean surface temperatures to the combined forcing shows a strong dependence on climate sensitivity. The fractional change in decadal averages ($[(\Delta T_G + \Delta T_S + \Delta T_O) - \Delta T_{GSO}]/\Delta T_{GSO}$) for the 1986-1995 period compared to pre-industrial times are 0.43, 0.90, and 1.08 with climate sensitivities of 3.0, 4.5, and 6.2 ^oC, respectively. The values of ΔT_{GSO} for these three cases are 0.52, 0.62, and 0.76 ^oC. The dependence of linearity on climate system properties, the role of climate system feedbacks, and the implications for the detection of climate system's response to individual forcings will be presented. Details of the model and forcings can be found at http://web.mit.edu/globalchange/www/.

A51G-06 1002h

Methods of Systematic Initial Model Tendency Error Detection

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The causes of systematic errors in the time mean fields of a GCM are difficult to detect from an analysis of the errors themselves. Direct estimates of systematic initial tendency errors (SITEs) determined by assimilation of reanalysis data will on the other hand point more directly to the model components causing the errors. For use with atmospheric climate models (GCMs) three different methods of SITE detection by assimilation of high resolution ECMWF reanalysis data have been tested. The horizontally truncated and vertically interpolated reanalysis data are available only every sixth hour, i.e. at the synoptic times. Between these times interpolated values are used. The initial tendency errors should be computed from initial model stages as close as possible to the reality, defined by the reanalysis. However, by the truncation and the interpolations in the vertical and in time, imbalances are created, which leads to spurious systematic tendency errors, when inserted directly. In the three different methods tested we have tried to reduce such spurious systematic tendency errors by allowing them to be balanced more or less during the assimilation, measuring the goodness of the method by to what extent a balancing of the real tendency errors is avoided. The first method, called "nudging", use simple Newtonian relaxation (nudging), with constant, less than 1 relaxation weights, toward the time interpolated reanalysis data. In the second method only the Slow Normal Modes, SNMs, (with frequencies below a certain cut-off frequency) of the reanalysis data are assimilated in the AGCM. Here we use a time window in the relaxation weights, meaning that the SNMs are inserted fully (relaxation weight 1) at the synoptic times only and in-between are relaxed toward the time interpolated SNMs with a weight decreasing to zero midway between the synoptic times. This method is called "SNMI with window". The third method investigated is similar to the second one except that the total fields, including all normal modes, are assimilated. This method is called "Total insertion with window". Determination of the underlying sources of systematic ECHAM model errors, deduced by these methods of SITE determination, will be discussed in an accompanying presentation.

URL: http://dmi.dk/pub/POTENTIALS/

A51G-07 1038h

Analyzing atmospheric general circulation models with a linearized single column model

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A systematic method for analyzing parameterizations of unresolved physics in atmospheric general circulation models (GCM) is developed. Here, a linear model of the unresolved physics in CCM3 (the atmospheric GCM developed at the National Center for Atmospheric Research) is constructed using a single column version of CCM3. We then examine the most unstable modes, the fastest growing perturbations, and the response to the most likely perturbations by the linear model. This method provides a precise metric with which to analyze the behavior of the totality of the unresolved physics. By systematically removing the effect of individual parameterizations, the behavior of those individual parameterizations is also analyzed.

Results are shown that illustrate the utility of this method and examine the limits for which the linear approximation is valid. For tropical conditions in which

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convection is suppressed, all normal modes of the system are stable or have very small growth rates (time scales of several days). However, rapid perturbation growth can still occur through non-modal interaction.

For conditions in which convection is promoted, large growth rates are found for normal modes in addition to the optimal perturbation. In that case, growth is primarily promoted by deep convection and vertical diffusion, while it is damped by large-scale stable condensation. The comparison of results from two different basic states demonstrate the nonlinearity of the single column model. Nevertheless, for even the convectively active basic state, linear behavior is apparent for several hours, comparable to the lifetime of tropical convection.

A51G-08 1054h

A Lagrangian Approach to the Validation of Spatio-temporal Cloud Properties in a General Circulation Model Using Geostationary Satellite Imagery

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The validation of cloudiness in general circulation models has traditionally been done by comparing time-averaged quantities, such as monthly mean cloud cover, with available observations. This approach ignores the dynamical aspects of clouds such as their lifecycle characteristics, areal coverage, temporal duration, and internal variability. Accurate simulation of the radiative forcing by clouds, and prediction of feedbacks associated with cloud cover require that the model properly represent of the spatial and temporal properties of clouds. In this study, a new Lagrangian approach to the validation of modeled tropical cloudiness is explored. An automated cloud detection and tracking algorithm is used to observe and track overcast decks of cloud with scales from 300 km to greater than 3000 km in a consecutive set of half-hourly METEOSAT-5 images and the NCAR Community Climate Model (CCM3). The algorithm is applied to the deep convective cloud systems of the tropical Indian Ocean region during the 1999 winter monsoon season. The standard diagnostic scheme used to predict cloud cover in the model is based on relationships with convective mass flux and relative humidity. The cloud fraction field is completely recalculated each model hour, hence the simulated clouds have no inherent lifetime, excepting for the time scales associated with large-scale moisture transport. This is consistent with the assumption that the lifetime of cloud material and cloud formation processes are of the same order as the model time step. In contrast, METEOSAT-5 imagery reveals that cloud cover associated with winter monsoon convection is dominated by semi-permanent overcast decks of anvil and cirrus cloud, which persist for durations of days to weeks. Statistics are compiled and compared based on large ensembles of both observed clouds and modeled clouds whose properties, such as areal coverage, temporal duration and cloud-averaged precipitation rate, have been observed throughout the lifecycle of each cloud. This approach tests whether the relationships between convection, relative humidity and cloud fraction presently applied in global-scale models of climate are sufficient to simulate the evolution and spatio-temporal scales of cloud systems associated with tropical deep convection.

A51G-09 1110h

A test for evaluating the downscaling ability of one-way nested regional climate models: the Big-Brother Experiment

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The purpose of this research is to evaluate the downscaling ability of one-way nesting regional climate models (RCM). To do this, a rigorous and well-defined experiment for assessing the reliability of the one-way nesting approach is developed. This experiment, baptised the Big-Brother Experiment (BBE), is used for addressing some important one-way nesting issues.

The BBE consists in first establishing a reference virtual-reality climate from an RCM simulation using a large and high-resolution domain. This simulation is called the "Big Brother". This big-brother simulation is then degraded toward the resolution of today's global objective analyses (OA) and/or global climate models (GCM) by removing the short scales. The resulting fields are then used as nesting data to drive an RCM (called the "Little Brother") which is integrated at the same high-resolution as the Big Brother, but over a sub-area of the big-brother domain. The climate statistics of the Little Brother are then compared with those of the big-brother simulation over the little-brother domain. Differences between the two climates can thus be unambiguously attributed to errors associated with the dynamical downscaling technique, and not to model errors nor to observation limitations. In this talk, we present results of BBEs showing the sensitivity of a RCM to the spatial resolution and temporal update frequency of the lateral boundary conditions.

A51G-10 1126h

Comparison of AMIP and CMIP: How Much Does a "Perfect Ocean" Help Climate Simulation?

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Results from several atmospheric general circulation models, run with sea surface temperature and sea ice amounts prescribed to observed values for the period 1979-1994, can be compared with "control run" simulations by the same atmosphere models coupled to ocean and sea ice models. The simulations with prescribed SST and sea ice are available from the Atmospheric Model Intercomparison Project (AMIP), and coupled ocean-atmosphere simulations are available from the Coupled Model Intercomparison Project (CMIP).

In preliminary work, we have compared CMIP runs from both the NCAR Climate System Model (CSM) and the US Department of Energy - sponsored Parallel Climate Model (PCM) with an AMIP run of their common atmosphere model, the NCAR Community Climate Model Version 3 (CCM3). All three simulations have common weaknesses that presumably orginate in the atmospheric model. Replacement of the CCM3's "perfect ocean" with either the CSM's or PCM's ocean model degrades the level of agreement with observations for most fields, but r.m.s. errors in atmospheric variables from the coupled simulations are surprisingly similar to those from the simulation in which SST and sea ice are prescribed to observations.

This work was performed under auspices of the U.S. Department of Energy by University of California Lawrence Livermore National Laboratory under contract No. W-7405- Eng-48.

URL: http://www-pcmdi.llnl.gov/cmip

A51G-11 1142h

Diagnosis of Climate Model Simulations by Downscaling With a High-Resolution Regional Model

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Climate models must parameterize subgrid-scale processes, and the diagnosis of the consequences of these simplifications and development of better parameterizations is an important part of improving climate simulations. Here we use a dynamical downscaling technique as a method of evaluating the performance of climate simulations from a coarse resolution general circulation model (GCM). For the eastern part of the United States, we first validate a regional model (Regional Atmospheric Modeling System - RAMS) by driving it with real atmospheric data (NCEP reanalysis) and compare the results to actual observations. RAMS is then nested within the NASA Goddard Institute for Space Studies GCM, using the same technique as when driven by reanalysis data. We compare the downscaled results to the GCM-scale simulations and to observations to identify systematic deficiencies in the GCM performance.

A51H MC: 133 Friday 0830h

Chemistry and Transport Near the Tropopause: Results From the 1999 and 2000 ACCENT WB-57F Field Campaigns I

Presiding: **R Friedl,** Jet Propulsion Laboratory**; K Rosenlof,** NOAA**; M Ross,** The Aerospace Corporation

A51H-01 0830h

Investigations of Rocket Engine Combustion Emissions During ACCENT

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The composition of rocket combustion emissions and the atmospheric processes that determine their stratospheric impacts are poorly understood. While present day rocket emissions do not significantly affect stratospheric chemistry, the potential for vigorous growth of the space transportation industry in coming decades suggests that rocket emissions and their stratospheric impacts should be better understood. A variety of in-situ measurements and modeling results were obtained during the Atmospheric Chemistry of Combustion Emissions Near the Tropopause (ACCENT) effort that will be used to evaluate the role of rocket exhaust in perturbing ozone chemistry in plume wakes and in the global stratosphere. We present a review of the ACCENT rocket emissions science objectives, summarize data obtained during the WB-57F plume wake sorties, and briefly discuss how the data will help resolve several outstanding questions regarding the impact of rocket emissions on the stratosphere. These include measurement of the emission indices for several important rocket engine combustion products and validation of plume wake chemistry models.

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Distributions of NOy, HNO3, and NO During the Sept. 1999 ACCENT Flights Including an Athena Rocket Plume

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Total reactive nitrogen NOy and NO were measured on 6 flights of the WB57 between 4 km in the troposphere and 18 km in the lower stratosphere during the September 1999 ACCENT missions. HNO3 was measured on several flights using a chemical ionization mass spectrometer instrument. The flights were based from Ellington Field, south of Houston, Texas and examined the region above hurricane Floyd, aircraft emissions in the neighborhood of the Dallas/Fort Worth airport, and the tropical upper troposphere/lower stratosphere on a flight which terminated in Costa Rica. An overview of the distributions and some aspects of the changes in partitioning of NOy between the troposphere and lower stratosphere are given. On one flight successive passes were made through the exhaust plume of an Athena II rocket that is powered by a solid rocket motor. The passes covered a plume age of 5 to 40 min. NO in the plume was depleted as expected but the HNO3/NOy ratio was apparently much larger than can be accounted for by known gas phase processes.