

A12E-06 1715h

EOS Microwave Limb Sounder Retrievals: New Approaches, Theory and Results From Simulations.

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An overview is given of the retrieval and forward model algorithms that form the 'Level 2 processing' for the Microwave Limb Sounder (MLS) instrument on the EOS Aura spacecraft. These are the algorithms used to convert the calibrated observations of thermal microwave emission from the Earth's limb into estimates of atmospheric temperature and composition for use by the scientific community. The algorithms take into full consideration atmospheric inhomogeneities along the line of sight, in what is essentially a 'tomographic' view of the limb sounding optimal estimation retrieval problem. Results are shown from pre-launch simulations of the EOS MLS measurement system. These give the expected precision of atmospheric data products from the MLS instrument, and have been key to the MLS science team's preparation for the EOS Aura launch. Great benefit has been obtained from designing the software to be flexible enough for both routine data processing and the generation of simulated data and various other pre- and post-launch support and testing.

A12E-07 1730h

PV Mapping and the Precision of Lower Stratospheric Ozone Column Measurements From Satellites

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AURA is expected to produce more precise measurements of ozone throughout the lower stratosphere than has been possible from previous satellite instruments. Excellent precision in lower stratospheric ozone measurements is needed in order to assess ozone changes in this region and, in particular, for inferring tropospheric ozone columns by the residual method. In this paper PV mapping (using the NCEP re-analysis data set) is combined with SAGE II and HALOE ozone measurements and the precision of these measurements and of PV mapping at mid-latitudes is assessed. It is shown for example that PV mapping in the lowest part of the stratosphere can account for a significant fraction of the differences between ozonesonde measurements at Hohenpeissenberg and Payerne which are approximately 400km apart. There is no difference between using SAGE or HALOE or a combination of all the ozonesonde measurements at mid-latitudes for defining the PV-ozone relationships. The calculations thus suggest that the limitation on the precision of PV-mapped ozone is determined by the precision of PV estimates from the assimilation models. The precision of PV-mapped, mid-latitude, lower-stratospheric ozone columns from SAGE and HALOE measurements is estimated.

A12E-08 1745h

The High Resolution Dynamics Limb Sounder (HIRDLS) Validation Plan

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HIRDLS is an infrared limb-scanning radiometer designed to sound the upper troposphere, stratosphere, and mesosphere to determine temperature; the concentrations of O₃, H₂O, CH₄, N₂O, NO₂, HNO₃, N₂O₅, ClONO₂, CFC11, CFC12, and aerosols; and the locations of polar stratospheric clouds and cloud tops. HIRDLS will obtain distributions of constituents with horizontal and vertical resolution superior to that previously obtained. The vertical resolution is approximately 1.2 km. The horizontal resolution is tunable depending on science and validation needs. Typical scan modes will have 5 x 5 degrees global coverage in a 12 hour period. Special scan modes are available for regional coverage with a horizontal resolution of approximately 1 x 1 degrees. The overall validation goals are to provide an assessment of the uncertainties of each HIRDLS data product. For this presentation, HIRDLS validation priorities and challenges will be discussed in conjunction with available correlative data. The importance of temperature, O₃, and H₂O sondes and lidar data will be discussed, with gaps in current measurement frequency and distributions highlighted. Special attention will be focused on the planned NASA aircraft campaigns (e.g., INTEx E, W; TC4, and POLAR). The need for aircraft data for validation of gravity wave structure will be shown. The inclusion of heavy lift balloons (e.g., for Mk IV) will be emphasized, especially for validation of correlative data where stratospheric distributions are limited (e.g., ClONO₂, N₂O₅, and HNO₃). The value of cross comparisons between HIRDLS and the other Aura instruments, along with available occultation and other limb viewing instruments will be outlined. The role of 3-D chemical transport models driven with analyzed meteorological fields for zero order validation, along with the use of chemical data assimilation techniques will also be discussed.

A12F MCC: 3016 Monday 1600h Chemistry and Physics of Clouds (joint with SA, AE, GC)

Presiding: G L Kok, Droplet Measurement Technologies; L M Avallone, Laboratory for Atmospheric and Space Physics, University of Colorado

A12F-01 1600h

Ground Based Remote Sensing of the First Aerosol Indirect Effect: An Update

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The first aerosol indirect effect can be defined as an increase in the shortwave albedo of clouds due to higher concentrations of atmospheric aerosol, whereby the aerosol acts as cloud condensation nuclei to produce increased cloud droplet concentrations and smaller, more reflective droplets. The current work is one step toward achieving a more complete understanding of the indirect effect, which will consequently allow for a better determination of how changes in cloud induced by aerosol may affect the radiation budget and thus the climate. We utilize a series of continuous ground-based measurements from the Southern Great Plains (SGP) Atmospheric Radiation Measurement (ARM) program to investigate the indirect effect. Days that exhibit ice-free, single layered, nonprecipitating clouds are analyzed, with the indirect effect quantified as the relative change in cloud droplet effective radius for a relative change in aerosol extinction (under conditions of equivalent cloud liquid water path). Several cases from the first six years of our analysis (1998-2003) are described here, and probable reasons for the differences in the cloud response to aerosol among the cases are discussed.

A12F-02 1615h

Effects of Arctic Haze on Low-level Cloud Microphysics

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Aerosol measurements from the NOAA Climate Monitoring and Diagnostics Laboratory (CMDL), and cloud retrieval data from the Atmospheric Radiation Program (ARM), located near Barrow, Alaska, are used to study the interactions between Arctic Haze and low-level arctic cloud microphysics. The measurements of aerosol scattering (σ_{sp}), droplet concentration (N), and droplet effective radius (r_e) show that haze events are associated with higher concentrations of droplets and smaller droplet effective radii. Nucleation events, with high Aitken nucleus concentrations but low (σ_{sp}), show no relationship with N or r_e . Therefore, long-range transported aerosols, rather than those produced by in-situ production, dominate the impact on cloud microphysical and radiative properties in Arctic winter through spring.

A12F-03 1630h

Airborne Digital In-Line Holographic System for 3-D Imaging of Cloud Particles

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We have developed an airborne holographic instrument for obtaining cloud particle sizes and shapes and their three-dimensional spatial distribution. The primary scientific motivation for the instrument is to provide *in-situ* measurements of cloud particle spatial correlations, conditioned on particle size. Accurate quantification of spatial correlations at cm-scales and below has been elusive, in part because only one-dimensional spacing data have been available. The optical technique used in the instrument is based on 'in-line holography,' where the same collimated light source serves as both the reference beam and the object beam. The system is completely digital, including the capture of holograms using a CCD array, transmission of the data via optical fiber, and subsequent reconstruction of the real image. During the recent IDEAS-3 field project hosted by the NCAR Research Aviation Facility the instrument was tested in flight for the first time, where holograms containing populations of cloud droplets, drizzle drops, and ice crystals were obtained.

A12F-04 1645h

A theoretical and modeling study of volatile chemical partitioning during cloud hydrometeor freezing

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Thunderstorms can significantly impact chemical distributions in the troposphere by 1) redistribution of air and hydrometeors containing trace chemicals and 2) providing a multi-phase environment for chemical phase changes and reactions. Interactions between ice-containing hydrometeors and chemicals are not well understood. Laboratory and field measurements of chemical partitioning during freezing provide greatly varying estimates of the retention efficiency of volatile solutes. In this work, we develop the theory of volatile chemical partitioning during hydrometeor freezing (termed 'freezing retention'), apply it for a variety of freezing conditions and chemicals, and compare the results to available experimental data. By analyzing the hydrometeor-scale processes involved in retention for non-rime freezing, dry-growth riming, and wet-growth riming, we investigated the factors that control it. For non-rime freezing, we developed a theoretical dimensionless number to indicate retention, derived its dependence on conditions and chemical properties, and calculated its value for several freezing cases for SO₂, H₂O₂, NH₃, and HNO₃. Retention is apparently highly chemical specific, controlled largely by the effective Henry's constant (and hence the drop pH for dissociating chemicals). Chemicals with high effective Henry's constants (HNO₃) will be fully retained during freezing, while chemicals with lower effective Henry's constants (SO₂) will undergo some loss. For chemicals that undergo loss, the degree of retention depends on freezing conditions. Retention likely increases with decreasing temperature and exhibits a maximum at intermediate drop sizes and ventilation. For dry-growth riming, we extended the development to predict retention and compared predicted values to experimental data from several measurement studies. The model agrees well with the data and provides a quantitative explanation for the differences in measured retention. For wet-growth riming, we developed a steady-state retention model. It suggests retention is dependent on the fraction unfrozen water in the riming hydrometeor. This work provides theory-based hypotheses regarding the dependence of retention on physical factors and chemical properties that can be used to develop robust parameterization in cloud models.

A12F-05 1700h

Equilibrium Cloud Droplet Size due to Competitive Condensational Growth on Mono-size Plural CCN

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Introduction

The equilibrium size of the cloud droplet condensed on cloud condensation nucleus (CCN) is commonly estimated by traditional Köhler model derived from the thermodynamic equilibrium between liquid and gas phases separated by a curved interface. However, Köhler model is based on the idealistic assumption that the cloud droplet grows in an infinitely large reservoir of water vapor at constant pressure and constant temperature. As for application of Köhler model to more realistic air parcels, this brings about two major faults in the droplet size estimation. The first is the limitation of the maximum allowable saturation ratio. The limitation produces curious situation as the equilibrium size cannot be decided in case of larger saturation ratios than the critical saturation ratio. The second is the inability to distinguish competitive growth of plural droplets from non-competitive growth of a single droplet. In other words, Köhler model fails to consider the effect of CCN number density on droplet size. A new model free of these faults has been made by supplementing mass and heat conservation equations to the traditional Köhler equation.

Mathematical Model for Cloud Droplet Size

The new model consists of three governing equations

and enables to estimate radius a_e , saturation ratio S_e and temperature T_e in equilibrium state. The governing equations are derived from (1) equilibrium of the droplet chemical potential μ_w with the vapor one μ_v as: $\mu_w(S_e, T_e, a_e) = \mu_v(S_e, T_e, a_e)$, (2) mass conservation represented by liquid water mass m_w and vapor water mass m_v as: $d(m_w + m_v) = 0$, and (3) heat energy conservation represented by enthalpy h_x per unit mass of m_x (suffix x gets w, v and a for liquid water, vapor water and air, respectively) as: $d(m_w h_w + m_v h_v + m_a h_a) = 0$. The equilibrium gives the relation between a_e and S_e (similar to Köhler equation). The mass conservation enables to obtain S_e from initial saturation ratio S_0 . The heat energy conservation relates T_e with initial temperature T_0 .

Conclusions

Numerical simulations by the new model show that: (1) Reduction of equilibrium cloud droplet radius a_e with increase in CCN number density N , i.e., reduction due to competitive growth, becomes more remarkable with increase in initial saturation ratio S_0 ; (2) Equilibrium temperature increase $T_e - T_0$ from initial temperature T_0 with increase in N is not monotonous but has the minimum value; (3) $T_e - T_0$ by competitive growth is too small to have an effect on a_e , but, it seems to be large enough to make the air parcel statically unstable and trigger off the thermal convection in the atmosphere.

A12F-06 1715h

Turbulent coagulation of cloud droplets - Impact on sulfate production in clouds

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Clouds represent a major environment for atmospheric chemistry and measurements have shown that cloud droplets of different size can differ substantially in their composition and pH value. Mixing of droplets of different sizes, however, as occurs when cloud droplets collide and coalesce, averages differences in their composition. This clearly impacts the chemical processes in clouds, such as the sulfate production and other important aqueous phase reactions, and eventually also affects the aerosol phase when the cloud drops evaporates. Laboratory and numerical work have shown that the velocity and spatial distributions of particles may be modified significantly in a turbulent flow field. Although there is a general agreement in current literature that turbulence enhances the collision frequency of cloud droplets, this process is not yet well understood and therefore ignored in most current cloud models. This study therefore addresses the question of how the collision rate of cloud droplets is enhanced due to turbulence in the cloud and how this process impacts the in-cloud chemistry. Since experimental data of the droplet-turbulence interactions is difficult to obtain, modeling studies are an important tool for investigation. Recently, direct numerical simulations (DNS) have been carried out to obtain expressions for the coagulation kernels due to the turbulent flow. Important mechanisms of the turbulence-droplet interaction can be identified. The key mechanisms are that: (1) coagulation rates should be enhanced due to local concentration increases for particles with response times on the order of the Kolmogorov scale, (2) particle inertia leads to relative velocities and less correlated velocity directions and hence to higher collision rates, (3) wind field shear produces collisions between particles even with the same inertia. For this study, a box model is developed to carry out simulations of cloud droplet evolution incorporating the effects mentioned above and the results from the DNS. On the basis of the resulting droplet distribution the in-cloud chemistry is calculated focusing on the conversion of SO₂ to sulfate. The individual processes are quantified and their importance for the sulfate production is assessed for different atmospheric conditions.

A12F-07 1730h

Sensitivity Study of the Vertical Velocity Variation on Cloud Droplet Nucleation Process Using an Adiabatic Parcel Model

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Eleven profiles through liquid water cloud obtained during RACE (Radiation, Aerosol and Cloud Experiment) and NARE (North Atlantic Regional Experiment) are used to study the sensitivity of cloud droplet nucleation to the vertical gust velocity. Selected cloud microphysical data, size-distributed aerosol properties and particle chemistry are applied in an adiabatic parcel model to predict the activated cloud droplet number concentrations (N) using the frequency distribution of the measured in-cloud vertical velocities and again using a vertical velocity characteristic of observations. The simulated adiabatic value of N obtained using the standard deviation of the vertical velocity distribution agrees with the observed maximum N (the cloud droplet number in an adiabatic core) to within 5%. If the parameterization derived by Lin et al. [1997] is applied to obtain the cloud-average N from the maximum N , the average N agrees with the observed cloud-average N to within 20%. The simulated N obtained using the full probability density function of the vertical gust velocities is one approach that has been used to represent the cloud average N . This is based on the assumption that the average N is controlled by all variations in the updraft and not by the mixing process [Leitch et al. 1996]. The value of N obtained in this manner is found to be higher than the observed average N by a factor of two. We believe that this result is because low vertical velocities do not contribute effectively to the cloud droplet nucleation. If we neglect the lowest 45% of all vertical velocities, then the difference between the simulated average N and the observed mean N is reduced to within 13%. These results suggest that it is appropriate to use a characteristic vertical velocity to predict the cloud droplet number concentration in climate models as done by Lohmann et al. [1999], where the subgrid variation of vertical velocity is diagnosed from the turbulent kinetic energy. The frequency distribution of vertical velocity is needed to represent the in-cloud vertical fluctuations. However, this work suggests that a modified frequency distribution of vertical velocity accounting for only the larger vertical velocities to predict the cloud droplet number concentration is more appropriate than using the full frequency distribution. To obtain a better understanding of this issue, it will be important to use both modeling and observations to investigate which updrafts in clouds contribute to cloud droplet nucleation. References: Lin, H., and Leitch, W.R., Development of an in-cloud aerosol activation parameterization for climate modeling, in Proceedings of the WMO workshop on Measurement of Cloud Properties for Forecast of Weather, Air Quality and Climate, pp. 328-335, Geneva, World Meteorol. Organ, 1997. Leitch, W.R., Banic, C.M., Isaac, G.A., Couture, M.D., Liu, P.S.K., Gultepe, I., Li, S.-M., Kleinman, L.I., Daum, P.H. and MacPherson, J.I., Physical and chemical observations in marine stratus during the 1993 NARE: Factors controlling cloud droplet number concentrations, J. Geophys. Res., 101, 29123-29135, 1996. Lohmann, U., Feicher, J., Chuang, C.C., and Penner, J.E., Predicting the number of cloud droplets in the ECHAM GCM, J. Geophys. Res., 104, 9169-9198, 1999.

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Aerosol Physics and Chemistry Within a Convective Cloud

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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. Observational data as well as weather center reanalyzed data have been used to initialize the model simulations. To evaluate the model, the results are compared with observed concentrations of aerosols and certain key chemical species, particularly in the upper troposphere. A number of model runs with various different settings in physics and chemistry as well as aerosol profiles have been carried out. The sensitivities of deep convection related physics and chemistry and its impact on tropospheric chemistry to various aerosol profiles will be discussed.