for the infrared emissivity approach, the mesoscale radiative transfer differs from solvers employed in GCMs because more details are available in mesoscale models. This presentation concluded with the presentation of several current radiative transfer schemes in use.

Dean Churchill's presentation concentrated on the phenomenology of mesoscale flows as influenced by radiation. He gave a short summary of interactions between radiation and cloud physics, radiation and dynamics, and radiation and convection. He discussed Houze's (1989) paper stressing the differences between convective and stratified parts of mesoscale convective systems and their implications for large-scale heating. He then reviewed the work of Churchill (1992) discussing the role of solar and infrared radiation in stratified regions of tropical cloud clusters (an EMEX case study), and that of Churchill and Houze (1991) concerning the interaction between turbulence and radiation. Finally, he mentioned some implications of mesoscale circulations in tropical cloud clusters for large-scale dynamics and climate (Hartman et al. 1984).

Robert Cess discussed lessons learned from the intercomparisons of GCM radiative transfer codes. He discussed an international project to isolate and understand interactive processes in general circulation models as well as in observational data. To date 12 GCMs have been used to produce 24 simulations of global warming caused by a doubling of atmospheric carbon dioxide. Cess enumerated possible reasons for model disagreement, namely differences in radiation codes, differences in atmospheric temperature structure, differences in radiative overlap by atmospheric water vapor, differences in the radiative impact of clouds, and coding errors. He warned to "never adjust more than one thing at a time or it will be impossible to tell which adjustment produced what result".

3.9 Chemistry on the mesoscale: modeling and measurement issues

Anne Thompson

John Pleim:	*RADM - A coupled chemistry/mesoscale model*
Christopher Walcek:	*Convection in RADM (Regional Acid Deposition Model)*
Jason Ching:	*Unresolved issues for mesoscale modeling with chemistry: non-precipitating clouds*
Frank Binkowski:	*Unresolved issues for mesoscale modeling with chemistry: aerosols*
Wei-Kuo Tao:	*Tracer Studies with GCEM (Goddard Cumulus Ensemble Model)*
Russell Dickerson:	*Field observations of trace gas transport in convection*
Kenneth Pickering:	*Photochemical consequences of convection*

The talks in this session pointed out that we have only begun to investigate the consequences of mesoscale meteorological features for atmospheric chemistry. *Uncertainties that exist in many modules of regional and cloud-scale chemical models could be reduced by incorporating chemical measurements and modeling into a Coordinated Multiscale Experiment (CME). Conversely, the use of chemical tracers in a CME can much better define air motions on both cloud and mesoscale.*

Jonathan Pleim discussed the various applications of the RADM (Regional Acid Deposition Model) coupled chemistry/mesoscale model and issues such as the amount of cloud cover produced by the model, PBL processes, biosphere/atmosphere interactions, and subgrid-scale photochemistry. Chris Walcek then presented the transilient matrix convective parameterization that is now in one version of RADM. Jason Ching and Frank Binkowski emphasized unresolved issues for mesoscale chemistry modeling, with regards to nonprecipitating clouds and aerosols, respectively. In particular, Ching discussed parameterizing the fraction of boundary layer air that is vented to the free troposphere by nonprecipitating clouds, describing the modeling of cumuli as flow through chemical reactors. Binkowski discussed development of the Regional Particulate Model, which will facilitate studies of the distribution of sulfate particles, with particular emphasis on the importance of ammonia. Wei-Kuo Tao described the GCEM (NASA/Goddard Cumulus Ensemble Model) and an associated tracer advection model, and showed a video tape of the 3-D redistribution of CO by a major squall line. Russell Dickerson's talk was concerned principally with aircraft chemical observation capabilities for a multiscale experiment, and he showed observations of stratosphere/troposphere exchange in a major MCS. Kenneth Pickering summarized convective enhancement of ozone production in the free troposphere for several case studies, and also showed possible flight strategies for verifying tracer and photochemical model results.

These talks all reviewed the current knowledge and research needs for chemistry on the meso and cloud scales. These needs closely parallel recommendations of a National Research Council (NRC, 1992) report which focuses on the ozone pollution problem in the U. S.. The NRC report points out that tropospheric ozone is a multiscale problem (urban, regional, global) and emphasizes that treatments of surface and boundary layer processes (including natural HC emissions from vegetation) and cloud venting are required for understanding the production and distribution of ozone in the troposphere.

1) Coupled Chemistry/Atmospheric Models

Although coupled models are now available for scales ranging from cumulus cloud scale to global scale, the colloquium emphasized coupled cloud scale and mesoscale (regional) models.

- *Regional models*

One coupled mesoscale-chemical model is the Regional Acid Deposition Model (RADM), which was developed during the 1980's to study source-receptor relationships between pollution emission and acid deposition [Chang et al. (1987); Walcek et al. (1990); Pleim et al. (1991); Pleim and Chang (1992)]. The model now resides at the Environmental Protection Agency in Research Triangle Park, NC, but versions exist at the State University of New York at Albany for a variety of atmospheric chemistry applications. Advection and dispersion of pollutants in RADM is driven by meteorological fields produced by the MM4 version of the Penn State/NCAR Mesoscale Model. Considerable effort was employed in developing the RADM chemical mechanism, although other mechanisms may be substituted into the model. Two model components in particular that are fairly crude and require additional work: (1) parameterizations of surface and boundary layer processes, and (2) parameterizations of boundary layer venting by convective clouds. Surface and boundary layer processes represent important components of the budgets of many trace species. For example, vertical fluxes of species such as HNO_3 are critical in estimating dry deposition of acidic material to surfaces. Emissions of natural hydrocarbons from vegetation are important in determining the amount of ozone production in some regions. The methods of determining the top of the mixed layer and its diurnal variation in the model have critical chemical implications because the depth of the mixed layer determines the initial volume into which pollutant gases and aerosols are mixed. Similarly, convective motions rapidly redistribute heat, momentum, moisture and trace chemicals in conditionally unstable areas. In addition, precipitation formed by condensation and coalescence removes water substance from the atmosphere, and latent heat resulting from this removal warms the atmospheric column. These mixing and condensation processes are initiated by nonhydrostatic, buoyancy-induced, cloud-scale dynamics. Any numerical models employing a horizontal resolution greater than ~10 km cannot resolve these processes. As a result, larger-scale models of atmospheric processes must parameterize these processes based on some assumed relationships between the convective-scale processes and resolvable processes.

Other significant uncertainties in coupled multiscale modeling associated with clouds include radiative effects, heterogeneous chemistry, and production of NO_x by lightning.

Perturbations of photolysis rates in and near clouds significantly alter the ozone production chemistry [Thompson 1984]. Some aqueous reaction schemes (e.g., Lelieveld and Crutzen 1990) show reduction of ozone production in clouds due to heterogeneous processes. Field observations are necessary to verify these theoretical calculations. NO_x production by lightning remains a large uncertainty because of the wide range of emissions per lightning flash that have been measured and because the number of intracloud and cloud-to-cloud flashes have generally not been counted.

- *Cloud models*

Cho et al. (1989) developed a coupled convective cloud model with gas and aqueous phase chemistry and fairly detailed microphysics, designed primarily for acid deposition studies. Chatfield and Delany (1990) developed a convective cloud/chemistry model that primarily simulates convective redistribution and fairly complete ozone photochemistry. Both of these models are essentially one-dimensional models designed for eventual incorporation into 3-D Eulerian transport/chemistry models.

Estimation of ozone formation in the free troposphere after redistribution of precursor gases by deep convection has been the objective of Pickering and coworkers at NASA/GSFC (Pickering et al. 1992c). These studies are based on running convective cloud and photochemical models in tandem. The detailed 2-D GCEM model (e.g., Tao et al. 1991) is run to simulate a particular deep convective event and wind fields generated by this model are used to advect and disperse the trace gases. Subsequently, particular profiles from the 2-D trace gas fields are used in a 1-D photochemical model (e.g., Thompson and Cicerone 1986) to estimate ozone production rates in cloud processed air. The largest uncertainties associated with this model stem from the treatment of the boundary layer, the representation of cloud microphysics and radiational characteristics, and the lack of interaction with meso- or larger-scale processes. For example, the only surface characteristics represented in the model are surface fluxes of heat and moisture. The photochemical model can either be run with photolysis rates for a clear sky or for the case of a single slab cloud, obviously an oversimplification of the real atmosphere.

2) *Chemical effects*

- *Ozone Production*

Deep convective clouds are a major means of transporting insoluble O_3 precursor gases (e.g. CO, NO_x, and hydrocarbons) from the boundary layer to the middle and upper troposphere [Dickerson et al. 1987; Luke et al. 1992]. These species, once detrained from a convective cloud, can react to produce O_3 in the free troposphere downwind from a convective system. (Note that O_3 in the upper troposphere is an effective greenhouse gas [Fishman et al 1979].) Because of higher winds and a longer photochemical lifetime than it has in the boundary layer, O_3 in the free troposphere may be transported large distances from the precursor source region [Pickering et al 1989]. More important, O_3 production in the boundary layer may actually be more efficient following dilution of polluted boundary layer air by deep convection when cleaner air descends in downdrafts. In this case the potential for O_3 production in the entire tropospheric column is enhanced. The major factors affecting the degree of enhancement of O_3 production by convection are the available boundary layer NO_x, strength and structure of the convective cells, presence of lightning-generated NO_x and the amount of background pollution in the free troposphere. An example of convective redistribution of NO_x and its consequences for O_3 production is given in Fig. 12 [Pickering et al 1992b]. The illustrations are all model-derived analyses of an episode from NASA/GTE/ABLE 2B, with pre-convective profiles of NO_x based on measurements. *There have been very few research flights with extensive sampling of cloud-outflow air to confirm such model predictions of convective transport and of O_3 production rates in cloud-processed air.*

Some fraction of nonprecipitating cumulus clouds also transport trace gases from the boundary layer to the free troposphere. Because these clouds do not typically occur in an organized weather system, their overall effectiveness for vertical transport is much more difficult to determine. Ching and Alkezwenny (1986) investigated the transport properties of a field of cumulus using SF_6 as a tracer and Ching et al. (1988) observed significant vertical exchange of ozone and aerosols between the mixed layer and the free troposphere during cumulus cloud activity using an airborne UV-DIAL system. Vukovich and Ching (1990) developed an empirical approach to estimate vertical transport by an ensemble of nonprecipitating convective clouds in a regional oxidant model. Very little verification data are available for this algorithm.

On a regional basis over a season, deep convection in the tropics may vent a significant fraction of CO, NO_x and hydrocarbon emissions from biomass burning to the free troposphere [Pickering et al 1992a]. It is clear that parameterizations of deep convection in 3-D global and regional chemical models need to capture chemical consequences of convective redistribution.

- *Aerosol-heterogeneous chemistry*

Over the eastern and central U. S. sulfates are a major, if not dominant, aerosol particle species. Water content and optical characteristics of clouds are crucially dependent upon the NH_4^+/SO_4^{--} molar ratio. Cumulus clouds are major chemical reactors where SO_2 gas is transformed into sulfate aerosol. Anecdotal data have suggested that the sulfate is not completely neutralized; thus, field studies are necessary to evaluate the level of neutralization of the aerosol and the amount of sulfate production in nonprecipitating cumulus clouds. These processes must be further elucidated in support of the development and validation of a Regional Particulate Model that will be adapted from RADM.

N94-24391

3.10 Validation of mesoscale models

Bill Kuo: Verification of mesoscale models

Tom Warner:	*Verification of cloud prediction from the PSU/NCAR mesoscale model*
Stan Benjamin:	*Results from MAPS/NGM verification comparisons and MAPS observation sensitivity tests to ACARS and profiler data*
Steve Koch:	*Systematic errors and mesoscale verification for a mesoscale model*
Andrew Staniforth:	*The COMPARE Project and the CME*

Bill Kuo opened the session by giving a review on the general methodologies used in the verification of mesoscale models. He then described the recent verification of an experimental mesoscale numerical weather prediction model during STORM-FEST, in which a 20-km version of the PSU/NCAR mesoscale model was used to provide experimental numerical guidance. His results showed that this model (which was not tuned prior to its use in support of STORM-FEST) gave a superior performance over the NMC Nested Grid Model (NGM). *This indicates that a mesoscale model which employs advanced physical parameterizations and more realistic topography has a strong potential to improve short-range local forecasting.* By verifying the model forecasts against the three-hour special rawinsondes and hourly profiler observations, Kuo was able to examine the model's systematic biases. He showed that the PSU/NCAR model has a wet bias in the humidity fields above 500 mb. By 36-h, the accumulated positive bias can be as high as 30%. He also noted that the model has a weaker diurnal variation in the surface-air temperature than that shown by the surface station observations. These results showed that *further improvement in model precipitation and planetary-boundary-layer parameterization is needed.*