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Project Summary

Tracer Studies of Transport and Transformation in Cumuli

J. L. Stith, A. J. Alkezweeny, and D. A. Burrows

Airborne measurements near Champaign, IL and Milwaukee, WI were made during the summers of 1990 and 1992 to study pollutant transport and transformation by clouds. Measurements of the aerosol size distributions, wind, turbulence, cloud microphysics parameters and trace gases were made from 31 research flights. During the 1990 study SF_6 was used as a tracer to determine cloud transport and entrainment.

In large clouds air from below the cloud bases was transported without dilution through the mid-levels of the clouds. On the other hand, in smaller clouds a more uniform dilution was observed as a result of outside air entrainment. The dilutions in the lower levels of the small clouds could be explained by a simple buoyancy sorting model.

An increase in the relative sizes of aerosol in the accumulation mode was observed in an area that was likely affected by the venting of cumuli in the area. Similar increases in size were not observed in evaporating regions of stratiform clouds. A hypothesis is proposed to explain the measurements. The cumuli activate much smaller aerosol which, after aqueous phase reactions and evaporation, have a much greater relative increase in size than the larger aerosol activated by stratus clouds.

The results from the entrainment experiments suggested that, during the early stages of entrainment, air from above the rising cloud is carried alongside the upper cloud region by the circulation present there. Later, the air is mixed into the main portion of the cloud and rapidly diluted with the cloud interior. The observations are consistent with the hypothesis that entrainment occurs through a vortex-like circulation that brings air from above the rising cloud top into the central region of the cloud.

The eddy correlation method was used to determine the transfer velocities of gases and aerosols over Lake Michigan downwind of Chicago. The results show downward transfer velocities (deposition) of 0.15 and 0.86 cm s^{-1} for 0_3 and aerosols in the size range of 0.1 to 3.0 µm in diameter and upward transfer velocities of 0.04 and 0.54 cm s^{-1} for CO_2 and water vapor about 7.5 km from the shoreline. At mid-lake much lower transfer velocities were measured. The turbulence intensity, in the subrange, was found to decrease as the air traveled over the cooler water.

This Project Summary was developed by the National Exposure Research Laboratory's Atmospheric Modeling Division, Research Triangle park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

Clouds cover about half of the earth's surface and occupy much of the troposphere. During their formation and dissipation, they interact in several ways with atmospheric pollution. Cumuliform type clouds (Cumuli) are especially effective at vertically redistributing pollutants. The largest variety, cumulonimbus, transport large amounts of polluted lower level air into the upper troposphere and bring cleaner mid- and upper-level air downward. Smaller cumuli play a similar role, except that they influence the vertical distribution of pollutants in the lower atmosphere. Although the effects from these smaller clouds are not as dramatic as those from the larger storms, their much greater number makes them very important in atmospheric chemistry. Both types of clouds transport pollutants away from the earth's surface, a major sink for most pollutants. Dry deposition is reduced, leading to long range transport of pollutants thus impacting further away receptors. On the other hand, precipitating clouds remove aerosols and trace gases from the troposphere.

Clouds also change the chemical and physical states of pollutants. For instance, the formation of acidic species by oxidation of SO_2 in clouds is much faster than in clear air. Clouds modify the size spectra of the atmospheric aerosol, changing its physical properties (e.g. light scattering, residence time, etc.). Pollutants that are lifted to higher elevations will experience higher light intensity which may alter their photochemistry.

Modeling the fate of pollutants on regional and larger scales is a major area of research for the US EPA. These models must properly account for the effects of clouds. For example, RADM, the Regional Acid Deposition Model, must account for cloud processes as well as gas-phase chemistry and deposition.

This report summarized the work that was done under a cooperative agreement between the US EPA and the University of North Dakota (UND). The objectives of the program were to better understand cloud processes (entrainment, cloud - aerosol interactions, etc.), and to collect measurements in support of the EPA regional modeling, especially the cloud module for the RADM and related models. During the period of the program the RADM and related code was extended by the EPA to address other concerns besides acid deposition (e.g. the fate of atmospheric aerosols). Consequently, the data from this program also support this broader effort.

Cloud and Aerosol Interactions

Measurements of in-cloud scavenging were made using the University of North Dakota Cessna Citation research aircraft on June 12, 1992, approximately 60 nautical miles southwest of Green Bay, WI. The aircraft was instrumented to measure several cloud physics and standard meteorological parameters. The cloud droplet size distributions were measured using the Particle Measuring System (PMS) forward-scattering spectrometer probe model 100 (FSSP). The FSSP is capable of measuring droplets in the diameter range of about 3-50 µm. Another PMSs probe, the passive cavity aerosol spectrometer probe model PCASP-100X, was used to measure the aerosol size distributions in the range from 0.1 to 3.0 µm. The probes complement each other and cover the range from 0.1 to 50 µm in a total of 30 channels. A 2DC PMS probe was used to determine ice crystal concentrations in the cloud.

The droplet concentration just above the cloud base is only about half of the aerosol concentration measured in the cloud inflow. This indicates that only about half of the aerosols act as cloud condensation nuclei active at the supersaturation of the cloud under study. To further investigate this problem we have examined aerosol size distributions from all sampled altitudes. The distribution in the cloud inflow (where the relative humidity was 96%) shows the normalized concentration decreasing with increasing aerosol diameter. However, within the cloud the size distributions are relatively flat. When compared to the inflow region, the in-cloud distributions show a marked increase in the concentrations of aerosols greater than about 0.4 mm. This means that a significant portion of the aerosols did not grow larger than 3.0 mm. The cloud droplet size distribution is slightly shifted toward larger droplets at the higher altitudes.

The results show that for cumulus clouds aerosol sizes are shifted toward slightly larger sizes after evaporation. On the other hand, no significant change in the aerosol sizes was observed after the aerosols were processed by stratiform clouds. We believe that this result is due primarily to the differences in cloud supersaturation in stratiform cloud droplets compared to droplets in cumuli.

Transfer Velocities of Gases and Aerosols Across the Lake Michigan Surface

On June 18, 1992 a constant altitude flight was made at 300 m above Lake Michigan near the Chicago shoreline and about 50 km downwind of it. The eddy correlation method was used to calculate the fluxes of CO_2, O_3, water vapor and aerosols in the diameter range of 0.1 to 3.0 µm. The fluxes near the shoreline were found to be significantly higher than those in the middle of the lake. The turbulence intensities, as measured by $e^{1/3}$, were 2.92 ± 0.75 cm$^{2/3}$s^{-1} and 1.34 ± 0.39 cm$^{2/3}$s^{-1} near the shoreline and mid-lake respectively. Fluxes measured near the shoreline were likely to be representative of those at the surface of the lake, because of the strong turbulence during the measurement. However, this is not true for the fluxes measured at mid-lake.

The fluxes near the shoreline for O_3 and aerosols were directed toward the surface and corresponding to transfer (deposition) velocities of 0.15 cm s^{-1} and 0.86 cm s^{-1}, respectively. For CO_2, and water vapor, the fluxes were directed upward and corresponding to transfer velocities of 0.04 cm s^{-1} and 0.54 cm s^{-1}, respectively.

A west-to-east constant altitude flight over the lake, starting from Chicago, showed that turbulence, as measured by $e^{1/3}$, decayed slowly along the flight track. The O_3 concentration steadily increased from 39 ppb to about 52 ppb as the air moved away from the shoreline.

Transport of Air by Convective Clouds

Large volumes of air from the mixed layer were lifted by a severe convective storm with very little mixing with air from the mid and lower levels of the free troposphere. Thus, these very strong storms may be best modeled with little or no entrainment in their lower levels. On a much smaller scale, the small clouds that were sampled contained a more uniform distribution of mixtures of air from below the cloud with air from near the level of entrainment. The upper portions of these clouds seem to be more dilute than their lower part, but this result may also be a function of the age of the cloud.

The techniques of buoyancy sorting are used in recent convective parameterization techniques. This approach was tested against observations and used to explain the behavior of a cloud base region that was tagged with a tracer. The results of the buoyancy sorting model are encouraging in that they offer a simple explanation for the behavior of the tagged region and they are able to explain the distribution of conserved parameters fairly well in the lower levels of the clouds that were studied, and less well in the upper regions. This may be improved when more information on the dynamics of the clouds are included. The results to-date suggest that a comparison of our results with the results from a more dynamic model—that also includes the buoyancy sorting method—should be conducted.

Entrainment

SF_6 tracer was released during single aircraft passes above the top of growing turrets associated with three different cumuli to study the entrainment of air by the cloud. The clouds ranged in size from a vigorous convective turret associated with a small thunderstorm, to a small, fair-weather cumulus.

The results suggest that, during the early stages of entrainment, the tracer remained mostly out of the cloud and was carried outward and down alongside the upper cloud regions by the circulation present there. In each experiment, concentrated SF_6 was first found on the edges of the cloud turrets. Later, the tracer mixed into the main portion of the turrets and rapidly diluted. The observations are consistent with the hypothesis that entrainment occurs through a vortex-like circulation that brings air from just above the rising cloud top into the central region of the cloud. Analysis of buoyancy considerations in cumuli suggest that much of the entrained air should remain relatively close to, or just slightly lower than, the altitude of entrainment. This helps explain the behavior of the tracer in the above study; a portion of the entrained tracer should be found just below the altitude where it was released after being entrained into the cloud. The tracer was indeed found in these locations (although it may have also been located in other parts of the cloud that were not sampled).

Jeffrey L. Stith, Abdul J. Alkezweeny, and D.A. Burrows are with the University of North Dakota, Dept. of Atmospheric Sciences, Grand Forks, ND 58201-9007.
***EPA Project Officers,** Dr. Jason K.S. Ching and Dr. Jonathan E. Pleim are on assignment to the National Exposure Research Laboratory (formerly Atmospheric Research and Exposure Assessment Laboratory, see below), from the National Oceanic and Atmospheric Administration.*
The complete report, entitled "Tracer Studies of Transport and Transformation in Cumuli," (Order No. PB95-255717; Cost: $19.50, subject to change) will be available only from:
National Technical Information Service
5285 Port Royal Road
Springfield, VA 22161
Telephone: 703-487-4650
The EPA Project Officer can be contacted at:
Atmospheric Modeling Division
National Exposure Research Laboratory
U.S. Environmental Protection Agency
Research Triangle Park, NC 27711

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