

Vertical Distribution of Pollutants in the
Presence of Convective Storms

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INTRODUCTION

Vertical profiles of trace gasses and aerosol particles were measured between the surface and 9.5 km during the PRECP (Processing of Emissions by Clouds and Precipitation) V field program conducted during June 1987 in the vicinity of Columbus, Ohio, USA. Measurements were made primarily in the vicinity of convective storms. Vertical motions occurring in these storms can transfer many pollutants from their source near ground level to the upper troposphere where they are more likely to travel long distances and influence the global environment (Gidel, 1983).

Observed vertical profiles are interpretable in terms of the pollutant source region and the vertical redistribution mechanism. Peak concentrations are observed at the altitude of peak emission or formation rate; near the surface except for H_2O_2 and sometimes O_3 which have mid-tropospheric and stratospheric sources, respectively. Vertical motions which transport air from the boundary layer to free troposphere are accompanied by cooling, condensation of cloud water and ultimately precipitation. Soluble species such as SO_2 (taking into account aqueous phase oxidation) and aerosol particles find themselves incorporated into cloud water during upward vertical motion. Conversion of cloud droplets into precipitation then limits the continued upward transport of dissolved pollutants in much the same way as the upward transport of water vapor is limited. There is no similar barrier to the upward transport of insoluble pollutants such as NO_y ($NO_x + PAN +$ organic nitrates) and CO and measurements in the upper troposphere show elevated concentrations due to storm venting.

RESULTS

Examples of vertical profiles generated from data collected during the PRECP V field program are presented here to illustrate the effects of 1) altitude of source region, 2) detrainment by convective clouds and 3) removal by precipitation scavenging. Data for these profiles were gathered from two instrumented aircraft used in the PRECP V experiments; the NOAA King Air (KA) and NCAR Sabreliner (SL). Figures 1a - 1e contain vertical profiles of CO, NO_y , O_3 , submicron aerosol particles and water vapor as measured from the SL on the days indicated in the figures. Figures 2a and 2b are vertical profiles of H_2O_2 and SO_2 measured from the KA. The vertical profiles do not include data collected while sampling in clouds or in precipitation.

The substances that are emitted or formed primarily or exclusively in the boundary layer (CO, NO_y , SO_2 , water vapor and aerosol particles) all have their highest concentrations in the boundary layer. The occurrence of these substances above the boundary layer is in general an indication of transport, though some contribution from in situ production is possible -

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for example, production of CO from oxidation of CH₄ and production of NO_x from lightning. Vertical transport due to convective storms was occurring during most of the PRECIP V flights. The occurrence of elevated concentrations of boundary layer pollutants at high altitudes is therefore interpreted in terms of the boundary layer composition during the sampling period. However, it is recognized that some fraction of the pollutants above the boundary layer were transported there at indeterminate times in the past.

High O₃ concentrations are observed in the boundary layer and/or upper troposphere. The boundary layer maximum is due to NO_x/HC photochemistry; the upper altitude source is transport from the stratosphere. O₃ at mid-altitude could be the result of transport from above or below. Under the relatively polluted conditions of the present experiments there is probably in situ generation of O₃ also occurring in the mid-troposphere.

The peak in H₂O₂ occurs above the boundary layer. This feature is observed on most days and shows up also in composite plots for the whole experimental period. Photochemical calculations identify the peak in H₂O₂ as occurring in a region of maximum H₂O₂ production - due to a relatively high concentration of H₂O and a relatively low concentration of NO_x (Kleinman and Leslie, 1988). The decrease in H₂O₂ at higher altitudes is attributed to a decrease in water vapor which causes a proportionate reduction in radical production from the short wavelength photolysis of O₃.

Elevated concentrations of CO and NO_x were often observed in the upper troposphere, at the same altitude, or slightly below the tops of convective clouds in the area. On 6/09 the concentration of CO at 290 mb was 1.5 times that observed in the mid troposphere. High concentrations of NO_y (> 1 ppb) were also encountered at 290 mb; background NO_y, however, was low (~ 0.1 ppb) and the resulting average vertical profile shows a nearly constant NO_y level above 700 mb. The C shaped vertical profile of CO in Fig. 1a is similar to that observed by Dickerson et al. (1987), which was attributed to venting from a thunderstorm. In the present experiments, the elevated concentrations of boundary layer pollutants usually occurred in patches, which could not be associated with an individual convective cloud. Rather, the patches appear to be a cumulative result of prior convection and venting.

In contrast to the insoluble pollutants CO and NO_y, the concentration of SO₂ and aerosol particles measured outside-of-cloud in the upper troposphere was always low. Figure 2b indicates that SO₂ decreases with respect to altitude up to the mid-troposphere. Measurements made from the SL at higher altitudes were not significantly different than zero. The number concentration of sub micron aerosol particles shown in Fig. 1d decreases by a factor of 200 at 290 mb as compared with 975 mb. The patches of boundary layer air encountered in the upper troposphere usually had increased numbers of particles. However, absolute concentrations remained low. While NO_y would increase to 0.5 - 1 ppb, a significant fraction of the boundary layer value, the number of particles might change from 10 cm⁻³ to 20 cm⁻³, resulting in an upper tropospheric concentration still two orders of magnitude lower than the boundary layer.

Figure 1d contains a vertical profile for water vapor obtained from dew point measurements made at the same time as the particle measurements. Similar data, averaged over 12 SL flights, are shown on a log scale in Fig. 3a. Figure 3b, which displays a vertical profile of the ratio of

particles to water, shows that there is a qualitative proportionality between these two substances. This proportionality is particularly noticeable in view of the 200 fold decrease in the individual concentrations between the surface and upper troposphere. The observed relation between particles and water can be explained as being a consequence of vertical transport of two species which both have boundary layer origins and which are both removed from the atmosphere via precipitation. Vertical motions which transport air from the boundary layer to free troposphere result in adiabatic cooling and the formation of clouds. A large fraction of aerosol particles is incorporated into cloud droplets as cloud condensation nuclei (Jensen and Charlson, 1984; ten Brink et al., 1987). The removal of cloud droplets in the form of precipitation then removes a roughly proportional number of aerosol particles. The proportionality between water and particles is maintained during microphysical transformations of cloud droplets (Flossmann et al., 1985) and during evaporation, which returns both pollutants and water vapor to the gas phase. These relations are consistent with the large scale behavior of particles and water vapor found in the present experiments.

SUMMARY

During the PRECP V field program vertical profiles of CO, NO_y, SO₂, O₃, H₂O₂, aerosol particles and water vapor were measured in the vicinity of convective storms or in air masses which had experienced convection in their recent past. Peak concentrations occur at the altitudes at which the various substances are emitted or formed in the atmosphere. For water vapor, SO₂, CO, NO_y and submicron aerosol particles this is the boundary layer. High concentrations of O₃ occur in the boundary layer where it is photochemically formed and in the upper troposphere as a result of downward transport from the stratosphere. The peak formation rate and the highest concentration of H₂O₂ occurs above the boundary layer.

The insoluble pollutants CO and NO_y can be efficiently transported from the boundary layer to upper troposphere. Soluble species such as SO₂ and aerosol particles cannot be similarly transported from the boundary layer to upper troposphere. The observed proportionality between aerosol particles and water vapor is consistent with limits on the upward transport imposed by the sequence of cooling, cloud formation, and removal in precipitation.

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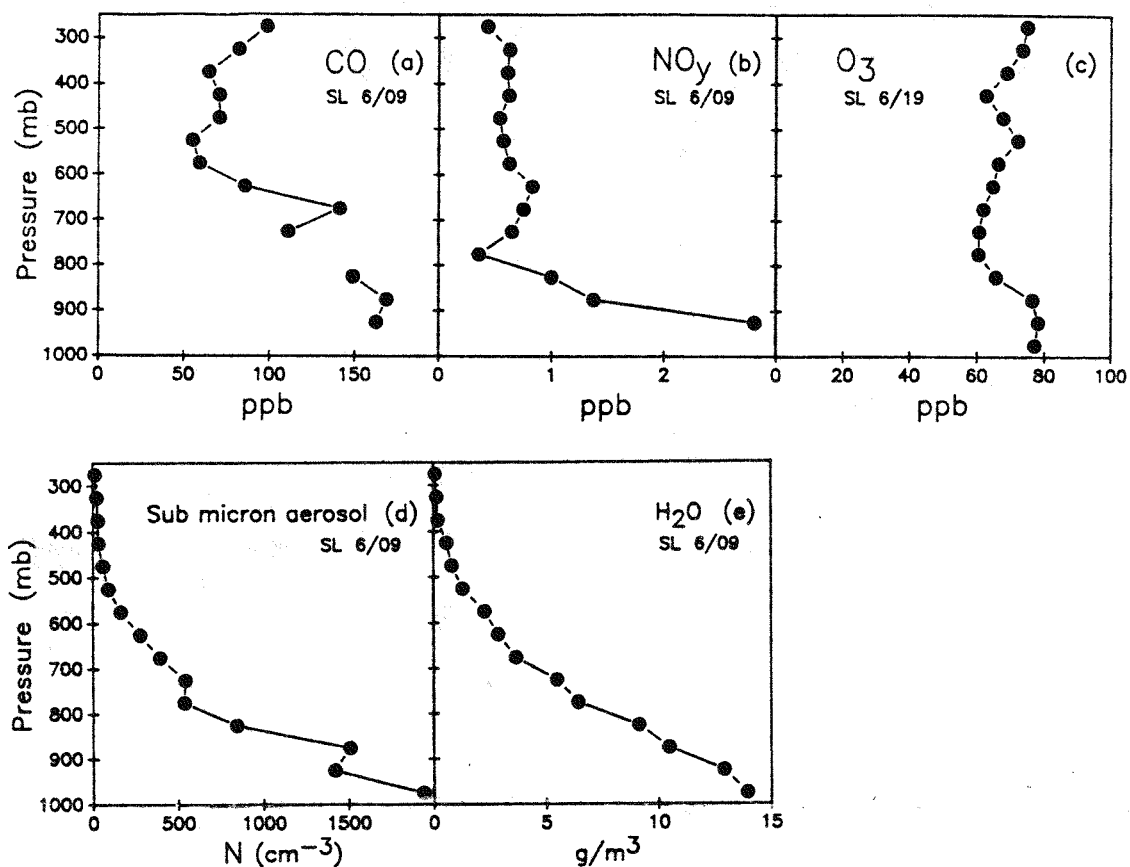


Figure 1. Vertical profiles of CO, NO_y, O₃, submicron aerosol particles and water vapor. O₃ not measured on the same day as the other four species.

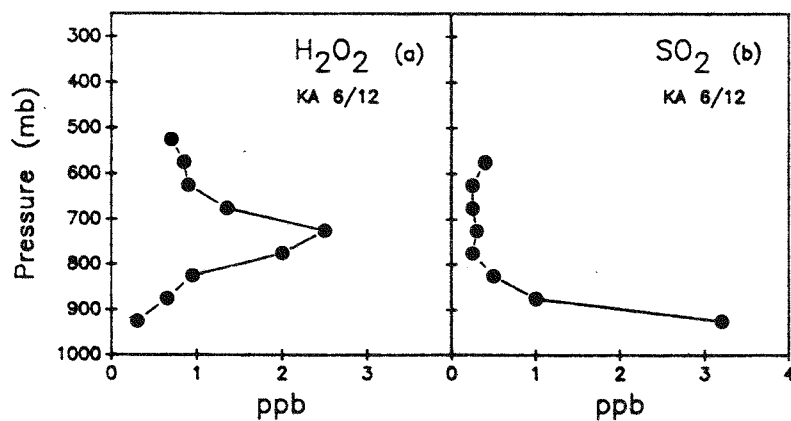


Figure 2. Vertical profiles of H₂O₂ and SO₂.

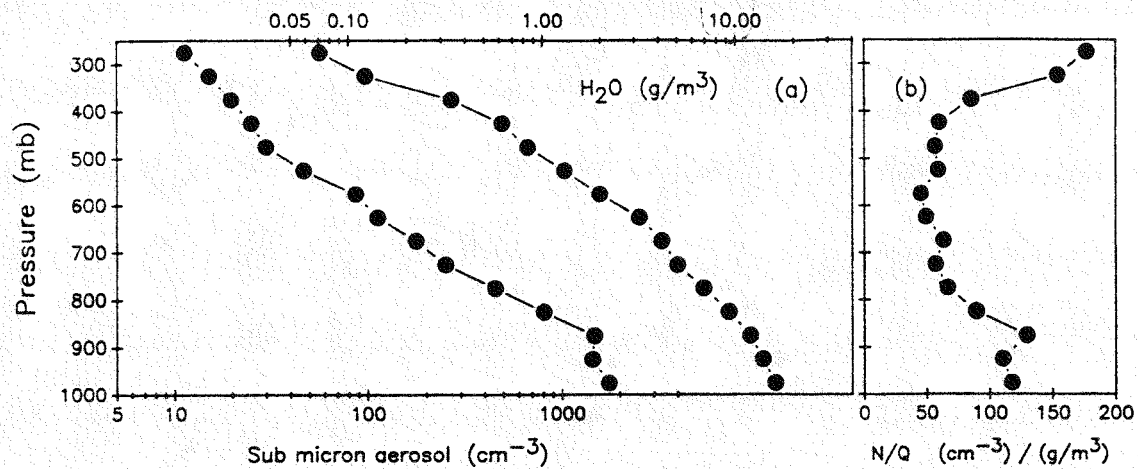


Figure 3. Composite vertical profiles for number concentration of submicron aerosol particles (N), water vapor concentration (Q), and the ratio N/Q ; from 12 flights of the SL.