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TURBULENT TRANSPORT AND PRODUCTION/DESTRUCTION OF OZONE IN A BOUNDARY LAYER OVER COMPLEX TERRAIN

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ABSTRACT

The first Intensive Observation Period (IOP) of the Swiss air pollution experiment POLLUMET took place in 1990 in the Aare River Valley between Bern and Zurich. During the IOP, fast response measurements of meteorological variables and ozone concentration were made within the boundary layer aboard a motorglider. In addition, mean values of meteorological variables and the concentrations of ozone and other trace species were measured using other aircraft, pilot balloons, tethersondes, and ground stations.

Turbulent flux profiles of latent and sensible heat and ozone are calculated from the fast response data. Terms in the ozone mean concentration budget (time rate of change of mean concentration, horizontal advection, and flux divergence) are calculated for stationary time periods both before and after the passage of a cold front. The source/sink term is calculated as a residual in the budget, and its sign and magnitude are related to the measured concentrations of reactive trace species within the boundary layer. Relationships between concentration ratios of trace species and ozone concentration are determined in order to understand the influence of complex terrain on the processes that produce and destroy ozone.

1. INTRODUCTION

POLLUMET is an ongoing air pollution and meteorology experiment in Switzerland that focuses on interdisciplinary studies of emissions, transport, and atmospheric chemistry in the boundary layer on distance scales ranging from regional to international and time scales ranging from hours to days (Neininger and Dommen, 1990). In the summer of 1990, aircraft of the Deutsche Forschungsanstalt fuer Luft- und Raumfahrt (DLR: German Aerospace Research Establishment) participated in the first Intensive Observation Period of POLLUMET, collecting data on boundary layer turbulence (motorglider) (Willeke et al., 1991), trace gas mean concentrations (Queen Air) (Paffrath et al., 1991), and winds and other meteorological parameters (King Air and pilot balloons) (Enderle et al., 1991; Hoelle et al., 1991).

2. FLUX MEASUREMENTS AND THE BUDGET OF MEAN OZONE CONCENTRATION

On three days in July 1990, the motorglider flew 20-km legs in a T-pattern at various altitudes within the boundary layer centered about 30 km northeast of Bern, Switzerland, in the vicinity of the Aare Valley. The complex terrain under the flight legs is shown in fig. 1. The motorglider flew at an air speed of 35 m s-1. Fast-response data were sampled at a rate of 10 Hz. The flight data analyzed in this paper were obtained between 1200 and 1530 LT (local time) (1000 and 1330 GMT), which corresponds approximately to the midday stationary time period in the evolution of the daytime boundary layer.
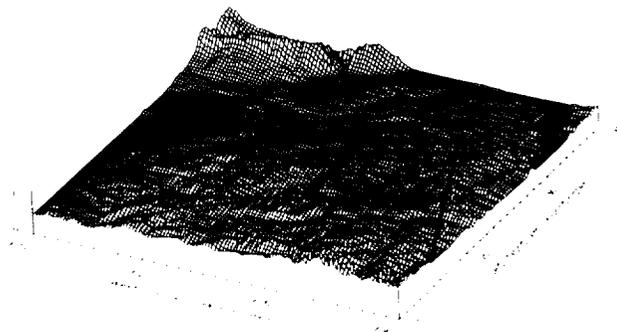

Fig.1. Terrain under the T-pattern flown by the motorglider during the Intensive Observation Period of POLLUMET 1990.

The profiles of potential temperature, specific humidity, ozone concentration, and the fluxes of sensible and latent heat and ozone are shown in figs. 2, 3 and 4 for 27, 28 and 30 July, respectively. There was only one motorglider flight on 29 July, when a cold front passed through the region, and the data from this flight are not included here.

The motorglider flew at the height of the inversion (z_i) on both 27 and 28 July, as evidenced by large, intermittent nonstationarities in the time series caused by sampling both above and below z_i. Although the data from the flights at these levels cannot be used for flux computations, the flight altitudes are denoted by z_i in figs. 2 and 3. The points in figs. 2, 3 and 4 are averages over five legs at each height (ten legs at the lowest level on 30 July) in the T-pattern, and the error bars are the standard deviations of the mean.

The fluxes of ozone were upward (positive) at a height of approximately 0.5 z_i on 27 and 28 July. More typical ozone fluxes were obtained on 30 July after the passage of the cold front, with downward (negative) values observed throughout the boundary layer, decreasing in magnitude up to the height of the inversion, which was at approximately 1300 m MSL (above mean sea level) on this day. (It was substantially higher on 27 and 28 July.) The average height of the terrain under the T-pattern was approximately 700 m MSL.

The budget of mean ozone concentration, neglecting horizontal flux divergence, is given by (Lenschow et al., 1981)

$$\frac{\partial c}{\partial t} = -\frac{\partial}{\partial z}(\overline{w'c'}) - V \cdot \nabla c + S$$

where the term on the left is the local time rate of change of ozone concentration, and the terms on the right are vertical flux divergence, horizontal advection, and boundary layer source/sink, respectively. Data from the motorglider, Queen Air, King Air, and pilot balloons were used to calculate the terms in the ozone concentration budget, except the source/sink term, which was obtained as a residual. The results are given in Table 1.

Although there are large uncertainties in the values of the source/sink terms in Table 1, the chemical processes in the boundary layer appear to change from a sink of ozone before the cold front passage on 29 July to a source of ozone afterward. The change in sign of the source/sink term is governed mainly by the change in sign of the flux divergence term, which is directly related to the change in direction of the mid-boundary layer ozone fluxes.

Along with the passage of a cold front, it is possible that the change in the sign of the source/sink term is
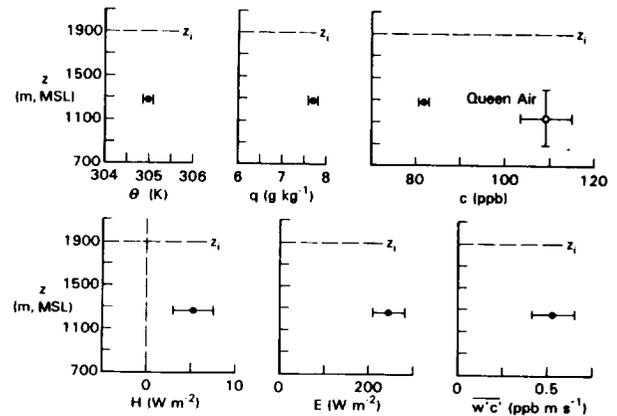

Fig. 2. Potential temperature (theta), specific humidity (q), ozone concentration (c), and the fluxes of sensible heat (H), latent heat (E) and ozone (w'c') measured aboard the motorglider and Queen Air on 27 July 1990.
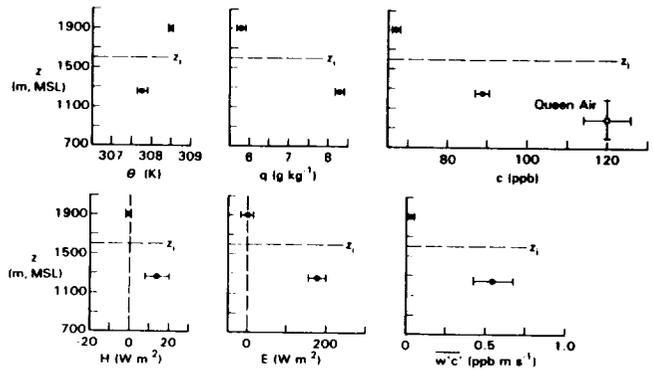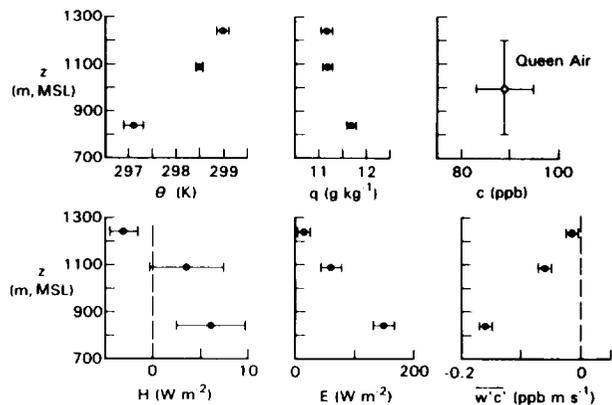

Fig. 3. Same as fig. 2 for 28 July 1990.

Fig. 4. Same as fig. 2 for 30 July 1990.

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associated with the timing of the flights: the midpoint of the flight on 30 July was 3 hours earlier than the midpoints of the flights on 27 and 28 July.

From the data taken aboard the Queen Air (discussed below), the only large change that occurred in mean meteorological variables and trace gas concentrations after the passage of the cold front was in average relative humidity, which increased by about 35% in the morning and 24% in the afternoon from before to after the passage of the front. The average pre- to post-frontal temperature change was approximately -3 deg. C in both the morning and afternoon; the average pre- to post-frontal change in ozone concentration was -4 ppb (parts per billion) in the morning (93 to 89 ppb) and -17 ppb in the afternoon (114 to 97 ppb).

Thus, in terms of mean quantities, conditions in the boundary layer were not changed very much by the frontal passage. The change in the boundary layer from being a sink of ozone to being a source, and the related change in the sign of the flux divergence, may therefore be at least partly due to the earlier time in the day for the flight of the motorglider after the frontal passage.

3. DAYTIME VARIATIONS OF METEOROLOGICAL VARIABLES AND CONCENTRATIONS OF CHEMICAL TRACE SPECIES

On 27, 28, and 30 July, the Queen Air flew at two levels within the boundary layer (at approximately 90 m and 550 m above the surface) in the morning and in the afternoon. The mean quantities measured during each flight were averaged over the three days. The results are shown in Table 2.

The distribution of meteorological quantities and concentrations of chemical species within the boundary layer

	27 July 1990	28 July 1990	30 July 1990
$\dfrac{\partial c}{\partial t}$	-2.7 ± 2.7	3.4 ± 3.4	1.9 ± 1.1
$\dfrac{\partial \overline{(w'c')}}{\partial z}$	-3.1 ± 0.7	-5.8 ± 1.4	1.3 ± 0.3
$V \cdot \nabla c$	-1.7 ± 0.7	-2.5 ± 3.0	-2.2 ± 1.2
S	-7.5 ± 4.1	-4.9 ± 7.8	1.0 ± 2.6
local flight time (midpoint)	1530	1530	1230

Table 1. Terms in the ozone mean concentration budget calculated from data obtained aboard the motorglider, Queen Air, and King Air, and from pilot balloon soundings. The source/sink term (S) is calculated as a residual. Units are ppb h-1.

can be inferred from the averaged Queen Air data in Table 2. Potential temperature, specific humidity, and the concentration of ozone were well-mixed in the boundary layer in both the morning and the afternoon. Temperature and ozone concentration increased and relative humidity decreased from morning to afternoon. Visibility decreased with height in the morning, but was approximately uniform in the boundary layer in the afternoon.

The profiles of the concentrations of NO2, NO, and SO2 had large vertical gradients within the boundary layer in the morning. The concentrations of NO2, NO, and SO2 at the upper level in the boundary layer remained relatively small from morning to afternoon. However, there was a marked decrease in the concentrations of these species at the lower level in the afternoon, resulting in a decrease in the vertical gradients.

4. PRODUCTION/DESTRUCTION RELATIONSHIPS BETWEEN TRACE SPECIES

A comparison was made between the coincident variations in the concentrations of ozone, NO2, NO, and SO2, and visibility over all 11 flights of the Queen Air, and also over the 5 low-level flights, on 27, 28, and 30 July. The resulting correlation coefficients are given in Table 3.

Although the number of flights is small, six correlation coefficients in Table 3 are significant at or above the 95% level. Ozone concentration was negatively correlated with the concentrations of NO2, NO, and SO2 and with visibility, with larger correlation coefficients occurring at the lower level in the boundary layer. The correlation coefficients between the concentrations of NO2, NO, and SO2 and visibility were all positive, with the strongest correlations occurring between NO2 and SO2, and between NO2 and visibility.

The correlation coefficients in Table 3 indicate that the production (destruction) of ozone in the boundary layer

	local time (midpoint)	altitude (m, ASL)	T (°C)	RH (%)	O₃ (ppb)	NO₂ (ppb)	NO (ppb)	SO₂ (ppb)	visibility (10⁻⁴ m)
morning lower level	1156	750	20	58	84	6.3	1.6	2.6	1.3
morning upper level	1141	1230	18	57	97	1.3	0.1	0.8	0.8
afternoon lower level	1653	830	25	43	111	2.8	0.7	1.3	0.9
afternoon upper level	1653	1270	21	46	106	1.4	0.4	0.4	1.0

Table 2. Mean meteorological variables and mean concentrations from flights of the Queen Air averaged over 27, 28, and 30 July 1990.

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was directly related to the destruction (production) of NO2, NO, and SO2. High concentrations of the latter three chemical species occurred low in the boundary layer (approximately 90 m above the surface) in the late morning (at about 1150 LT; see Table 2). These concentrations were reduced by a factor of 2 to 3, with a corresponding increase in ozone concentration by about 30% by mid-afternoon (at about 1650 LT).

At the upper level (550 m above the surface), the production/destruction relationships between ozone and NO2, NO, and SO2 are not as clear. From Table 2, between late morning and mid-afternoon, the ozone concentration at the upper level increased slightly (by about 10%), while the NO2 concentration remained constant, the (small) NO concentration increased by a factor of 4, and the (small) SO2 concentration decreased by a factor of 2.

5. DISCUSSION

Jumps in the magnitude of potential temperature, specific humidity, and ozone concentration indicate the presence of well-developed capping inversions at the top of the boundary layer on each observation day. For 27 and 28 July, these jumps were determined from the large, intermittent non-stationarities in the time series obtained during the flights of the motorglider at the height of the inversion (z_i), and from the jumps in the profiles of mean quantities across the inversion (28 July only, fig.3).

Representative changes in mean quantities from below to above the inversion were, for potential temperature, +0.4 deg C and +0.8 deg C; specific humidity, -2.4 g kg-1 and -3.1 g kg-1; and ozone concentration, -16 ppb and -30 ppb, for 27 and 28 July, respectively. A temperature jump similar

	O_3	NO_2	NO	SO_2	visibility
O_3 :	—	-0.49	-0.61	-0.44	-0.51
NO_2 :	-0.76	—	0.68	0.88	0.70
NO :	-0.72	0.35	—	0.46	0.56
SO_2 :	-0.61	0.92	-0.08	—	0.44
visibility :	-0.94	0.88	0.74	0.68	—

Table 3. Correlation coefficients for variations of mean concentrations and visibility from data obtained during flights of the DLR Queen Air within the boundary layer on 27, 28, and 30 July 1990. The coefficients above the diagonal are for all 11 flights; those below the diagonal are for the 5 low level flights (approximately 90 m above the surface) only. Highlighted correlation coefficients are significant at or above the 95% level.

to those on 27 and 28 July was seen in profile data from the King Air obtained on 30 July. (No other inversion jump data are available for this day.)

The presence of a well-defined capping inversion on each day, combined with the presence of anthropogenic sources and sinks at the surface and the complex terrain associated with the Aare Valley (fig. 1), tended to trap pollutants in the experimental region. Interactions between these pollutants gave rise to the production/destruction relationships between ozone and NO2, NO, and SO2 discussed in the previous section.

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