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BOUNDARY LAYER STRUCTURE AND PHOTOCHEMICAL POLLUTION IN THE HARZ MOUNTAINS—AN OBSERVATIONAL STUDY

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Abstract—Results from a field campaign that has been performed in summer 1993 to study photochemical pollution in the Harz Mountains, Germany, are presented. During a five-day, fair-weather period, a steady increase of the daily maximum ozone concentration up to values around 100 ppb was observed in the Harz region. The results of ozone measurements at two surface stations (404 m a.s.l. and 1142 m a.s.l., respectively) are discussed with respect to both the transport and dispersion conditions during the period and the local structure of the atmospheric boundary layer. Remarkable similarities in the time series of the ozone mixing ratio at the two places have been found which indicate some kind of quasi-homogeneity even over complex terrain. The trace gas concentrations at the top of Mt. Brocken, the highest summit of the Harz Mountains, are shown to be strongly influenced by vertical transports due to convection (at daytime) and subsidence of inversion layers (at nighttime).

Key word index: Photosmog, ozone, mountain meteorology, atmospheric boundary layer, vertical transports.

1. INTRODUCTION

Studies of photochemical pollution have been of increasing interest in many countries during the last few years (e.g. Güsten *et al.*, 1988; Spanton and Williams, 1988; Wakamatsu *et al.*, 1990; Lutz, 1994; Stohl and Kromp-Kolb, 1994; Hanna and Chang, 1995) because of the negative effect which high concentrations of photooxidants exert on human health and plant growth. The occurrence of photochemical pollution is favoured by strong emissions of the precursor substances, intensive insolation and a stagnant weather situation. The most common indicator of the so-called photosmog is the concentration of ozone which is formed in the lower atmosphere by photochemical reactions from precursors like nitrogen oxides and volatile organic compounds (VOCs). On a regional scale, the highest ozone concentrations have often been found some tens or even hundreds of kilometres away from large centres of industry and traffic (e.g. Bower *et al.*, 1994; Fabian *et al.*, 1994).

Whereas the photosmog problem has been recognized in the highly industrialized countries of Western Europe and North America for a longer time, attention in the Eastern European countries (former G.D.R., Czecho-Slovakia, Poland) was basically focused on the winter smog which is characterized by high concentrations of sulphur dioxide and soot. However, aircraft measurements in 1990 (Schaller *et al.*, 1992) revealed surprisingly high concentrations of ozone in the atmospheric boundary layer (ABL) over Eastern Germany. It had thus been argued that the problem of summer smog already existed in the former G.D.R., but had not attracted as much attention due to the more severe winter smog problem and also due to the absence of relevant measurements.

In 1990, the SANA programme has been launched in order to study the changing immission situation over Eastern Germany as it had been expected to result from the reconstruction of industry, energy production, and traffic that occurred since that time, as well as the consequences from those changes for highly sensitive ecosystems (Schaller and Seiler, 1993). The SANA programme includes both long-term monitoring of wet and dry deposition within special networks and field experiments dealing with detailed process studies under certain weather situations. Experimental activities of the SANA programme focus on the central southern part of the former G.D.R. around the cities of Halle, Leipzig, and Bitterfeld, one of the major industrial regions (Fig. 1). Related to the SANA-project, a field experiment has been carried out

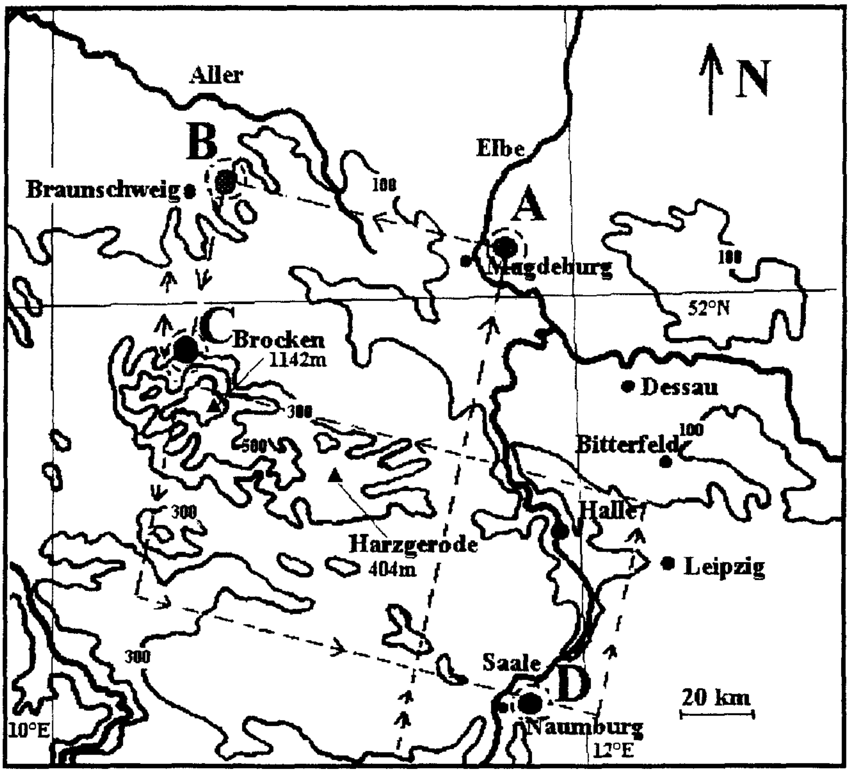

Fig. 1. Schematic map of the experimental region around the Harz Mountains and of the aircraft flight pattern (dashed line: level flight at about 150 m above ground, filled circles A–D: spiral profiling).

in the more rural areas west of the Halle–Leipzig–Bitterfeld triangle in June and July, 1993. This experiment was aimed at studying the occurrence and intensity of photosmog in the federal state of Sachsen-Anhalt and in the Harz Mountains. It was the first regional-scale study on photochemical air pollution over Eastern Germany apart from the major industrial centres. The study is of particular social relevance, since the Harz serves as a recreation area and partially has been declared as a natural reservation.

A general description of the field experiment, including a discussion of the regional distribution of pollutants over Sachsen-Anhalt, and an assessment of the regional contribution to the ozone production based on numerical simulations with a photochemical box model, will be published elsewhere (Möller *et al.*, 1995). The discussion in this paper is directed to the interpretation of the observed ozone concentration in the Harz Mountains with special focus on the structure of the ABL and on the role of vertical mixing and transport processes.

2. DESCRIPTION OF THE EXPERIMENT

2.1. *Site characterization*

The Harz Mountains area (see Fig. 1) is situated in a rural environment at the former borderline between West and East Germany, 50–100 km away from major industrial activities and most of the highways — except in the west, where highway E45 passes the foothills of the Harz at a distance of a few kilometres. In the Harz, ground-based measurements of standard meteorological parameters, trace gas concentrations, and ABL-structure were performed at two sites: at the top of Mt. Brocken and near to the little town of Harzgerode.

Mt. Brocken, the highest Harz mountain (1142 m a.s.l.), can be characterized as an isolated summit with steep slopes especially to the north (height difference of about 900 m over a distance of about 10 km). The top of the Brocken is a small plateau with an area of about 0.25 km^2. Due to its exposed position, the summit lies above the tree line and is covered by grass and scrub. Several tourist and administrative activities (restaurants, broadcasting station, terminus of a steam-powered, narrow-gauge railway) are concentrated at the Brocken plateau.

The Harzgerode station was temporally set up about 40 km to the southeast of the Brocken at an altitude of 404 m a.s.l. The terrain around this little town is characterized by moderate slopes directed from the WNW to the ESE. Harzgerode lies at roughly half the distance between the Brocken and the Halle-Leipzig industrial region.

2.2. *Description of measurements*

At the Brocken, the German Weather Service operates a meteorological station. In addition, a permanent field station has been installed there in 1991 basically to investigate the chemical processes within clouds (Möller *et al.*, 1993). At this station, continuous *in situ* concentration measurements of SO_2, O_3, NO_x, and H_2O_2 (using Thermo Env. Model 43 S, Environics S-300, Ecophysics and Aerolaser analyzers, respectively) have been performed since 1992 regularly between April and October. In addition, standard meteorological parameters are measured and cloud water sampling is performed. The data are recorded as 30-s averages. The field site at Harzgerode was equipped in a similar way — except measurements of H_2O_2 and using a Dasibi-analyzer for ozone measurement.

Doppler sodars were operated at both ground stations over the period of the experiment for continuous monitoring of the ABL-structure, turbulence intensity and winds. The sodars of type ECHO-1D have a technical height range of 50–800 m with a vertical resolution of 25 m. Single pulse data were recorded every 20 s.

Aircraft measurements using a Dornier-228 research aircraft have been performed on 30 June, and 1 July, 1993. The aircraft was instrumented for continuous measurements of meteorological parameters (pressure, temperature, relative humidity, wind) and concentrations of several trace gases. Ozone was measured with a Bendix analyzer (response time: 2 s, lower detection limit: 2 ppb, relative accuracy: 10%). Two flights were carried out on both days, one in the morning (between 07 CET and 10 CET) and one in the afternoon (between 12 CET and 15 CET). The flight pattern consisted of level-flights at about 150 m a.g.l. along the route indicated in Fig. 1 (for regional mapping) with spiral ascents and descents at the points A–D up to the lower free troposphere (for ABL-profiling).

3. METEOROLOGICAL SITUATION AND DISPERSION CONDITIONS

3.1. *Synoptic situation*

The weather conditions over Central Europe during summer 1993 were rather non-typical since only a very few fair-weather periods favourable for the formation and accumulation of photochemical pollutants within the ABL had been observed. One of these periods occurred between 29 June and 3 July.

The synoptic situation over Northern Germany during that time was characterized by dominating anticyclonic weather conditions. At the beginning of the period, an upper-air ridge was situated above the North Sea causing advection of cool air masses into the northern part of Germany (29 and 30 June). Later, the upper-air anticyclone moved towards the southeast, crossing Northern Germany thereby weakening, and finally it disappeared. Under the influence of weak pressure gradients at the surface, the polar air masses became stagnant and warmed up (1 and 2 July). At the end of the period, the north-atlantic frontal zone reached the European Continent from the north-west. At the surface the anticyclonic influence remained dominant caused by a large high pressure system over southern Central Europe. Strongly weakened fronts crossed the experimental area in the evening of 30 June, and on 3 July shortly after midnight. These fronts were not associated with any significant weather or even with clouds, but they led to modifications of the character of the air masses and of the ABL-structure (see Section 4.2). On 4 July larger amounts of clouds formed in the boundary layer behind another cold front. However, due to the anticyclonic influence they disappeared during the day and in the afternoon strong insolation was observed again. An active cold front on 5 July associated with widespread rainfall terminated the fair-weather period.

3.2. *Wind field and turbulence intensity*

The temporal evolution of wind speed and wind direction at both field sites derived from the Doppler–Sodar measurements for the lowest available range gate (50 m at Harzgerode, 75 m at Mt. Brocken) is shown in Fig. 2(a) and (b). In general, the wind behaved in a similar way at both places. The wind speed at Mt. Brocken was significantly larger than in Harzgerode due to the exposed location of the mountain station. The overall winds were weak and mainly came from easterly directions on 29 June. On 30 June, the wind blew from around north and was somewhat increased in speed. On 1 and 2 July, weak easterly winds were observed again, at times veering to south (or even southwest — in Harzgerode) on 2 July. Passage of the weak front around midnight on 3 July caused the wind to turn to westerly directions for the last two days of the period, thereby increasing in strength.

The time series of the vertical velocity variance (σ_w^2) derived from the sodar data are presented in Fig. 2(c). σ_w^2 reflects the intensity of vertical wind fluctuations and is therefore a suitable parameter to characterize the intensity of turbulent vertical exchange processes. The curves for both sites exhibit a pronounced diurnal cycle during all six days with small values during the nights ($\sigma_w^2 \leqslant 0.2\ \mathrm{m^2\,s^{-2}}$) and maximum values at daytime ($\sigma_w^2 \geqslant 0.8\ \mathrm{m^2\,s^{-2}}$), which is typical for fair-weather conditions.

3.3. *Mixing height*

A composite picture of the mixing layer height (MLH) evolution during the six-day period has been constructed mainly based on the sodar observations at Harzgerode and at Mt. Brocken, but considering information from the aircraft vertical soundings, too (Fig. 2(d)). For comparison, the inversion base height in the temperature profiles of the nearest routine radiosonde station at Hannover (about 150 km to the NNW from the Harz) is plotted (data from Deutscher Wetterdienst, 1993).

The diurnal MLH-evolution shows the typical picture to be expected under clear-sky, anticyclonic conditions. A shallow nocturnal boundary layer was observed during all nights at Harzgerode. Its depth was estimated to be between 80 and 150 m for the undisturbed nights (29 and 30 June, 1 and 2 July) and somewhat higher (150–250 m) for the nights, when the weak fronts crossed the experimental area. The breakup of the nocturnal inversion and the onset of intensive vertical mixing at Harzgerode occurred between 06 CET and 08 CET. It was followed by a rapid mixed layer growth. The MLH exceeded the sodar probing range (800 m) between 08 CET and 10 CET on most days (except on 1 July). A MLH of more than 800 m above Harzgerode implies growth of the mixing layer beyond the top of Mt. Brocken, as it can be also inferred from the sodar data at this location. For most days the data from both sites fit quite well to each

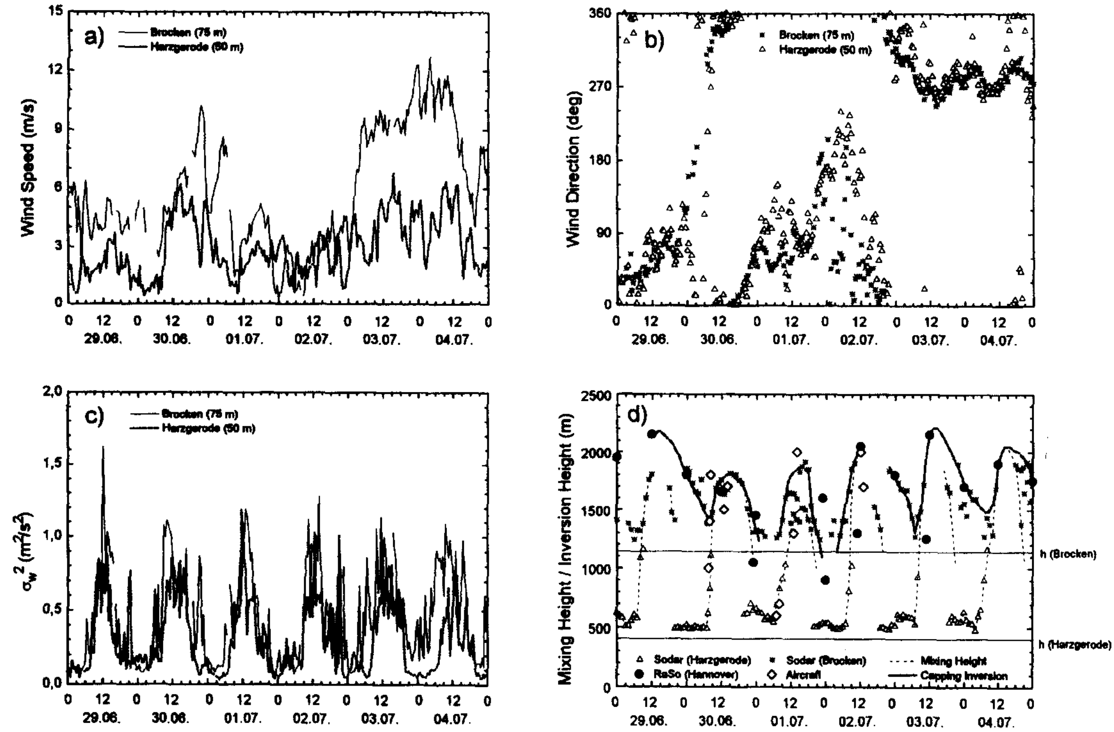

Fig. 2. Time evolution of meteorological parameters between 29 June and 4 July at Harzgerode (404 m a.s.l.) and at Mt. Brocken (1142 m a.s.l.) from sodar measurements: (a) wind speed, (b) wind direction, (c) vertical velocity variance, (d) mixing height (the dashed and solid lines are a "fit by hand" through the available data).

other. However, on 1 July and 2 July, the MLH observations from the top of Mt. Brocken and from Harzgerode are partially decoupled. On these days, the aircraft vertical soundings (compare with Section 4.2, Fig. 5) revealed a multi-layer internal structure of the ABL with different reservoir and inversion layers.

During nighttime, the Brocken was normally situated within the turbulence-free residual layer (remnant of the well-mixed convective boundary layer from the day before). During the nights between 30 June and 1 July, and between 1 July and 2 July, the elevated inversion, capping the ABL, subsided below the summit, which therefore became temporally placed within the free atmosphere.

Maximum MLH-values during the afternoon were between 1200 and 2200 m which can be considered typical for the region and for the season (cf. Gutsche and Lefebvre, 1981).

3.4. *Local convective boundary layer at the Brocken plateau*

The summit plateau of Mt. Brocken is normally situated in the upper ABL or even in the free troposphere due to the steep slopes at all of its edges. However, in case of weak winds and strong insolation, the plateau is obviously large enough to create a local, shallow convective boundary layer (CBL). The presence of a CBL can be inferred from the appearance of typical spiky structures in the time-height diagram of the backscattered signal intensity (sodargram) measured with the acoustic sounder (cf. Section 4.3 and Fig. 7b). Moreover, averaged signal intensity profiles have been checked with respect to the decrease of the backscatter signal (S) with height (z). For a well-developed CBL one should expect a decrease of S proportional to $z^{-4/3}$ (Wyngaard *et al.*, 1971). Averaged vertical profiles of S for some time periods of 1 July are shown in Fig. 3. The first one (08:30–09:00 CET) still corresponds to stable stratification at Mt. Brocken, and only a weak decrease of S with z is observed. After the onset of convection (09:00–10:30 CET), the decrease of S with height fits quite well the $z^{-4/3}$-law up to a height of about 300 m. Between 10:30 CET and 12:00 CET, when the mixed layer from the surrounding flatlands grew up to the height of the summit, the local CBL became destroyed. In the afternoon (14:30–16:30 CET) it formed again but it remained quite shallow due to the capping inversion at about 300 m above the Brocken plateau. The decrease according to the $z^{-4/3}$-law now extended up to about 175 m. Further evidence for the existence of a shallow local CBL comes from the time series of σ_w^2 (Fig. 2(c)) which shows a diurnal evolution that is typical for the CBL over flat homogeneous terrain.

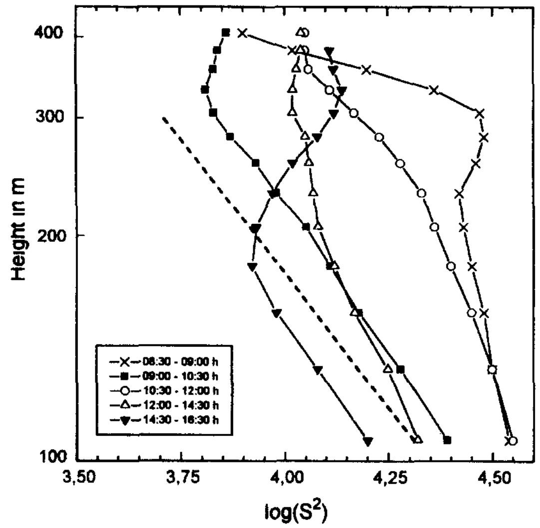

Fig. 3. Sound backscatter intensity profiles from observations with the acoustic sounder at the top of Mt. Brocken on 1 July compared to the $z^{-4/3}$-law (dashed line) valid for free convection.

4. TIME EVOLUTION OF THE OZONE MIXING RATIO

4.1. *General trend and diurnal cycle*

The time series of the near-surface ozone concentration measured at Harzgerode and at Mt. Brocken are shown in Fig. 4, and some key values are summarized in Table 1. At both sites, the daily maximum and the averaged O_3-concentration increased by roughly a factor of two between 29 June and 3 July. A pronounced diurnal cycle of the ozone concentration was observed at Harzgerode. In contrast, the data from the top of Mt. Brocken exhibit a general positive trend superimposed by variations which do not show a clear diurnal behaviour.

The daily maxima of the O_3-concentration are very close to each other at both sites (cf. Table 1). This is explained by the fact that on most days the maximum MLH was beyond the top of Mt. Brocken which was therefore in the afternoon situated within the well-mixed CBL, and similar concentration values like those at Harzgerode were observed. However, a time delay in the occurrence of the peak values can be noticed, especially for the days with easterly wind directions.

The highest ozone concentration values were observed at both sites on 3 July although insolation was reduced due to larger fields of Ci clouds in the afternoon. In addition, the MLH of that day was higher than the days before, and the regional ozone formation potential was presumably reduced due to the weekend (this is confirmed by the observed NO- and NO_2-concentrations at Harzgerode). Considering the change in wind direction between 2 and 3 July and the relatively high wind speed on the latter day, it can be assumed that these maxima in the ozone mixing ratio are basically due to long-range transport processes. Indeed, the computation of backward trajectories on isentropic surfaces§ has shown that the air masses arriving at Mt. Brocken on 3 July had been advected over a distance of 400–600 km from the west within 24 h. In contrast, the transport paths on 1 July and 2 July are only about 100 km long thereby showing a strong curvature thus limiting the possible source areas to the region around the Harz mountains (Naumann, 1995—personal communication).

The pronounced diurnal cycle of the ozone mixing ratio observed at Harzgerode is typical for flat-terrain locations. At daytime, intensive vertical mixing due to convection causes a rather uniform distribution of ozone and its precursors through the whole depth of the ABL. Ozone depletion (chemical destruction, dry deposition) during the night, however, is limited to the lowest few tens or hundreds of metres since the residual layer from the day before is decoupled from the near-surface layers due to stable stratification and a reduced level of turbulence. During the next morning, after the onset of convection and the break-up of the nocturnal surface inversion, intensive vertical exchange rapidly mixes down the pollutants from the residual layer to the surface (rapid increase of O_3-concentration at Harzgerode during the morning mixed layer growth period—cf. Figs 2(d) and 4). The initial concentration values from which photochemical ozone formation starts again are therefore higher than the day before. In this way ozone may accumulate within the ABL from day to day as observed.

4.2. *Frontal passages*

It has been mentioned earlier that weak fronts have passed the Harz region during the period of the experiment in the evening of 30 June, and around midnight from 2 July to 3 July (see triangles in Fig. 4). These fronts lead to a reconstruction of the ABL-structure and to a change in the chemical composition of the prevailing air mass.

On 30 June, the weak cold front brought some fresh and less polluted (polar) air to northern Germany. As a result, the minimum and maximum O_3-concentrations on 1 July at Harzgerode were lower than the day before (cf. Fig. 4). At Mt. Brocken, the frontal passage was associated with a jumplike decrease in the O_3-mixing ratio by about 15 ppb. The changes in the ABL-structure due to the front can be clearly seen from the aircraft profiles measured in the afternoon of 30 June and in the morning of 1 July (Fig. 5). Prior to the passage of the front, a simply structured ABL with a well-mixed layer extending up to a clearly-marked

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§The computation of the three-dimensional trajectories has been performed using the TRAP simulation package (Reimer and Scherer, 1991). The wind and potential temperature fields used for the analysis are based on an interpolation of the data from all standard radiosonde stations in Europe.

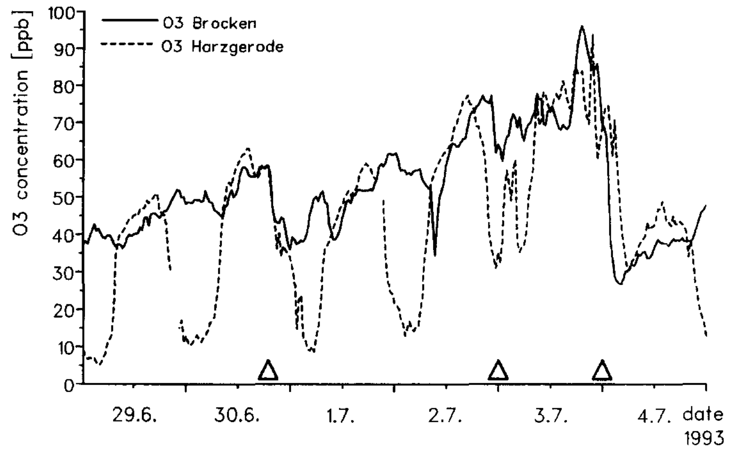

Fig. 4. Time series of near-surface ozone mixing ratio measured at Harzgerode and at Mt. Brocken between 29 June and 4 July 1993 (frontal passages are indicated by the open triangles).

Table 1. Key parameters characterizing the time evolution of ozone mixing ratio (in ppb) for the period 29 June to 4 July 1993

	Diurnal average		Minimum concentration (h)		Maximum concentration (h)	
Date	Brocken	Harzgerode	Brocken	Harzgerode	Brocken	Harzgerode
29.06	43	30	36 (07:35)	4 (05:55)	52 (21:55)	53 (16:55)
30.06	50	39	36 (23:35)	10 (00:35)	59 (18:25)	64 (14:05)
01.07	49	36	37 (01:25)	7 (05:05)	62 (23:35)	60 (18:05)
02.07	62	48	32 (09:00)	12 (02:25)	78 (22:55)	78 (16:35)
03.07	75	66	57 (00:45)	28 (04:55)	97 (19:05)	98 (21:45)
04.07	38	42	26 (04:05)	12 (23:55)	69 (00:15)	78 (01:05)

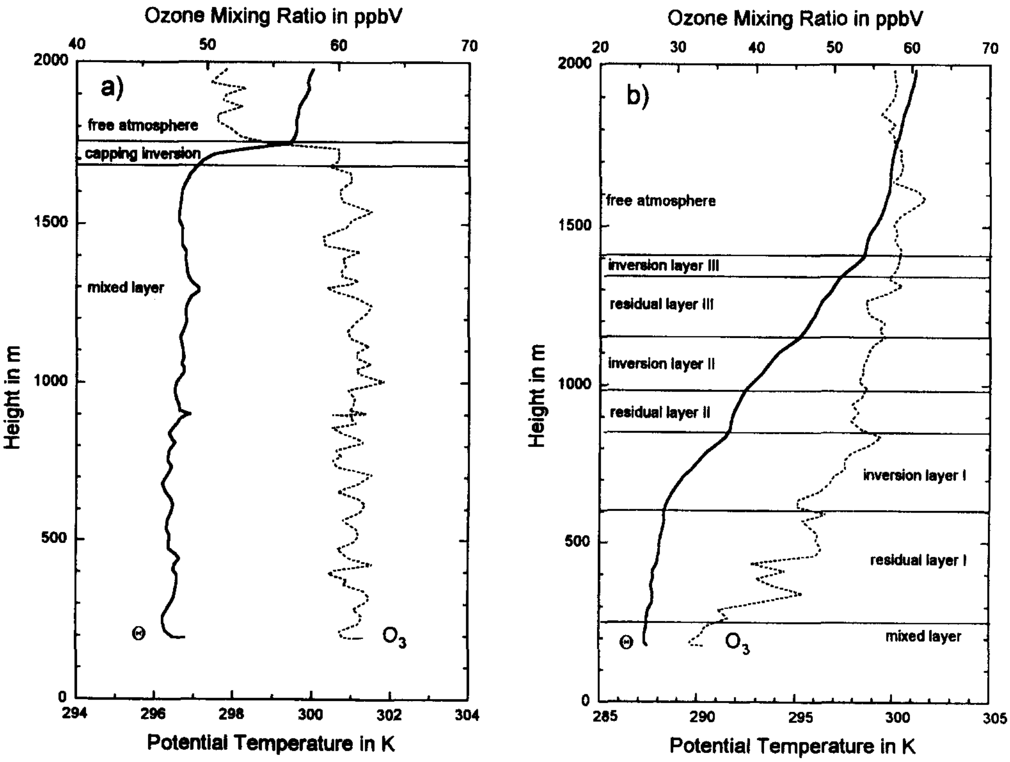

Fig. 5. Vertical profiles of temperature and ozone mixing ratio measured with the research aircraft on 30 June, and on 1 July near to Magdeburg.

capping inversion at about 1700 m a.s.l. was observed. On 1 July, in the morning, the ABL showed a rather complex structure with multiple inversion and mixing layers. The different layers are characterized by different values for the ozone mixing ratio representing different reservoir layers. Similar observations of reservoir layers have been described, e.g. by McElroy and Smith (1993).

During the night between 2 July and 3 July, the secondary ozone maximum at Harzgerode is attributed to the passage of the second weak front. In this case, vertical mixing at the front transported the ozone from the residual layer down to the surface. The mixing, associated with the frontal passage can be clearly seen from the sodargram recorded at Harzgerode, and from the time series of σ_w^2 during this night (Fig. 6). At Mt. Brocken, the passage of this front was associated again with a sudden decrease of the O_3-mixing ratio (cf. Fig. 4).

Passage of another cold front during the early morning of 4 July resulted in a rapid decrease of the ozone concentration levels at both sites due to the arrival of a new air-mass which was less polluted. At Mt. Brocken, the sudden decrease was much more sharp than at Harzgerode. This can probably be explained by the role of ozone depletion in clouds (e.g. Acker *et al.*, 1995) since the summit was within clouds for a few hours just behind the front.

4.3. *Vertical motions at the top of Mt. Brocken*

The observed time evolution of the ozone mixing ratio at Mt. Brocken is characterized by several jumplike changes superimposed on the generally increasing trend. The time series of SO_2, air temperature and humidity show similar steplike features during the period 30 June to 2 July (Fig. 7(a)).

The sodargram plot from Mt. Brocken for the time between 30 June, 16 CET, and 2 July 16 CET illustrating the local ABL-structure is presented in Fig. 7(b). The spiky, vertically oriented echo structures during daytime on 1 and 2 July correspond to convection (cf. Section 3.4). The horizontal echo layers visible especially in the evening of 30 June and 1 July are interpreted as inversion or stable layers with strong vertical temperature gradients and small-scale turbulence due to vertical wind shear. Vertical displacement of these layers is a dominant feature to be inferred from Fig. 7(b).

To support interpretation of these vertical motions, the time series of vertical velocity (w) measured with the sodar at Mt. Brocken is also shown schematically in Fig. 7(b). It should be noticed that the general accuracy of w-measurements with sodar is not assumed to be better than $0.2\,\mathrm{m\,s^{-1}}$. Besides, it is not trivial to correct w-measurements from the top of a mountain for terrain influences. The analysis of the w-time series presented in Fig. 7(b) is therefore only a qualitative one. However, it can be seen that during nighttime, when the downward displacements of echo layers have been observed, indeed downward vertical velocities were measured. These are attributed to large-scale subsidence under the anticyclonic weather situation. During daytime the plume structure of the convective ABL can be also inferred from the w-measurements. The morning rise of echo layers is related to the growth of the convective mixed layer.

Based on the most pronounced changes in trace gas concentrations and meteorological parameters (arabic numbers in Fig. 7), the period has been divided

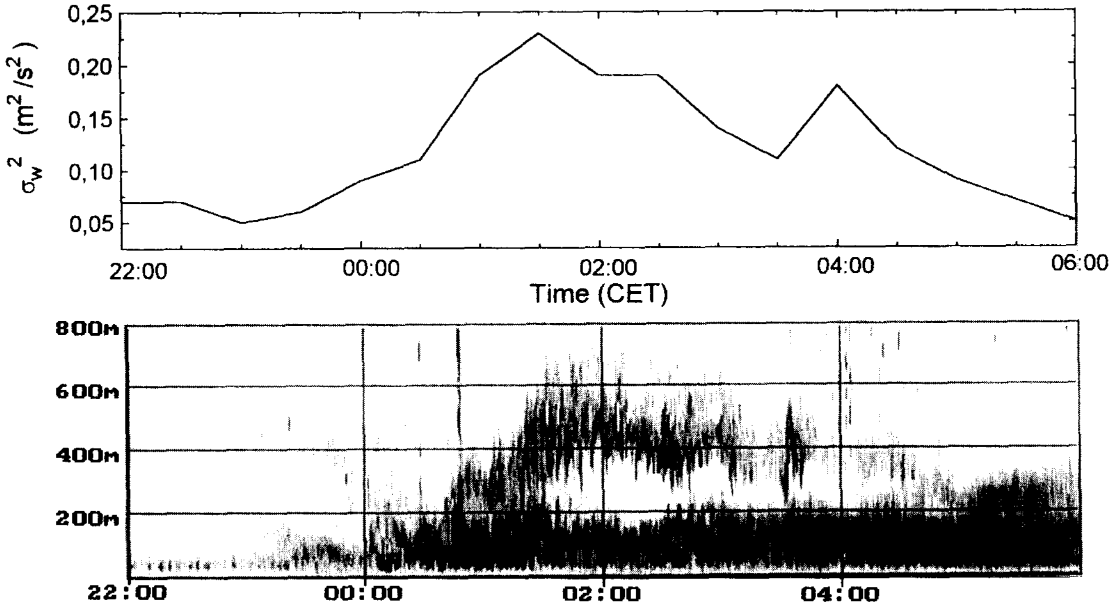

Fig. 6. Frontal mixing at Harzgerode during the night between 2 and 3 July as observed with the acoustic sounder: (a) sodargram, (b) time series of the vertical velocity variance.

into a number of phases (roman numbers in Fig. 7). These phases and events can be briefly characterized as follows.

On June, 30th, between 19 CET and 20 CET the weak front mentioned above passed the Brocken site (1). This could be clearly detected from the pressure trace (not shown here), and from the subsequent decrease in the O_3-mixing ratio and temperature, and the increase in humidity. At about 23 CET (2) the sodargram shows a horizontal echo layer arriving at the top of Mt. Brocken. The O_3-concentration decreased from 43–45 ppb to below 40 ppb. During the following hours (phase III), the Brocken station was situated within a turbulent layer with strong wind shear (the wind speed at the mast was higher than at the first sodar level). This turbulent layer moved down, leaving the sodar range between 03 CET and 04 CET (3). The summit now became situated within the capping free-tropospheric inversion (phase IV), as can be inferred from the temperature increase and from the drop in relative humidity down to values lower than 30%. Within this layer, ozone had been accumulated (jumplike increase by about 10–12 ppb), whereas SO_2 was nearly completely absent (mixing ratio below 0.05 ppb, the lowest values during the whole campaign).

After the onset of convection on 1 July, upward vertical motions became dominant (cf. Fig. 7(b)). At around 8.30 CET (4) the first mixed layer again reached the top of the mountain (phase V). This event is marked by a negative jump of the O_3-concentration down to the same level as during phase III at night, and a strong increase in humidity and SO_2 mixing ratio. Between 11 CET and 12 CET the flatland mixed layer grew up to the height of the Brocken plateau (strong positive w-values—cf. Fig. 7(b), (5)), followed by a slow rise in temperature, a decrease in relative humidity and a steady, nearly linear increase in the O_3-concentration for the next hours (phase VI). It should be noticed, that the growth rate of the ozone mixing ratio between 11 CET and 16 CET at Mt. Brocken (+ 11 ppb) was nearly the same as in Harzgerode (+ 10 ppb).

During the afternoon (phase VII), two layers can be seen in the sodargram. The O_3-mixing ratio and also most of the other meteorological and chemical parameters, which had been measured, remained nearly constant between 15 CET and 19 CET. The decrease of σ_w^2 after 16 CET (cf. Fig. 2(c)) indicates a decrease in the efficiency of vertical mixing during that time. The two layers visualized in the sodargram subsided again in the evening, and the top of Mt. Brocken became located in different layers between 19 CET and 20 CET (7), and also at around 22 CET (8). In each case, this was associated with a jumplike increase in O_3-concentration by 6–8 ppb (up to 57 ppb) and 3–5 ppb (up to 61 ppb), respectively (phases VIII, IX). The ozone mixing ratio in these layers thus was considerably higher than in the morning, but again, the upper layer was more polluted with ozone than the lower one. It seems noteworthy, that the SO_2-concentrations were rather high during this phase (up to 10 ppb). This might probably be attributed to the wind direction E to S, hence coming from the Halle–Leipzig industrial region.

Between midnight and 01 CET (9) continuing downward motions brought the Brocken into the free troposphere (as during the night before), characterized

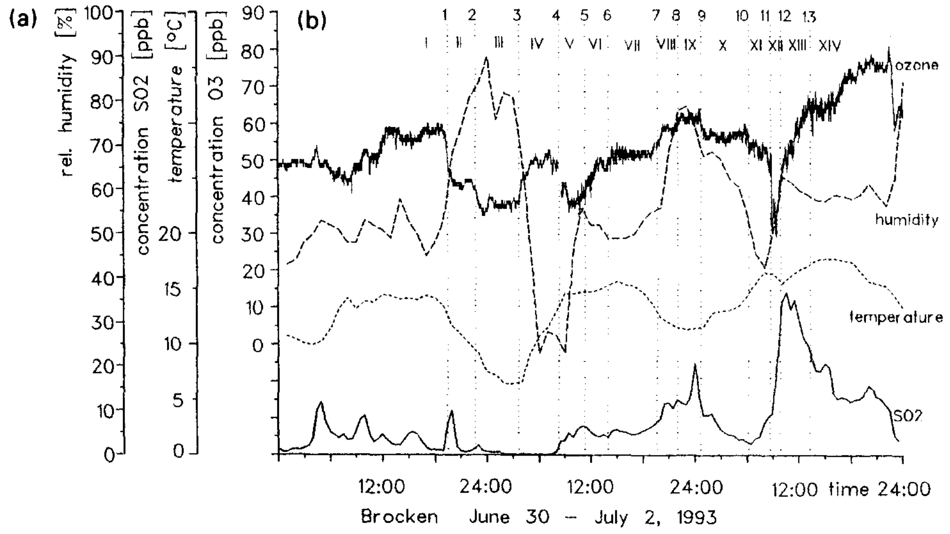

Fig. 7. Time series of meteorological and chemical parameters, and ABL structure observed at the top of Mt. Brocken between 30 June and 2 July 1993: (a) time series of temperature, relative humidity, and the O_3 and SO_2 mixing ratios; (b) sodargram plot and time series of the vertical velocity at 75 m a.g.l. for the time period between 30 June 1993, 16 CET, and 2 July 1993, 16 CET.

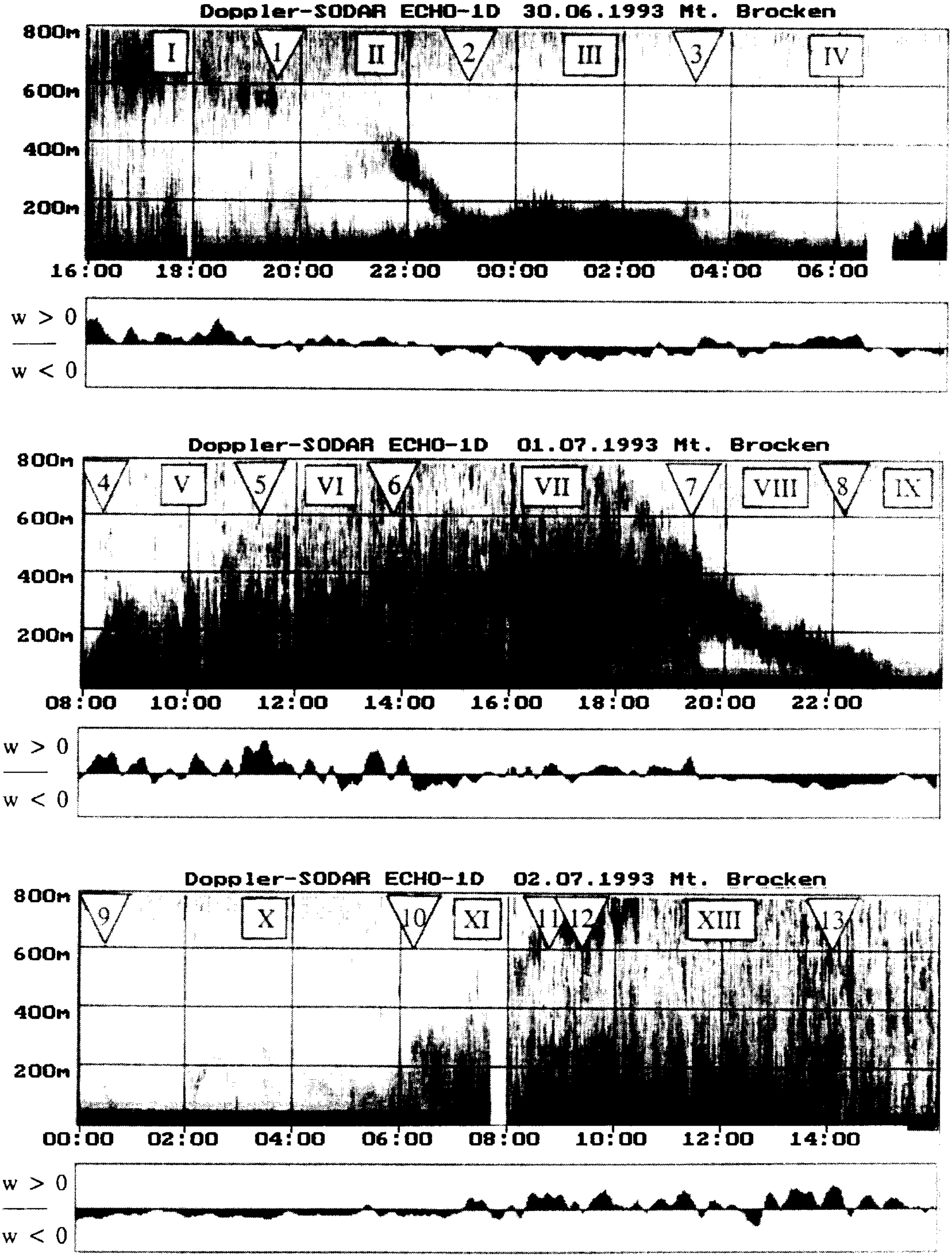

Fig. 7(b).

by a nocturnal rise in temperature and a corresponding decrease in humidity and SO_2 mixing ratio (phase X). In the morning of 2 July, the winds at the Brocken plateau were very light ($< 2\,m\,s^{-1}$) allowing the formation of a local CBL and rapid heating of the ground (phase XI). The temperature rise stopped at about 8.45 CET (11). For a short period of about half an hour duration (phase XII) the ozone mixing ratio dropped down drastically by nearly 20 ppb, at the same time a dramatic increase in NO_2 mixing ratio and relative humidity occurred. The reasons for this are not fully clear. Due to the very light winds they must have been of local origin.

Between 9.30 CET and 16 CET, the Brocken was situated within the well mixed CBL (phases XIII and XIV), as can be seen from the behaviour of temperature and humidity. Again the mixed layer winds were small (2–$4\,m\,s^{-1}$), and vertical mixing should have been as effective as during phase VI on the previous day. The growth rates of the ozone mixing ratio between 10 CET and 16 CET are again nearly the same at Harzgerode (+ 17 ppb) and at Mt. Brocken (+ 16 ppb).

5. SUMMARY AND FINAL DISCUSSION

A field experiment has been performed in summer 1993 in order to investigate the horizontal and vertical distribution of photooxidants in a mountainous region, namely the Harz Mountains in the federal state of Sachsen-Anhalt, Germany, which is a recreation and partially natural reservation area.

For that purpose, concentration measurements of several chemical constituents have been carried out during a five-day fair-weather period (29 June to 3 July) under anticyclonic conditions at two surface stations separated horizontally by about 40 km and with a difference in elevation above sea level of about 750 m. The structure of the ABL has been monitored with two Doppler sodars, and a research aircraft has been used for regional mapping and vertical profiling of meteorological and chemical parameters within the ABL.

The basic findings from this study can be summarized as follows:

(i) The daily maximum ozone concentration and the rise in the average ozone mixing ratio from day to day are similar at Harzgerode and at Mt. Brocken. This is attributed to the fact that the top of the convectively mixed ABL exceeded the top level of Mt. Brocken regularly during the afternoon. Obviously, under these conditions, some degree of horizontal homogeneity in the ABL-characteristics and the pollution level does exist even over complex, heterogeneous terrain. Similar observations but for less structured terrain and over a much shorter distance have been reported by Gay (1991).

(ii) The temporal evolution of the ozone concentration during the measurement period was characterized by a steady increase of the afternoon maximum mixing ratio, reaching values between 80 and 100 ppb on the last day. This seems remarkable in so far as the fair-weather period has covered only five days. Therefore it is evident that in the Harz area concentrations of photo-oxidants are observed which have to be considered in environmental policy, planning, and decision-making.

(iii) Studies of the ABL structure at the top of Mt. Brocken have shown that a relatively small summit plateau obviously may be large enough to create a shallow local convective ABL under conditions of strong insolation and light ambient winds.

(iv) Vertical displacements of different inversion and reservoir layers characterized by different photochemical equilibrium conditions are of considerable importance for the interpretation of the temporal evolution of the ozone concentration at the top of Mt. Brocken. The dominating role of vertical transports for the ozone concentration, especially at mountain stations, has been also stressed, e.g. by Wege and Vandersee (1991), and Dietze (1993).

(v) During nighttime, the Brocken summit was normally situated within the residual layer where only weak ozone depletion takes place. It has been shown by different authors (e.g. Feister and Balzer, 1991; Neu *et al.*, 1994), that the peak values of near-surface ozone mixing ratio on a given day strongly depend on the ozone concentration within the residual layer from the day before. Thus the background data measured at a mountain station like e.g. Mt. Brocken can probably contribute to the improvement of short-time forecasts of the maximum daytime ozone concentration during photosmog episodes even on a regional scale if the above-mentioned horizontal quasi-homogeneity exists. When using background concentration data from an elevated location for regional forecasting of air pollution, the possibility of a layered structure of the upper ABL as observed temporally during the experiment has to be carefully considered. The operation of sodars is very helpful to identify these structures.

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