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Lidar Studies of Tropospheric Transport

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Summary

The ozone and aerosol lidar systems of IMK-IFU (IFU) have been used within TOR 2 for tropospheric transport studies. The investigations have been based on measurements at the institute, during field campaigns and on model simulations provided by TOR-2 partners. An important topic has been transport in the Alpine wind system, such as the advection of urban pollution into the Alps, transport in and above Alpine valleys, and trans-Alpine ozone transport during Föhn. In particular, a significant contribution of the orographic wind system in the Alps to the pollution export from the boundary layer (PBL) to the free troposphere was verified. The measurements of the diurnal variation of the vertical aerosol distribution in the local Loisach valley and the Swiss Mesolcina valley showed that the main export from the PBL typically reaches heights about 1 to 1.5 km above the summit heights in the vicinity of the upper parts of the valleys under conditions of moderate humidity. The export efficiency may be as high as 80 %. Another important mechanism for lifting of polluted air from the PBL is upward transport by frontal systems. It was found from the analysis of the lidar sounding series that upward transport in warm conveyor belts may, in part, reach altitides even next to the tropopause and is a key step for long-range transport. Ozone sounding in May 1996 yielded the first clear evidence of intercontinental transport of North-American O_3. Subsequent studies have confirmed the characteristic peak ozone mixing ratios of 80 to 110 ppb in the middle and upper troposphere in air masses imported from North America. These values mostly agree with suface data in the United States or measurements in the outflow regions of the warm conveyor belts during MOZAIC flights. Simulations by TOR-2 partner have visualized the air flow and have identifed the most likely source regions. Numerous measurements were also carried out during stratospheric air intrusions. It was found that direct intrusions mostly reach 3000 m which underlines the significance of the long-term work at high-lying Alpine stations, in particular Zugspitze. The lidar measurements have yielded the vertically best resolved dense time series of subsiding stratospheric air tongues so far available. This has been an ideal basis for model validation and model intercomparision. It was found that the spatial resolution of the models is crucial for a satisfactory agreement with the measurements. Although the underlying ECMWF data allow one to catch most structures seen in the measurements the vertical resolution is obviously still limited. Finally, a climatology of the free-tropospheric aerosol was carried out. A pronounced spring maximum was found. Since long-range transport should be the principal source of aerosol above the PBL its role for the formation also of the ozone spring maximum must be taken into consideration.

Objectives

The main objective of this contribution has been to elucidate the factors which influence the vertical distribution of ozone above the Central European station Garmisch-Partenkirchen, in particular vertical and long-range transport. It had been proposed to contribute to the TOR-2 work packages on trends, vertical exchange and seasonal cycles. This goal was subsequently corrected when it became obvious that the proposal of routine measurements of free-tropospheric ozone in addition to the dense sounding series during specific transport episodes and the field campaigns were beyond our present capability. As a consequence, no data for free-tropospheric ozone trend studies were generated.

Main Results

The focus of this section is on recent work since most of the investigations during the early phase of TOR 2 are already described in previous reports or in scientific journals. Nevertheless, a brief overview is also given on the early work.

a) Air-Pollution Transport into and across the Alps

Within the VOTALP (Vertical Ozone Transport in the Alps, parts 1 (1996-1998) and 2 (1998-2000)) project three international field campaigns to study the ozone transport from adjacent polluted source regions into/across the Alps. IFU participated in these campaigns with its aerosol and ozone lidars, in particular the newly developed mobile ozone lidar (Fig. 1).

Figure 1: The mobile ozone lidar of IMK-IFU during the VOTALP Milano Experiment (Barni, Prov. di Como, 620 m a.s.l.)

In May 1997 the VOTALP Föhn Campaign was carried out with instrumention located at many sites between the Po basin and Garmisch-Partenkirchen and aircraft operations (Seibert *et al.*, 2000). The IFU ozone lidars were operated at Garmisch-Partenkirchen and at Thaur near Insbruck. The measurements showed little vertical variation of ozone. However, in contrast to the TOR Föhn case (Carnuth *et al.*, 2002), a pronounced increase to 70 to 80 ppb was observed after the air from the Po basin intruded the area to the north of the Central Alps after the breakthrough of the Föhn wind (4 to 8 a.m. on May 5, see Fig. 2). During the following period the ozone mixing ratio gradually declined to less than 50 ppb.

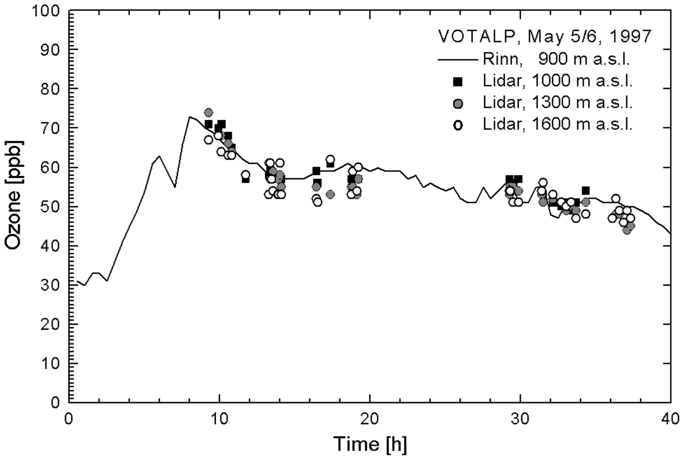

Figure 2: Lidar measurements in the Inn valley at Thaur selected for three different heights showing good agreement with *in-situ* measurements at the nearby station at Rinn. The time is given with respect to May 5, 0:00 CET (Central European Time = UTC + 1 h).

The two VOTALP ”Alpine foothill” field campaigns in 1998 and 1999 were devoted to the transport of polluted air from Milano and Munich into the Alps and to its potential uplifting by the first mountain range. The ”Munich Campaign” was of limited success due to a period of torrential rain preceding the measurements which resulted in rather moderate orographic winds. In contrast, the ”Milano Campaign”, carried out in co-operation with the LOOP project in the mountains between the two legs of Lago di Como, yielded excellent results. Throughout the week of the experiment (June 1 to 6, 1998) pronounced cycles of nighttime ozone reduction (wind from the Alps, 40 to 50 ppb of O_3) and daytime ozone advection from the Milano area in the ”valley wind” (see below) were observed, with a day-by-day concentration increase until June 4. Selected examples from the lidar measurements on June 4 are given in Fig. 3 and show a daytime ozone increase to 120 ppb, the highest PBL values ever recorded with an IFU lidar. The upvalley flow to Barni did not show any significant rise of the PBL height before 6 p.m.. In contrast, aircraft measurements by VOTALP partners showed some vertical uplifting downwind the mountain.

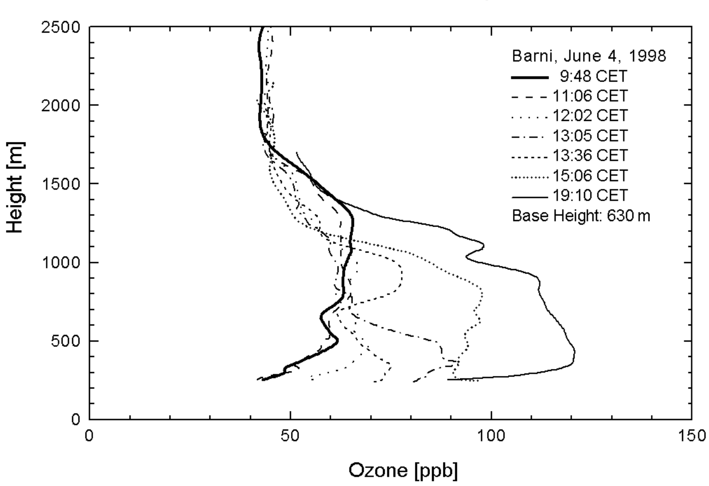

Figure 3: Ozone profiles selected from the measurements at Barni (Prov. di Como) on June 4, 1998

b) Vertical Transport in Alpine Valleys

The lidar measurements during the first TOR project had demonstrated the pronounced daytime vertical transport of ozone and aerosol to heights around 4000 m above the mountains next to IFU (Carnuth *et al.*, 2002). These results have been confirmed by aerosol sounding within the German Lidar Network, the European lidar network EARLINET and the German VERTIKATOR project in recent years. In the morning an upvalley air stream forms in the PBL of the local Loisach valley (”valley wind”) reaching IFU typically before noon and resulting in an increase of ozone and aerosol imported from outside the mountains. This flow rises along the slopes in the upper part of the valley and reaches heights of 1 to 1.5 km above the summits and crests of the adjacent mountains (2300 to 2962 m a.s.l.) under conditions of moderate humidity. A return flow may form at high altitudes (Carnuth *et al.*, 2002 (and references therein); Kreipl *et al.*, 2001) resulting in the formation of a second aerosol and ozone layer above the PBL. An example of a diurnal series of measurements within TOR 2 is given in Fig. 4, showing the simultaneous mid-day increase of ozone and aerosol above 3000 m due to the formation of this return flow. As we deduce from the simultaneous measurements with the tropospheric aerosol lidar (not shown here), revealing more details below 1.1 km, there is a delay of approximately 1.5 h between the arrival of aerosol at IFU (below 1.7 km, i.e., 1 km above the valley) and the return at higher altitudes.

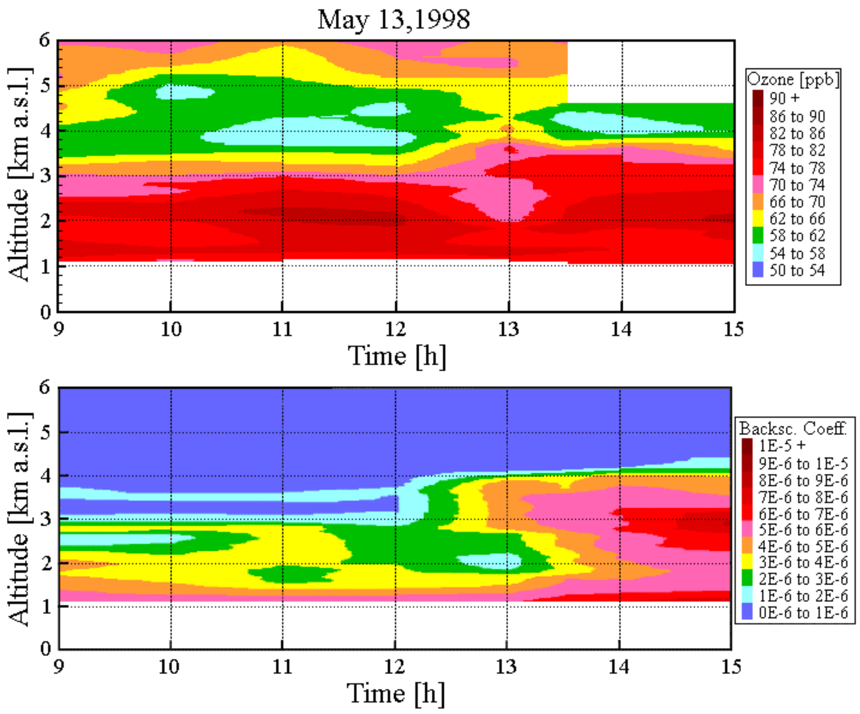

Figure 4: Simultaneous ozone and aerosol measurements with the ozone lidar at IFU on May 13, 1998, showing the influence of the orographic wind system in the Loisach valley

In general, the aerosol backscatter coefficients in the upper layer rarely drop to values below one third of those found in the PBL. To some extent, the decrease may be explained by the increase in valley cross section for growing altitude. The upward-transport efficiency is expected to exceed 50 %. A more accurate estimate has not been possible due the absence of wind data. Wind measurements were made by aircraft during the 2002 VERTIKATOR campaign, simultaneous to aerosol sounding at IFU and in the Murnauer Moos outside the mountains. The data analysis is still in progress.

The formation of a second aerosol layer above the PBL strongly depends on the degree of interference by the synoptic wind. The Munich radio-sonde wind data were used as a reference. It could be shown that all cases for which a bimodal vertical aerosol distribution was observed are associated with low synoptic wind speed and wind directions between east and south. For northerly advection the formation of an upper layer may be even completely suppressed.

The situation was different during the VOTALP field campaign in the Swiss Mesolcina valley (Furger et al., 2000; Carnuth et el., 2000). This valley is substantially deeper (300 m to 3000 m; Loisach valley below Garmisch-Partenkirchen: 650 to 2000 m). As a consequence the return flow was highly reproducible since it was, in part, channeled in the valley (Fig. 5). By incorporating the data from wind measurements onboard two Metair research aeroplanes a upward transport efficiency of roughly 80 % on July 19, 1996 (the only day with simultaneous aircraft and lidar measurements). This high value agrees well with the more extensive budget studies by researchers of the Paul-Scherrer-Institut (Furger *et al.*, 2000). It should be mentioned that the vertical distribution of the aerosol was not uniform across the valley. There was a pronounced difference in both PBL heights and extinction coefficients on both sides of the valley.

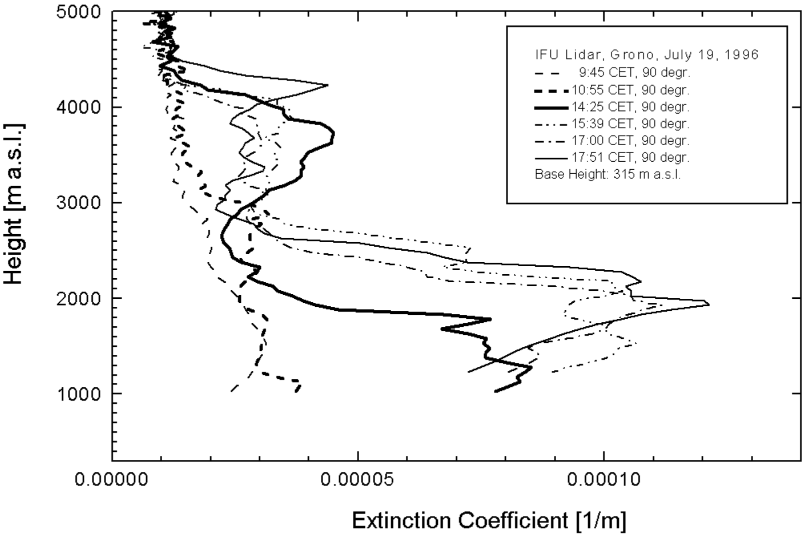

Figure 5: Aerosol extinction coefficients measured in the Mesolcina valley at Grono on July 19, 1996 (Carnuth and Trickl, 2000)

c) Stratospheric Air Intrusions

Downward transport of stratospheric air into the troposphere (STT) has been regarded as one of the principal natural sources of tropospheric ozone. A pronounced seasonal cycle with a maximum in spring has been found (e.g., Danielsen and Mohnen, 1977). In contrast to this, Beekmann *et al.* (1997) reported an almost constant frequency of tropopause folding over the year. The spring maximum should, therefore, be ascribed to a maximum in downward flux per fold in spring. Appenzeller *et al.* (1996) analysed the global ciculation between the tropics and the northern latitudes and found a maximum in the downward flux during the late spring months.

The investigations of STT within TOR 2 have been mostly based on research within the EU projects VOTALP and STACCATO (Influence of Stratosphere-Troposphere Exchange in a Changing Climate on Atmospheric Transport and Oxidation Capacity, Stohl *et al.*, 2003). Some of the key questions, which form the frame of ongoing activities on STT within the German ATMOFAST project, have been:

1. How is stratospheric air mixed into the troposphere? How much ozone is transferred to the upper and lower troposphere (in particular to the Alpine summit level)?
2. How can the ozone content in layers of stratospheric air in the troposphere be quantified?
3. How can we quantify the contribution of aged stratospheric air in the troposphere, which is presumably quite significant?
4. What is the contribution of STE to the spring-time ozone maximum?
5. Is there a trend in the frequency of STE events and in the input of stratospheric air into the troposphere, e.g., due to a changing climate?

The activites have comprised transport modelling with the EURAD, FLEXTRA and FLEXPART models, long-term measurements at the high-lying Central European summit stations Jungfraujoch, Zugspitze, Sonnblick and Mte. Cimone and ozone sounding with the lidar at Garmisch-Partenkirchen. Within STACCATO the vertical sounding activities, co-ordinated by IFU, have been extended to Mediterrenean area by including groups from Greece (Zanis *et al.*, 2002). The TOR-2 sounding stations Uccle and RIVM were also informed prior to common measurements. In this way an important intrusion area over Europe could be covered. Starting in November 2000 daily trajectory forecasts were provided by H. Wernli (ETH Zürich). Thus, a complete list of intrusions in the area of the participating stations could be generated.

Case Studies

The case studies within VOTALP and STACCATO yielded an excellent basis for the understanding of the different patterns of deep intrusions and for model validation. It was found from lidar measurements during a total of 66 cases in four years that all but a few direct intrusions reach heights around 3000 m in the Alps (Eisele *et al.*, 1999). Particularly spectacular examples of lidar measurements are shown in Figs. 6 and 7. Both measurement series were analysed in detail by comparison with FLEXPART model simulations (e.g., Stohl and Trickl, 1999; Zanis *et al.*, 2002). The FLEXPART result for the rather complex example in Fig. 7, showing at least two intrusions with an, in part, rather strange changing subsidence behaviour, is given in Fig. 8 and demonstrates the present capability of Lagrangian modelling based on ECMWF data. However, model intercomparisons within STACCATO based on the lidar and station results (e.g., Cristofanelli *et al.*, 2002; Roelofs *et al.*, 2002) showed substantial differences between the participating models in reproducing the details of the experimental findings, due to the differences in spatial resolution and numerical diffusion in Eulerian models.

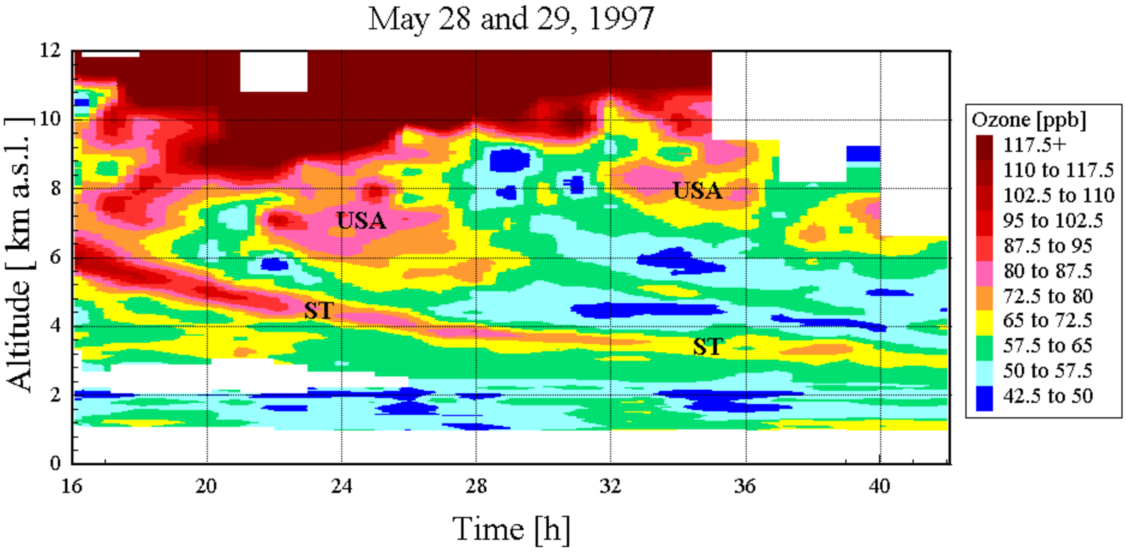

Figure 6: Lidar measurements at Garmisch-Partenkirchen between May 28 (16 CET) and May 29 (18 CET), 1997; the figure shows both a stratospheric air tongue (ST) descending to the lower troposphere and upper-tropospheric ozone maxima of North.American origin (USA) (Eisele et al., 1999; Stohl and Trickl, 1999).

Climatology Studies

In contrast to the expectations from earlier work the STT climatology obtained for the Alpine summit levels exhibits a winter rather than a spring maximum (Elbern *et al.*, 1997; Stohl *et al.*, 2000; James *et al.*, 2002). The seasonal cycle for the Zugspitze summit and seasonal mean values were obtained from both a re-analysis data from 1990 to 1999, and FLEXPART model runs yielded the annual cycles for the four VOTALP/STACCATO summit stations (James *et al.*, 2002). There is a summer minimum at all stations which is less pronounced at Jungfraujoch (3.5 km) than at the sites at lower elevations which suggest a height dependence of the intrusions. This seems to be confirmed by the lidar measurements: All intrusions registered with the lidar not reaching 3 km have, so far, taken place in summer. The obvious discrepancy between the lack of a summer minimum in the results by Beekmann *et al.* (1997) might be ascribed to their analysis approach based on sonde data covering also the height above 3500 m. The summer minimum in the lower troposphere indicates a lower penetration depth of stratospheric air intrusions during the warm season.

d) Intercontinental Transport

The first detection of intercontinental transport of ozone in May 1996 is one of the principal results of TOR 2 (Eisele *et al.*, 1999; Trickl *et al.*, 2002). The lidar measurements within TOR already showed that beginning anticyclonic periods may lead to a rich layer structure with ozone advection from rather different source regions (Carnuth *et al.*, 2002). However, the ozone mixing ratios of trans-

Atlantic origin did not exceed 60 ppb due to an air-mass passage over the southern United States (U.S.) at altitudes above 3 km. The examples in recent years have consistently shown subsiding layers with peak ozone values between 80 and 110 ppb in the free tropopsphere between roughly 4 km and 11 km during beginning anticyclonic conditions.

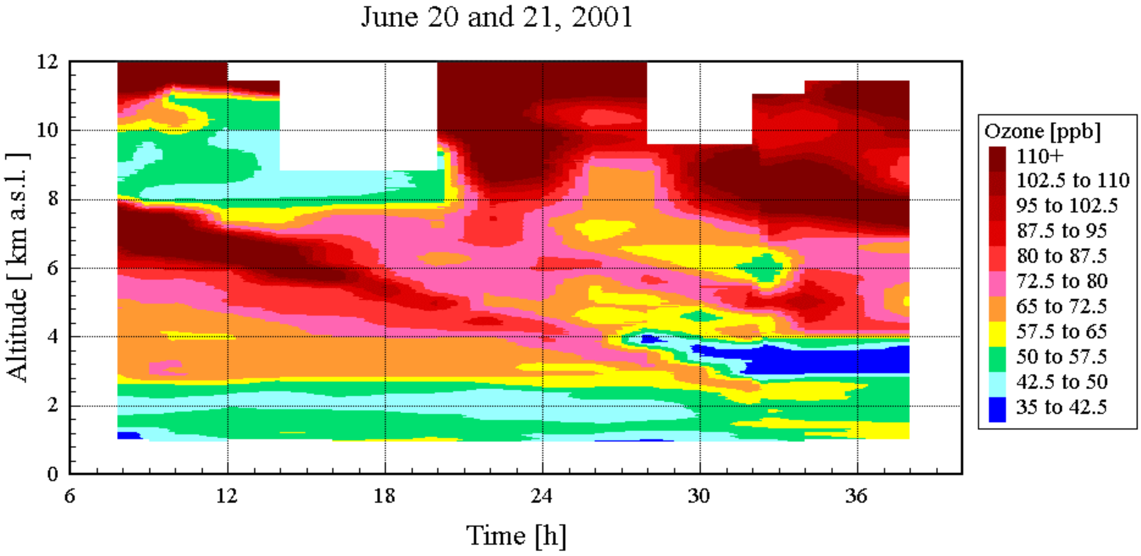

Figure 7: Lidar measurements at Garmisch-Partenkirchen on June 20 and 21, 2001, showing the influence of two major stratospheric air intrusions (labels 1 and 2) indicated by elevated O_3 concentrations (Zanis *et al.*, 2002; see Fig. 9). Intrusion 2 is the first STT event ever observed with the lidar from its beginning in the stratosphere.

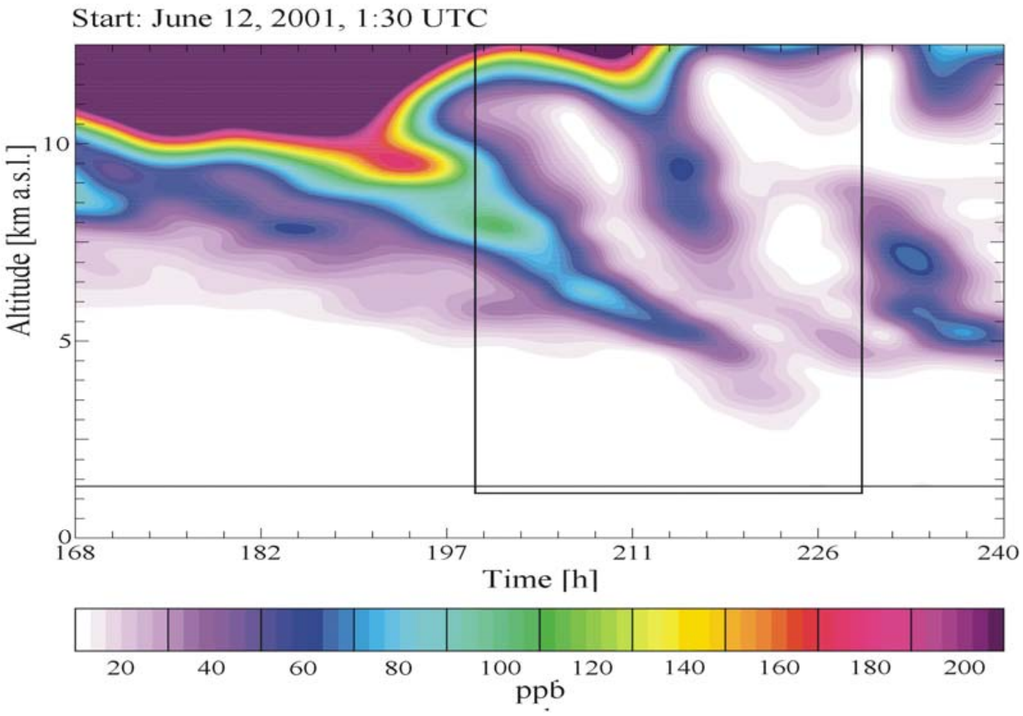

Figure 8: Simulation of the episode in Fig. 7 with the tracer model FLEXPART; the period of the lidar measurements is marked by a frame. Most structures of the measurements are reproduced by the stratospheric tracer (Zanis et al., 2002). Labels 1 and 2 correspoind to those in Fig. 7.

One case (May 1997), depicted in Fig. 6, yielded a very simple transport mechanism and a clear source-receptor relationship and was, therefore, the first example thoroughly analysed (Stohl and Trickl, 1999). A high-ozone episode in the eastern U.S. (up to 90 ppb) was terminated by a frontal passage exporting the polluted air mass from the continent where it got entrained in the warm conveyor belt (WCB) of this front and lifted to altitudes between 8 and 11 km. Here, the boundary-layer air was rapidly transported to Europe in the jet stream in an anticylonic pathway. Above Garmisch-Partenkirchen two descending upper-tropospheric layers with maximum ozone mixing ratios of the order of 90 ppb were registered, matching the values observed at the surface in the U.S. and during a MOZAIC flight across the North Atlantic. It is interesting to note the the normal atmospheric stratification is inverted, with stratospheric air at the bottom and boundary-layer air at high altitudes.

Other cases were found to be substantially more complex, including the ”classical” May 1996 case (Fig. 9). The general layer pattern in Fig. 9 has been reproduced by a number of more recent measurements during beginning anticycloning conditions. At the bottom a stratospheric intrusion is seen (May 29), followed by PBL formation with an ozone increase to 80 ppb and aerosol below 3.5 km. Above this there is a subsiding layer of air from the subtropical Atlantic with 25 to 35 ppb of O_3. The layers of enhanced O_3 in the middle and upper troposphere are of trans-Atlantic origin.

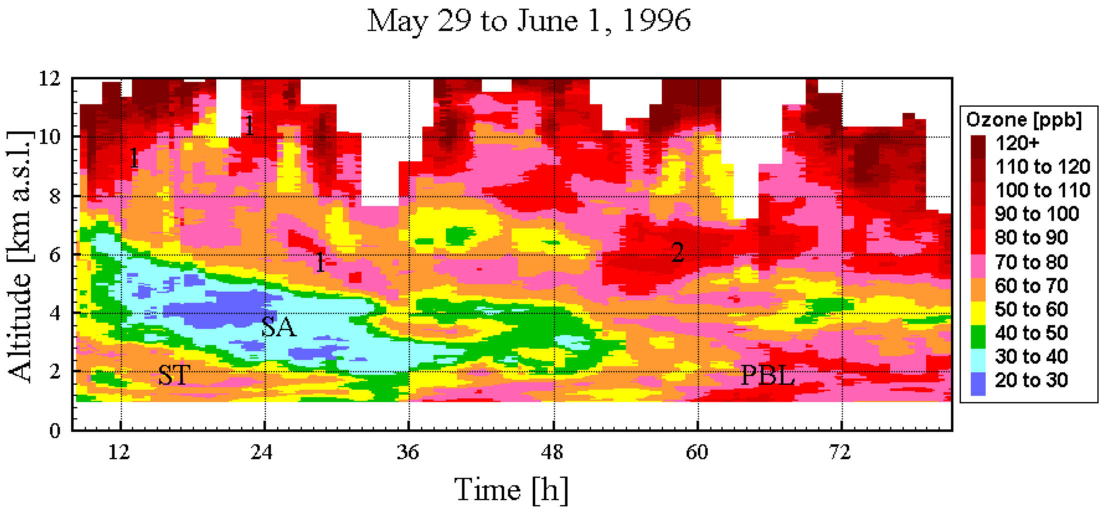
May 29 to June 1, 1996

Figure 9: Four-day measurement series with the IFU ozone lidar between May 29 (8 CET) and June 1 (9 CET), 1996, showing a rich layer structure throughout the observational period with contributions from the stratosphere (ST), the subtropical Atlantic (SA) and North America (labels 1 and 2). Labels 1 and 2 refer to contributions from the PBL and upper troposphere, respectively. After the end of the stratospheric intrusion a PBL formed (PBL) (Eisele *et al.*, 1999; Trickl *et al.*, 2002).

Although the advection pattern looks similar to that in the May-1997 case (see FLEXTRA backward trajectories in Fig. 10) the contribution of the eastern U.S. during the observational period is low since the air mass passes over that area above the PBL. Instead, contributions from Texas, Mexico and California could be identified by our analysis (area in Fig. 9 around labels 1). During the first half of the measurements an upper tropospheric branch (8 to 12 km) and a descending mid-tropospheric part of the North-American air may be distinguished, corresponding to the vertical steps at –40 h and –20 h (respectively) in the lower panel of Fig. 10. The export is complex, comprises mixing of different contributions and changes in composition every few hours (Trickl *et al.*, 2002). As in May 1997 a high-ozone episode in the eastern U.S. preceded the passage of the main front. Very interestingly, FLEXPART tracer simulations showed that the air mass directly lifted by the WCB of this front reached Central Europe prior to the beginning of the measurements, i.e., prior to the onset of the high-pressure period.

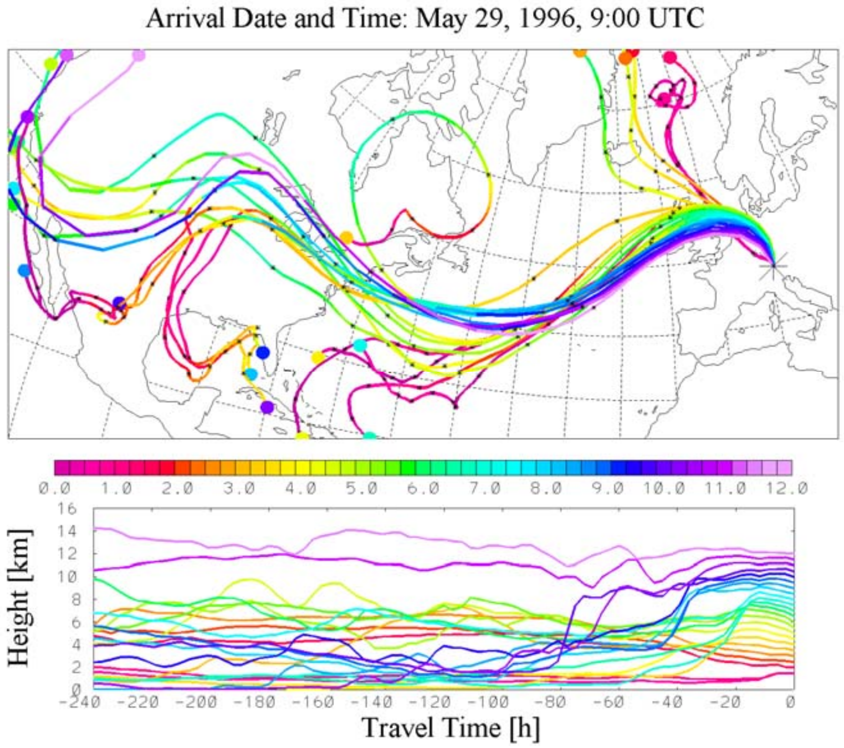

Figure 10: FLEXTRA backward trajectories for May 29, 9 UTC; the colour of the large dots at the end of the trajectories or their intersection indicates their height above Garmisch-Partenkirchen (Trickl *et al.*, 2002)

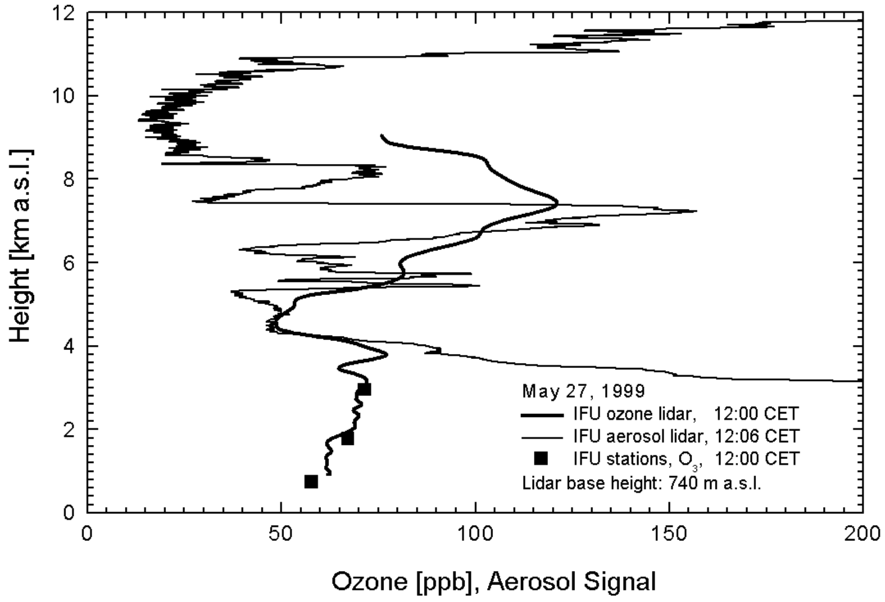

Figure 11: Ozone and aerosol measurements at IFU on May 27, 1999 (Trickl *et al.*, 2002); the (range-corrected) aerosol backscatter signal (wavelength 532 nm) in the free troposphere is substantially lower than that in the rather clean PBL.

The very dry layer around label 2 in Fig. 9 (relative humidity 10 %) has not been fully understood so far. The upper-tropospheric trajectories are traced back to the Pacific Ocean within just five days. However, it has not been possible to distinguish between an input from the stratosphere or from East Asia.

Trans-Atlantic transport is not restricted to the sine-wave-shaped advection pathway of the above examples associated with the onset of high pressure (see Fig. 10). During a four-day measurement period in May 1999 a layer above 5 km with up to 130 ppb ozone was almost straightly advected from the Great Lakes (Fig. 11). During this period aerosol was simultaneously measured with the big aerosol lidar of IFU and was found to be positively correlated with ozone in this layer. This verifies an export of PBL air from the central U.S. in a frontal system suggested by the trajectories. On the other hand the surface data in the U.S. do not support the high mixing ratios observed above our site. In addition, nearby humidity (and ozone) sounding by partners from the Paul-Scherrer-Institut revealed an anti-correlation of high ozone with relative humidity below 10 %. This could indicate an admicture of stratospheric air. However, some of the FLEXTRA trajectories show direct advection from the Pacific Ocean. Thus, again, also some input from Asia cannot be excluded.

e) Aerosol Spring Maximum

Aerosol is a suitable indicator of long-range air-pollution transport in the free troposphere. In contrast to other tracers such as CO it may be sensitively detected in the entire free troposphere by lidar remote sensing. Prefrontal advection of Saharan dust is the most important source of extra-European aerosol above our site. The air mass from the desert is lifted up to 5 km above the Mediterrenean Sea and may contain rather little ozone (e.g., Kreipl *et al.*, 2001; Carnuth *et al.*, 2002). It has also been known for some time that dust particles even of North American origin may occasionally reach Central Europe (e.g., Reiter *et al.*, 1984). In addition, also the influence of large-scale forest fires in North America must be taken into consideration (Wotawa and Trainer, 2000; Wotawa *et al.*, 2001; Forster *et al.*, 2001).

Low amounts of aerosol are expected for the advection pathways including warm conveyor belts (e.g., Stohl and Trickl, 1999; Kreipl *et al.*, 2001; Trickl *et al.*, 2002) due to washout. Indeed, the free tropospheric lidar measurements rarely show significant amounts of aerosol. Thus, only the frequency of aerosol layers occurring in the free troposphere may give some more quantitative information on the importance of long-range transport of air pollution. In order to determine the seasonal cycle of free-tropospheric aerosol three years of NDSC lidar measurements by H. Jäger (IFU) were analysed (Trickl and Wandinger, 2001). The structures were counted only if they exceeded 5 % of the Rayleigh backscatter coefficient, at a lidar wavelength of 532 nm. We selected only those measurements for which the aerosol layers may be clearly distinguished from boundary-layer aerosol or from residual cirrus clouds. For the period September 1997 to August 2000 the measurements may be distributed as follows:

Season	***Days with aerosol***	***All days***

Winter (Dec. 21 – March 20)	12 (36.4 %)	33
Spring (March 21 – June 20)	17 (65.4 %)	26
Summer (June 21 – Sept. 20)	12 (44.4 %)	27
Autumn (Sept. 21 – Dec. 20)	6 (18.2 %)	33

The maximum frequency of days with free-tropospheric aerosol occurs in spring. This stimulates a question about the importance of long-range transport also for the formation of the spring maximum of ozone. The overall fraction of cases with free-tropospheric aerosol layers is 39.5 % which is quite substantial and indicates a rather strong contribution of long-range advection from remote PBLs to the composition of the free troposphere over Central Europe. It should be mentioned that a spring peak of aerosol and CO was also reported on for the southern hemisphere by Jones *et al.* (2001).

Acknowledgements

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