Trans-Atlantic Transport of Aerosol and Ozone

P. James[2], A. Stohl[2], T. Trickl[1]

[1]Forschungszentrum Karlsruhe, IMK-IFU, Kreuzeckbahnstr.19, D-82467 Garmisch-Partenkirchen

[2]Lehrstuhl für Bioklimatologie und Immissionsforschung, Technische Universität München, Am Hochanger 13, 85379 Freising

Introduction

Elevated upper-tropospheric ozone mixing ratios (peak values 80 to 110 ppb) frequently ob-served above IFU under beginning anticlonic conditions in recent years could be reproducibly ascribed to trans-Atlantic transport. The layer sequence is rather similar (e.g., Figs. 1-4), due to a rather similar transnport pathway resembling a sine wave (Fig. 6). Upward transport from the North-American boundary layer (PBL) occurs within a warm conveyor belts (WCB) over the central Atlantic. The WCB belongs to a front which previously ended a high-O_3 period in the eastern United States (U.S.), However, not in all cases the eastern U.S. is the principal source region. Also other regions may contribute as well as input from the upper troposphere above the Pacific Ocean could be verified (Asia?). In 2002, a detailed analysis of several of these cases was completed, based on North-American surface data and MOZAIC flights (Figs. 6-19, Trickl et al., submitted to JGR, 2002).

As a consequence of the transport in the WCB, there is a high probability of washout of the PBL aerosols. In fact, the aerosol measurements at 313 nm have not shown clear aerosol signatures. Measurements with the sensitve NDSC lidar at IFU, simultaneously carried out with the ozone soundings in some of the cases, have revealed the presence of tiny amounts of aerosol (of the order of 0.1 of the Rayleigh values, or just slightly higher).

Aerosol was also detected in a case with a different advection pathway (May 26-30, 1999, Figs. 15-19). In this case, again, mixing with air from above the Pacific occurred. In addition, the formation of a mesoscale concective cell in the uplifting region makes the interpretation difficult. Up to130 ppb of O_3 were observed with the lidar which is beyond the values in the most likely source regions in the U.S.. Thus, admixture of stratospheric or Asian contributions must be considered.

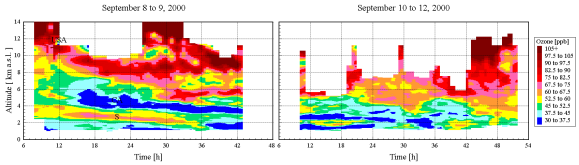


Fig. 2: Lidar soundings for Sept. 8 to 12, 2000, again showing a pattern as in Fig. 1; the interpretation is similar. PBL contributions from the U.S. are limited to Sept 8 (above 8 km), the main source region being in Alabama. Again,the model simulations suggest that the export from the east coast of the U.S. reaches Central Europe prior to the observational period.

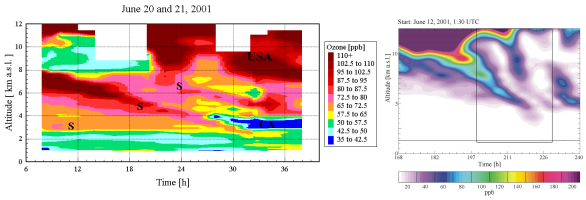


Fig. 3: Lidar measuremnts during the period June 20 to 21, 2001 showing ozone layers from 3 stratospheric air intrusions. Two intrusions are reproduced by a FLEXPART simulation (right panel). USA and CA denote input from the United States and the (sub)tropical Atlantic, respectively (as verified by model results by Roelofs et al. (manuscript submitted to JGR, 2003).

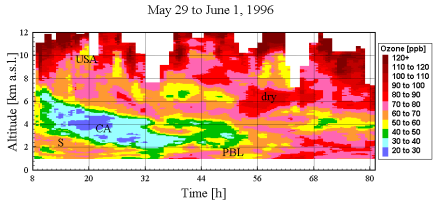


Fig.1: Four-day seroes of O_3 soun-dings at IFU showing for the first time North American influence (above 5 km). On May 29 and 30, the contributions above 8 km are ascribed to import from Texas or Mexico (Figs. 6 ,12). In the second half the high-O_3 period at around 6 km is associated with very low humidity (Fig. 14). The backward tra-jectories(Fig. 7) above 5 km mostly stay in the middle and upper troposphere 10 d backward in time and origi-nate in part beyond North America. The low-O_3 layer (CA) above the intrusion (S) is ascribed to air from the (sub)tropical Atlantic.

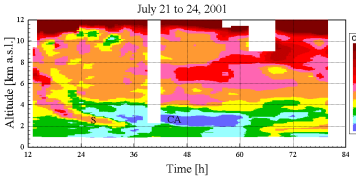


Fig. 4: Four-day lidar series, reproducing the general pattern in Figs. 1 and 2; the analysis is in progress. For the labels see Fig. 1.

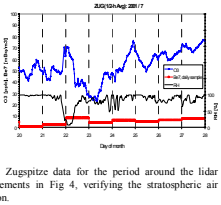
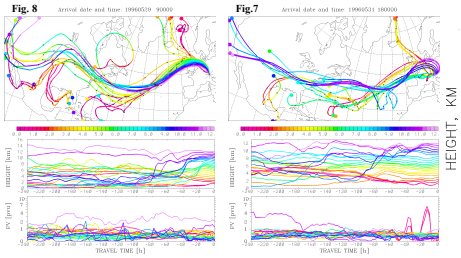


Fig. 5: Zugspitze data for the period around the lidar measurements in Fig 4, verifying the stratospheric air intrusion.



Figs. 6 and 7: FLEXTRA backward trajectories for May 29 and 31, 1996 (see Fig. 1)

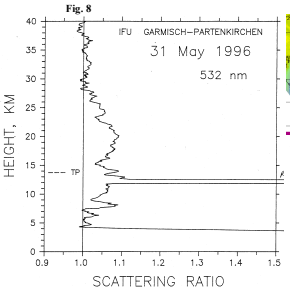


Fig. 8: Scattering ratio for a aerosol-lidar measurement on May 31, 1996, at about 22 CET, showing a peak at 8 km where input from beyond North America is suggested by the trajectories.

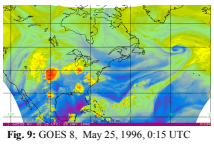
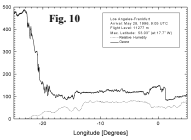
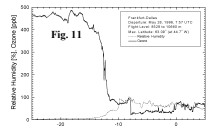
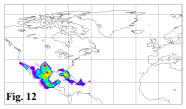
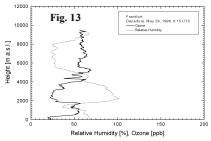


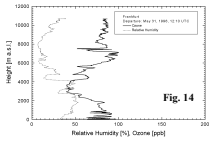
Fig. 9: GOES 8, May 25, 1996, 0:15 UTC











Figs. 10-11: MOZAIC measurements during transatlantic flights on May 28, 1996, through the WCB outflow region showing high-O_3 and low-O_3 contributions from different source regions

Fig. 12: 14-d integrated backward tracer simulation with the FLEXPART model (May 30, 3 UTC) folded with emission data from the EDGAR emission inventory for North America; the most likely source regions are in Texas and Mexico; the tracers are registered at a height of 500 m a.s.l..

Figs. 13-14: MOZAIC O_3 and RH profiles for Frankfurt for two relevant times in Fig. 1; the moderate ozone values in Fig. 13 are ascribed to lacking input from Texas for a few hours. In Fig. 14, the combination of low hunidity and high O_3 values above 5 km might suggest input from the stratosphere, but clear evidence is missing.

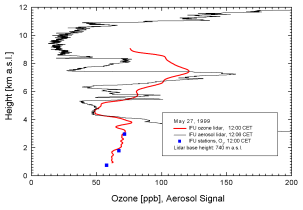


Fig. 14: Single O_3 profile from the measurements series May 26-30, 1999; simultaneous aerosol measurements show a good correlation of O_3 and particles indicating the presence of PBL air. However, an anti-correlation with humidity was found by nearby balloon ascents carried out by PSI (CH) which could suggest some admixture of stratospheric air.

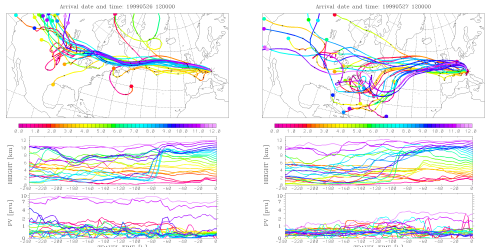


Fig. 16: FLEXTRA backward trajectories for May 26, 1999, 12 UTC

Fig. 17: FLEXTRA backward trajectories for May 27, 1999, 12 UTC, corresponding to the measurements in Fig. 15

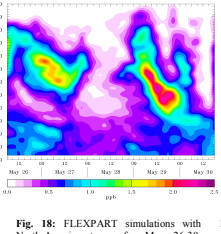


Fig. 18: FLEXPART simulations with North-America tracer for May 26-30, 1996, showing nice agreement with the high-O_3 layers observed with the lidar during this period

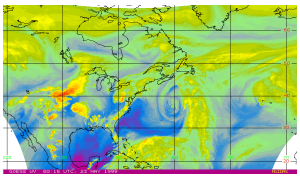


Fig. 19: GOES 8 water-vapour image (May 23, 1999, 0:15 UTC); the source region for the air arriving over IFU on May 26 and 27 is near 40º N, 100º W, where strong convective activity is seen. Surface data in the south west of the Great Lakes indicate O_3 below 80 ppb.

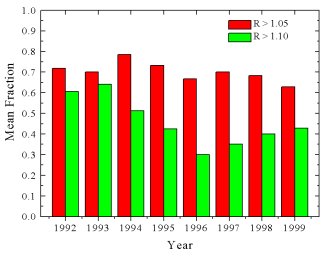


Fig. 20: see text

Long-range Transport and the Free-tropospheric Aerosol above Central Europe

Free-tropospheric aerosol backscatter coefficients are mostly very low (at 532 nm: < 50 % of the Rayleigh values). Exceptions are, e.g., Saharan dust or fire plumes not affected by washout or dilution. The fraction of the number of days with observations of free-tropospheric aerosol in the total number of measurement days should be much less affected by washout. Therefore, these fractions have been determined on an annual (Fig. 20) and monthly (Fig. 21) basis using the scattering ratios R from the highly sensitive NDSC lidar measurements at IFU (447 measurements since 1992, so far analysed for 1992-1999). This analysis will be refined within the next few weeks by intercomparison with FLEXTRA trajectories currently calculated for all 447 days (0.5 km vertical intervals). A clear influence of the threshold conditions (5 % and 10 % above the Rayleigh values) is seen. The results for R > 0.1 indicate stratospheric influence (positive correlation with the Mt.-Pinatubo aerosol, seasonal cycle less pronounced). The seasonal cycle peaks in spring, similarly to the spring maximum ozone seen at background station in the northern hemisphere.

Funding: European Union: VOTALP, STACCATO, EARLINET; German BMBF: German Lidar Network, ATMOFAST

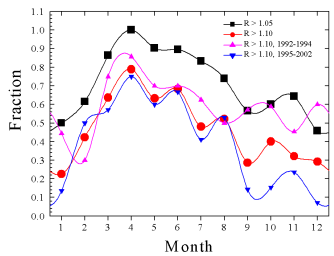


Fig. 21: see text