EPJ Web of Conferences **362**, 09002 (2026) https://doi.org/10.1051/epjconf/202636209002
ILRC31/CLRC22-2024

Is there a correlation between tropospheric ozone and climate?

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Abstract: Lidar vertical sounding of ozone, water vapour and aerosol at Garmisch-Partenkirchen in the German Alps and long-term in-situ measurements at local mountain stations up to 3 km a.s.l. have been the basis for investigations of atmospheric transport over several decades. The positive ozone trend in the lower free troposphere is dominated by downward transport from the stratosphere. The thin intrusion layers are extremely dry which suggests very low mixing with tropospheric air, in contrast to the traditional view. The related ozone trend at the Zugspitze summit (2962 m a.s.l.) follows the changes in solar radiation. This indicates a correlation of the vertical exchange with the climate. Parallel to this development the background air quality has improved, as demonstrated by the decrease of CO and NO_x since 1990.

1. Introduction

Lidar time series combined with high-resolution transport modelling have yielded considerable insight into long-range atmospheric transport. At Garmisch-Partenkirchen in the German Alps transport has been investigated over several decades by lidar vertical sounding of ozone, water vapour and aerosol and long-term in-situ measurements at the mountain stations Zugspitze (2962 m a.s.l.), Schneefernerhaus (UFS, 2570 m a.s.l.) and Wank (1780 m a.s.l.) [1-4]. High Alpine summits provide, around the clock, valuable additional information on constituents not accessible by lidar sounding. A topic of special interest is the downward transport of ozone from the stratosphere which has been regarded as the most important natural source of tropospheric ozone. The lidar measurements have yielded details of the stratospheric layers whereas the continuous measurements of O_3, relative humidity (RH) and ^7Be (an important tracer for stratospheric air) at 2962 m offer the chance to quantify the amount of O_3 related to stratosphere-troposphere transport (STT) based on observational data alone. Quite surprisingly, the humidity profiles have revealed that the descending layers are much drier than previously thought [5-7]. Thus, the influence of their mixing with tropospheric air can be regarded to be rather low.

In this contribution, we give a brief overview of the results. More details can be found in Ref. 8 and other publications cited.

2. Instrumentation

Tropospheric vertical sounding at Garmisch-Partenkirchen has been carried out with a three-wavelength ozone differential-absorption lidar (DIAL) [9], a water-vapour DIAL (at UFS) [10] and a mobile three-wavelength aerosol lidar system (within EARLINET, European Aerosol Research Lidar Network, 2000-2002), eventually converted into a high-spectral-resolution lidar [11,12]. The DIAL systems cover a vertical range up to the tropopause region and feature relative uncertainties of just a few per cent in a major part of the operating range [5,7,13].

The in-situ measurements at the three mountain stations have been carried out and routinely validated with conventional methods of the Global Atmosphere Watch (GAW) programme (see Ref. 8 for details).

3. The role of humidity

The low humidity in stratospheric intrusion layers mentioned in the Introduction has been verified in more than 80 free-tropospheric measurements with the UFS DIAL. In Fig. 1 we present one example from Ref. 7. These measurements were carried out during the LUAMI (Lindenberg Upper-Air Methods Intercomparison) campaign in October 2008 during which a dry intrusion layer was mapped by lidar sounding of water vapour at four Central European stations and onboard an aircraft. The minimum H_2O density was substantially lower than that for 1 % RH.

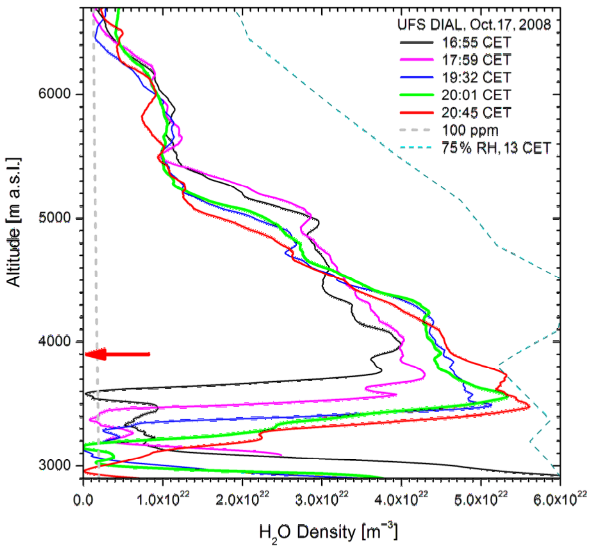

Figure 1. H_2O profiles from the measurements of Zugspitze DIAL on 17 October 2008 [7]; the red arrow marks the vertical position of the RH minimum from the noon "Munich" radiosonde. The grey dashed line marks a mixing ratio of 100 ppm.

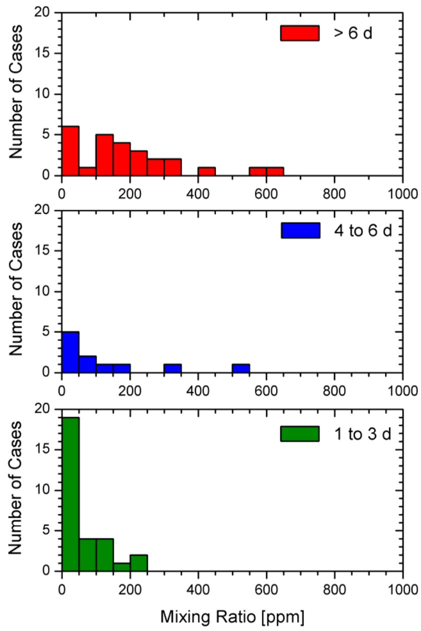

Figure 2. Histograms of the minimum H_2O mixing ratios on days with a deep stratospheric air intrusion (h $\leq$ 4.5 km) for three different ranges of air-mass travel times [5].

Figure 2 shows the statistical analysis of the minimum H_2O mixing ratios in deep stratospheric intrusions captured by the UFS DIAL [5]. For rapid descents from the tropopause (typically from areas around Greenland) the lowest mixing ratio is found. However, even for very long transport times occasionally extremely dry layers have been observed, although on average the minimum H_2O mixing ratio grows for longer transport. This behaviour could be verified for RS92 radiosonde measurements in a climatological study [4].

It is important to mention that extremely low H_2O values do not necessarily mean very high ozone. The ozone mixing ratio at the centre of an intrusion layer may even be quite moderate. The peak ozone depends on how far above the tropopause this layer originates [5,7].

The minimum relative humidity measured at the Zugspitze summit is rarely similarly low. These cases are limited to the cold season and night-time (compare validation exercise in Ref. 10). It is obvious that upslope winds [14] influence the station even under the condition of subsidence. Given the lidar results we can exclude tropospheric mixing to influence the results of the analysis shown below.

In Fig. 3 we show averaged Zugspitze RH percentiles. The lowest percentiles correspond to stratospheric intrusions. The RH descends until the end of the 20th century. After the beginning of decreasing solar activity in the late 1990s the negative trend stops. This is an intriguing observation and will be discussed in Sec. 6.

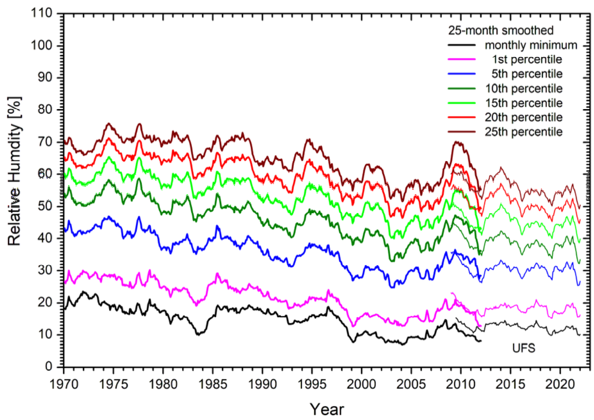

Figure 3. Monthly Zugspitze RH percentiles averaged over ±12 months [8]: The driest values correspond to stratospheric intrusion layers. The summit measurements terminate in 2011, but have been continued at UFS.

4. Stratospheric component of the Zugspitze ozone

The radioisotope ^{7}Be in the atmosphere is primarily produced in the stratosphere. At high latitudes, relevant for intrusions towards Central Europe, at least 80 % of the isotope originate in the stratosphere. We assume stratospheric portions of 80 % and 90% in our analysis.

Based on the RH and ^{7}Be time series the stratospheric component in the Zugspitze ozone was determined. We applied two data filtering criteria almost identical to those successfully validated previously [3]: RH < 50 % plus ^{7}Be > 60th percentile (^{7}Be criterion), or RH < 50 % plus RH_{min} < 30 % (RH criterion). Stratospheric contributions not identified by these criteria are named *indirect* intrusions and are identified by residual ^{7}Be up to 80 or 90 % of the values. The corresponding ozone is estimated by converting ^{7}Be to ozone based on comparing monthly ^{7}Be and intrusion O_3 averages (see Ref. 8). The results are depicted in Fig. 4.

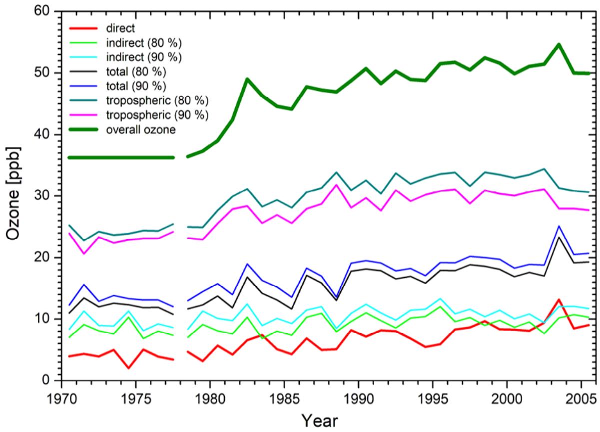

Figure 4. Annual average Zugspitze ozone mixing ratios in direct or indirect intrusions (for assuming 80 % and 90 % stratospheric origin of ^{7}Be), as well as tropospheric component [8]. Before the beginning of the quality-assured ozone measurements in 1978 we assume constant annual O_3 averages.

In contrast to the emphasis until the 1990s on the role of the anthropogenic ozone production the stratospheric influence on the Zugspitze ozone is rather remarkable (up to 40 %) and determines the positive trend starting in the mid-1970s. A closer look at the monthly means reveals that the positive trend is mainly caused by a wintertime rise in intrusion ozone.

The ^{7}Be time series ends in April 2006. Afterwards, the RH criterion must be applied. This is not a bad choice since the analyses for the two criteria yield not much different results. The RH criterion allows us to extend the analysis by using the GAW measurements at UFS (Fig. 5).

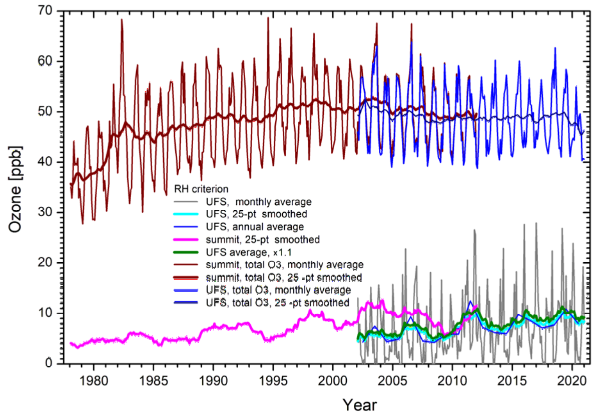

Figure 5. Monthly ozone averages and ±12-month running averages (total O_3 and direct stratospheric component) for Zugspitze summit (until the end of the series in 2011) and UFS (until 2020) [8]. The UFS RH data until 2008 are questionable (see discrepancy).

The figure reveals that the positive stratospheric trend in the Zugspitze ozone ends in 2003. The rise in the tropospheric component in the Zugspitze O_3 ends in the 1980s (Fig. 4). The measurements of CO [8] and NO_x started in 1990 reveal a considerable improvement of the air quality since that year.

5. Free Troposphere

The lidar measurements extend the information obtained at the summit stations to the rest of the free troposphere (FT). Due to the absence of ^{7}Be data and the limitation in measurement times a full quantification of the transfer of stratospheric O_3 into the FT has not been possible. Nevertheless, the routine measurements between 2007 and 2018 (H_2O until 2014, complemented by radiosonde profiles) have yielded a very high stratospheric influence above 4 km. The analysis until 2016 [4] revealed that at least one stratospheric layer was present in the FT on 84 % of the measurements days. The pronounced summer minimum of stratospheric ozone observed at the European summit stations [3] disappears. In fact, frequently very high ozone values are found in the middle and upper troposphere during the warm season. In this altitude range mixing ratios sometimes range between 100 ppb and 150 ppb, too high for justifying to assume an exclusively remote tropospheric source, despite a not thoroughly low humidity (for examples see Refs. 4 and 13.

6. Discussion and Conclusions

Lidar sounding at Garmisch-Partenkirchen, in combination with continuous (not fair-weather-biased) in-situ measurements at high-altitude sites has revealed a lot of detail on long-range transport, particularly on STT. STT has a considerable impact on the FT, which is higher than thought in the past. For the first time the stratospheric ozone contribution at a high-altitude summit station has been determined based on observational data alone, a consequence of the long time series of ^{7}Be and RH.

The ozone contribution from STT increased from the 1970s to the first years of the new century and then stayed rather constant or slightly increased. It is not unreasonable to assume a change in atmospheric dynamics caused by the change in solar irradiation as the source of this behaviour. STT explains the atmospheric drying at the Zugspitze summit observed until 2000. A drying FT has been reported even on a global scale in a study covering 40 years [15]. A dryer FT contradicts the expectations from climate modelling. However, coarse climate models do not have the capability to resolve thin intrusion layers [16,17].

7. References

[1] A. Stohl and T. Trickl, "A textbook example of long-range transport: Simultaneous observation of ozone maxima of stratospheric and North American origin in the free troposphere over Europe," J. Geophys. Res. **104**, 30445-30462 (1999).

[2] T. Trickl, O. R. Cooper, H. Eisele, P. James, R. Mücke and A. Stohl, "Intercontinental transport and its influence on the ozone concentrations over central Europe: Three case studies, J. Geophys. Res. **108**, 8530, doi: 10.1029/2002JD002735, 23 pp. (2003).

[3] T. Trickl, H. Feldmann, H.-J. Kanter, H. E. Scheel, M. Sprenger, A. Stohl and H. Wernli, "Deep stratospheric intrusions over Central Europe: case studies and climatological aspects," Atmos. Chem. Phys. **10**, 499-524 (2010).

[4] T. Trickl, H. Vogelmann, L. Ries and M. Sprenger, "Very high stratospheric influence observed in the free troposphere over the Northern Alps – just a local phenomenon?" Atmos. Chem. Phys. **20**, 243-266 (2020).

[5] T. Trickl, H. Vogelmann, H. Giehl, H. E. Scheel, M. Sprenger and A. Stohl, "How stratospheric are deep stratospheric intrusions?" Atmos. Chem. Phys. **14**, 9941-9961 (2014).

[6] T. Trickl, H. Vogelmann, H. Flentje and L. Ries, "Stratospheric ozone in boreal fire plumes – the 2013 smoke season over Central Europe," Atmos. Chem. Phys. **15**, 9631-9649 (2015).

[7] T. Trickl, H. Vogelmann, A. Fix, A. Schäfler, M. Wirth, B. Calpini, G. Levrat, G. Romanens, A. Apituley, K. M. Wilson, R. Begbie, J. Reichardt, H. Vömel and M. Sprenger, "How stratospheric are deep stratospheric intrusions into the troposphere? LUAMI 2008," Atmos. Chem. Phys. **16**, 8791-8815 (2016).

[8] T. Trickl, C. Couret, L. Ries and H. Vogelmann, Zugspitze ozone 1978 – 2020: The role of stratosphere-troposphere transport, Atmos. Chem. Phys. **23**, 8403–8427 (2023).

[9] T. Trickl, H. Giehl, F. Neidl, M. Perfahl and H. Vogelmann, "Three decades of tropospheric ozone-lidar development at Garmisch-Partenkirchen, Germany," Atmos. Meas. Tech. **13**, 6357-6390 (2020).

[10] H. Vogelmann, and T. Trickl, "Wide-range sounding of free-tropospheric water vapor with a differential-absorption lidar (DIAL) at a high-altitude station," Appl. Opt. **47**, 2116-2132 (2008).

[11] H. Giehl and T. Trickl, "Testing the IFU High-Spectral-Resolution Lidar at the 2009 Leipzig Field Campaign," pp. 920-923 in: *Proceedings of the 25th International Laser Radar Conference, St.-Petersburg (Russia), 2010*, G. Matvienko, A. Zemlyanov, Eds., V. E. Zuev Institute of Optics (Tomsk, Russia, 2010).

[12] U. Wandinger and many co-authors, "EARLINET instrument intercomparison campaigns: Overview on strategy and results," Atmos. Meas. Tech. **9**, 1001-1023 (2016).

[13] T. Trickl, M. Adelwart, D. Khordakova, L. Ries, C. Rolf, W. Steinbrecht and H. Vogelmann, "Local comparisons of tropospheric ozone: Vertical sounding at two neighbouring stations in Southern Bavaria," Atmos. Meas. Tech. **16**, 5145–5165 (2023)

[14] W. Carnuth and T. Trickl, "Transport studies with the IFU three-wavelength aerosol lidar during the VOTALP Mesolcina experiment," Atmos. Environ. **34**, 1425-1434 (2000).

[15] G. Paltridge, A. Arking, and M. Pook, "Trends in middle- and upper-level tropospheric humidity from NCEP reanalysis data," Theor. Appl. Climatol. **98**, 351–359 (2009).

[16] G. J. Roelofs, A. S. Kentarchos, T. Trickl, A. Stohl, W. J. Collins, R. A.. Crowther, D. Hauglustaine, A. Klonecki, K. S. Law, M. G. Lawrence, R. von Kuhlmann, and M. van Weele, M., "Intercomparison of tropospheric ozone models: Ozone transport in a complex tropopause folding event," J. Geophys. Res. **108**, 8529, doi: 10.1029/2003JD003462, 13 pp. (2003).

[17] S. D. Eastham and D. J. Jacob, "Limits on the ability of global Eulerian models to resolve intercontinental transport of chemical plumes," Atmos. Chem. Phys. **17**, 2543–2553 (2017).