



## In-situ ozone observations at Zugspitze (2962 m) from 1978 to 2008

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Ozone concentrations observed at the Zugspitze mountain site (47 °N, 11 °E) were analysed with respect to long-term trend, seasonal variations, and impact of different meteorological conditions. From concurrent measurements conducted at the site, data for carbon monoxide (CO), relative humidity (RH) and the stratospheric/upper-tropospheric tracer  $^7\text{Be}$  were used for filtering of ozone data.

The entire  $\text{O}_3$  record is characterised by seasonal cycling with a summer maximum and a winter minimum as well as pronounced short-term variations (from minutes to days) and year-to-year variability. Depending on specific meteorological condition, the  $\text{O}_3$  mole fractions have varied over a range from near zero to about 110 ppb. The annual average  $\text{O}_3$  levels have increased from about 35 ppb in 1978 to about 50 ppb in 2008.

According to the characteristics of the ozone increase, the record can be divided into three parts. The period 1978-1989 displays the strongest increase, for which the annual means yield an average growth rate of  $1.0 \text{ ppb yr}^{-1}$ . The 1990-2002 data still show a significant ozone growth of  $0.25 \text{ ppb yr}^{-1}$ . In contrast, from 2004 onwards approximately constant annual  $\text{O}_3$  levels around 50 ppb were observed, while  $\text{O}_3$  during 2003 was biased by three exceptionally high monthly values during summer. Taken together, this means that no trend can be detected for the last six years.

Comparisons with the surface ozone at the neighbouring Wank summit (1780 m a.s.l.) have revealed smaller and partly negative growth rates at the lower altitude during the 1990s, and a remarkable agreement in the trend behaviour from 2000 onwards.

Data analyses have aimed at the detection of differences in the mechanisms predominantly driving the trend in the first and the second part of the record. The annual distributions of the average growth rates were analysed with a temporal resolution of 11 days and indicate a shift of the seasons contributing most to the  $\text{O}_3$  increase from summer (1978-1989) to autumn/end of winter (1990-2002). This change can be related to the known general decrease of pollution emissions over Europe, leading to less photochemical  $\text{O}_3$  production in summer and less  $\text{O}_3$  loss during winter. Nonetheless, the average seasonal cycles for 1978-1989 and 1990-2002 agree in their only slightly asymmetric shape, displaying a broad spring/summer maximum extending from April to August, with similar peak-to-peak amplitudes of about 16 ppb. In contrast, the recent 2004-2008 average cycle is more asymmetric with a spring maximum (April/May) and a seasonal  $\text{O}_3$  decrease starting already around July.

Special regard was given to a comparison of  $\text{O}_3$  in polluted and unpolluted air masses as well as to the influence of air from the lower stratosphere/upper troposphere. The respective contributions to the observed ozone levels were estimated together with the associated growth rates. While all the average growth rates of the period 1978-1989 are close to  $1 \text{ ppb yr}^{-1}$  (range  $0.98 - 1.17$  for the different filter criteria), the relative differences are much greater among the generally smaller rates (range  $0.23 - 0.44 \text{ ppb yr}^{-1}$ ) obtained for the different conditions during the second part of the time series (1990-2002). The all-data rate has dropped to 23 % of its value for the first part, whereas the stratospherically flagged data display a slightly stronger trend, with average rates ranging between 35 % to 44 % of the respective previous growth rates. Generally,  $\text{O}_3$  data (1990-2002) filtered with respect to clean air (stratospheric criteria or CO criterion) have yielded trends above the all-data result. In contrast,  $\text{O}_3$  data (1990-2002) selected from polluted air (CO filter criterion) show growth rates below the all-data rate.

Based on the statistical parameters available from the data flagging for stratospheric/upper tropospheric air (humidity and  $^7\text{Be}$  criteria), the temporal development of ozone influence from the lower stratosphere was assessed. From the annual number and average duration of events it is suggested that the contribution of middle and upper

tropospheric air masses has been growing stronger than the more stratospherically influenced contributions.