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Ozone Climatology Studies for the Zugspitze and Neighbouring Sites in the German Alps

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Summary

Surface ozone data collected at the mountain sites Zugspitze (2962 m a.s.l.) and Wank (1780 m a.s.l.), 47°N, 11°E, since 1978 have been studied with respect to temporal changes in the characteristics of the long-term trend and seasonal cycle. Trend estimates over different periods between 1978 and 2001 have given evidence for a shift of the months with the highest growth rates from summer to winter. Zugspitze ozone data filtered by meteorological and trace gas parameters (1990 – 2000) indicate higher growth rates for clean air conditions, including subsidence, than the all-data value of 0.25 ppb yr^{-1}. The trend estimates based on different subsets of the data suggest that changes in the advection patterns may have contributed to the O_3 increase observed at Zugspitze during recent years. With 11-day moving medians averaged over the last decade, a representative shape of the seasonal cycle at high temporal resolution has been determined, which reveals structures that get lost with monthly means. The general pattern with a broad spring/summer maximum has not significantly changed at both sites, when comparing first and second part of the record. Using carbon monoxide as data filter, the seasonal cycles for 'unpolluted' and 'polluted' conditions at the Zugspitze have been determined. The cycle associated with low CO levels is assumed to represent O_3 background concentrations.

Introduction

Due to the location of the mountains sites Zugspitze (ZUG) and Wank (WNK), and the valley site Garmisch (740 m a.s.l.) inside the European continent, the ozone records are characterised by strong short-term variations and interannual variability brought about by the prevailing meteorological conditions. The valley site is strongly affected by local conditions. Since its O_3 record does not indicate any long-term trend and the seasonal cycle (day-time data) was previously shown in a European perspective (Scheel *et al.*, 1997), recent results form Garmisch will not be considered here.

By using appropriate data selection criteria, the ozone concentrations related to a number of atmospheric conditions were separated with the aim of detecting those ones that had a major impact on the overall ozone trend, and to quantify their contributions to the seasonal cycle. When using data filters, emphasis was given to the visualisation of each subset in comparison to the entire data on an annual basis. Besides the meteorological parameters relative humidity (RH) and visibility, data filters were based on ^{7}Be as tracer for stratospheric/upper-tropospheric air and on carbon monoxide (CO) as indicator of anthropogenic pollution. It is important to note that the choice of the selection criteria (cut-off levels) for data filtering remains somewhat subjective. However, it critically affects data coverage and the resulting ozone statistics. For higher temporal resolution than obtained with monthly means, 11-day moving averages (or medians) calculated from the half-hourly data points were a good compromise between sufficient resolution and necessary degree of smoothing.

Results

Based on dynamic harmonic regression, the seasonal component and the long-term trend of the O_3 records were determined (Fig. 1). Around the end of the 1980s the peak-to-peak amplitude at Wank started decreasing, and has recently stabilised at about 16 ppb. In contrast, the amplitude at ZUG has remained largely constant. The trend curves were at similar levels until the middle of the 1990s. Thereafter a concentration difference has developed, yielding ~ 5 ppb higher levels at ZUG. From about 1995 onwards the O_3 trend of the Wank site displays a small decrease.

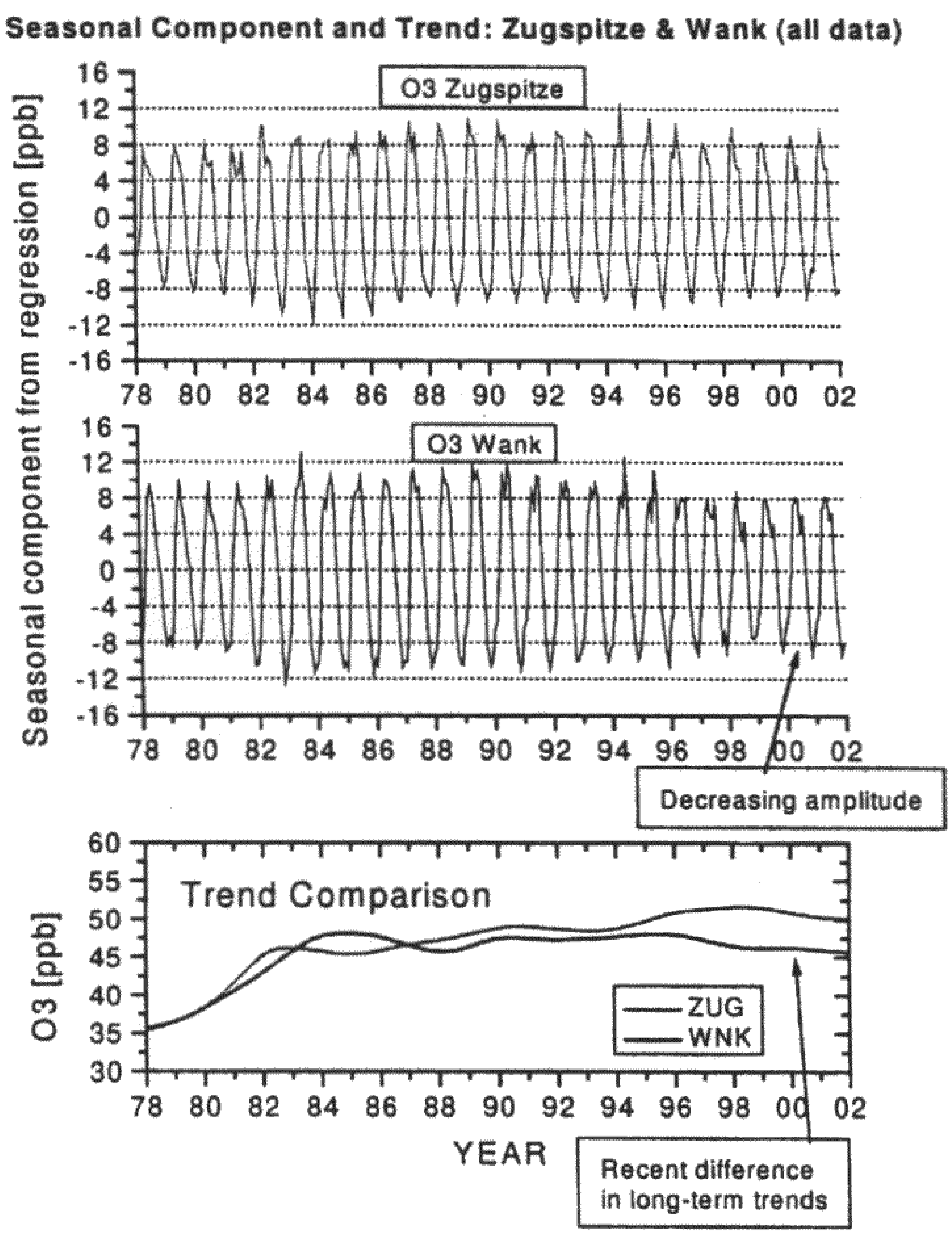

Figure 1. Results from dynamic harmonic regression on monthly means. Seasonal component and trend of O_3 at Zugspitze and Wank.

Changes in the trend characteristics of the time-series were detected by comparisons between the first and second parts of the 24-year long ozone records, using different statistical approaches. Linear regression performed on percentiles between 1st and 99th from an annual data basis already indicated systematic changes. For 1978 - 1989 the highest O_3 increase was associated with the higher percentiles, which mainly represent the summer levels. In contrast, for the period 1990 – 2001 a statistically significant increase for ZUG was only observed for the lower percentiles (largely winter values) up to the median. For the Wank site, only the lowest concentrations, as represented by the 1st percentile, showed a significant increase, whereas the 95th and 99th percentiles indicated a significant decrease.

By using 11-day moving medians, average growth rates were determined at high temporal resolution. Results for 1990-2001 are shown in Figure 2. The sudden decrease of the rates in March and the variability from July to September are noteworthy. The +/- signatures of March and August would not be represented by monthly statistics. On an annual scale the growth rates for Wank yield a negative value (cf. Fig. 1). The resulting positive rate for Zugspitze indicates major differences in the origin of the air at these sites. A stronger influence of subsidence at ZUG is suggested by the ^{7}Be and humidity records.

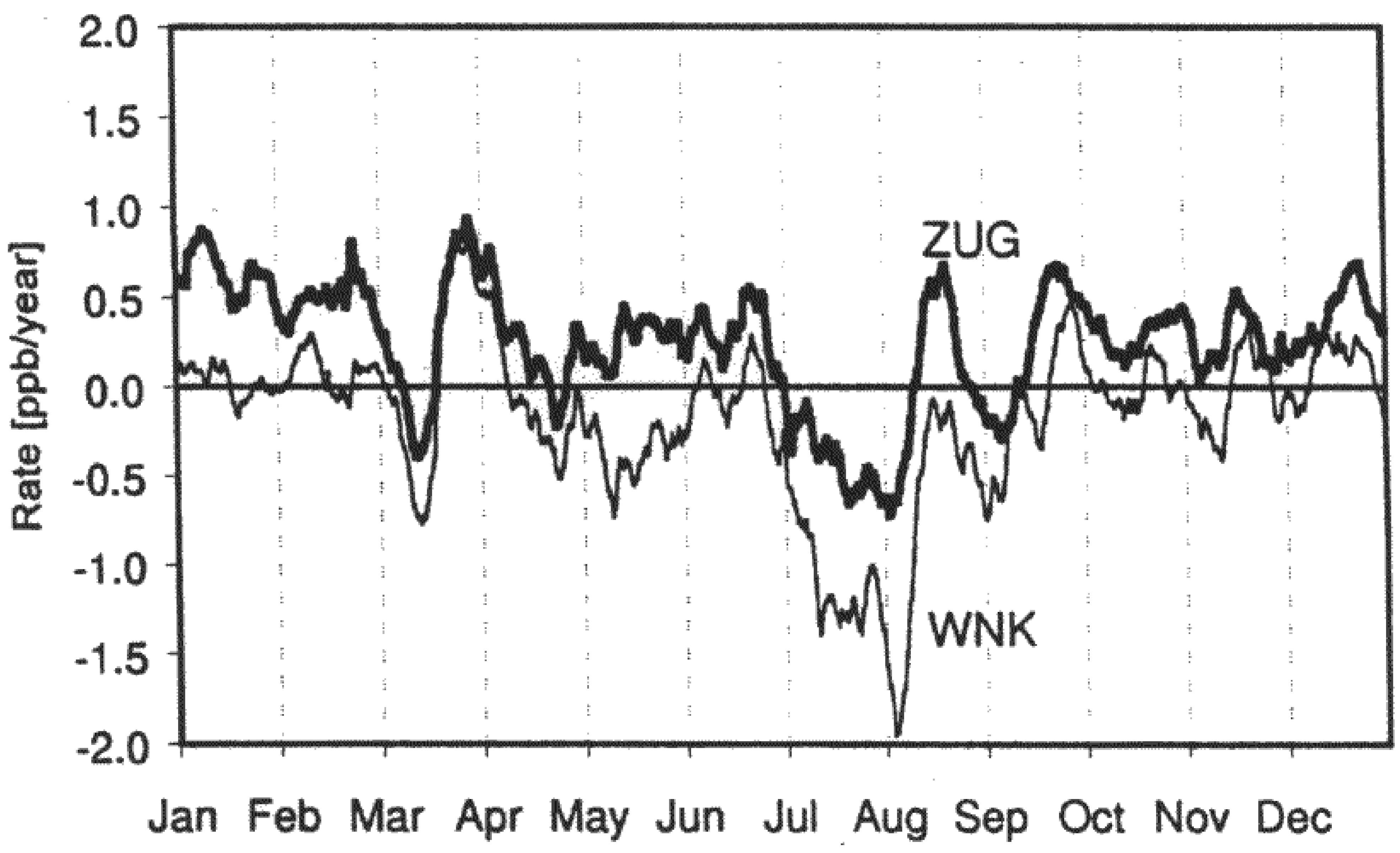

Figure 2. Seasonal distribution of the average O_3 growth rates [ppb yr-1] at Zugspitze and Wank (1990-2001) at a temporal resolution of 11 days.

For filtering of ozone data into the categories 'polluted' (boundary layer transport) and 'unpolluted/less polluted' (background concentrations) at ZUG, different CO cut-off levels were tested. Finally two sets of criteria were used for the climatology. Filter 1: CO > 1.1*11-day moving median; and CO < 0.9*11-day mov. median, respectively. Filter 2: CO > 30-day mov. median; CO < 30-day mov. median. Although less specific, filter 2 has the advantage of a much higher data coverage, yielding more representative O_3 statistics.

Trend estimates for ZUG obtained with filtered data representing different atmospheric conditions are shown in Table 1, where they are compared to the growth rate for unfiltered data of 0.25 ppb per year (1990 – 2000). Some of the trends and small differences in growth rates are lacking statistical significance. However, this point was neglected, since it was the aim to detect certain developments in the atmosphere at an early stage. In addition, in Table 1 growth rates are shown that were obtained with a purely statistical filter. It was intended to model O_3 values representing clean air conditions and polluted air by constructing an annual cycle from the 30th, 50th and 70th monthly percentiles. The ozone levels based on the sequence P70 (Jan, Feb), P50 (Mar, Apr), P30 (May-Aug), P50 (Sep, Oct), P70 (Nov, Dec) in the case of clean air, and P30, P50, P70 etc. for polluted air, were found to be a suitable proxy as was seen from a comparison with the respective CO-filtered ozone concentrations.

Table 1. Summary of growth rates for O_3 at the Zugspitze as obtained with linear regression on monthly mean values.

Atmospheric condition	Data filter	Data coverage	Average rate [ppb yr^{-1}]	Time period
"all"	none	all valid data	0.25	1990 – 2000
Stratospheric influence	RH < 60% AND ^7Be >85th perctl.	7%	0.55	"
Dry air (subsidence)	RH < 60 %	22%	0.37	"
Humid conditions excluded	RH < 90 %	49%	0.15	"
Polluted air, CO filter 1	CO > 1.1*11-day mov. median	19%	0.01	"
Polluted air, CO filter 2	CO > 30-day moving median	38%	0.08	"
Unpolluted air, CO filter 1	CO < 0.9*11-day mov. median	17%	0.34	"
Unpolluted air, CO filter 2	CO < 30-day moving median	37%	0.40	"
"Polluted", statistical filter "Unpolluted", statistical filt.	combination of high/low monthly percentiles	all valid data	0.22 0.33	"
"all"	none	all valid data	0.32	1991 – 2000
Fine weather	visibility > 60 km	34%	0.51	1991 – 2000

The average seasonal cycle of O_3 at the two mountain sites is characterised by lowest concentrations from November to January and highest levels occurring during spring and summer (Fig. 3). A comparison of the first and second part of the time-series did not indicate significant changes in the overall shape of the cycle. In Figure 3 a high-resolution, average annual profile is plotted together with the monthly values. The additional information provided by the finer resolution is particularly valuable for the months April, July and August, when the interannual variability is strongest. The major ozone variations observed at the Zugspitze with a duration of days to weeks are representative of O_3 concentrations over Central Europe. This was concluded from a year-by-year comparisons with the annual ozone profile of Mt. Cimone in Northern Italy (Bonasoni *et al.*, 2000).

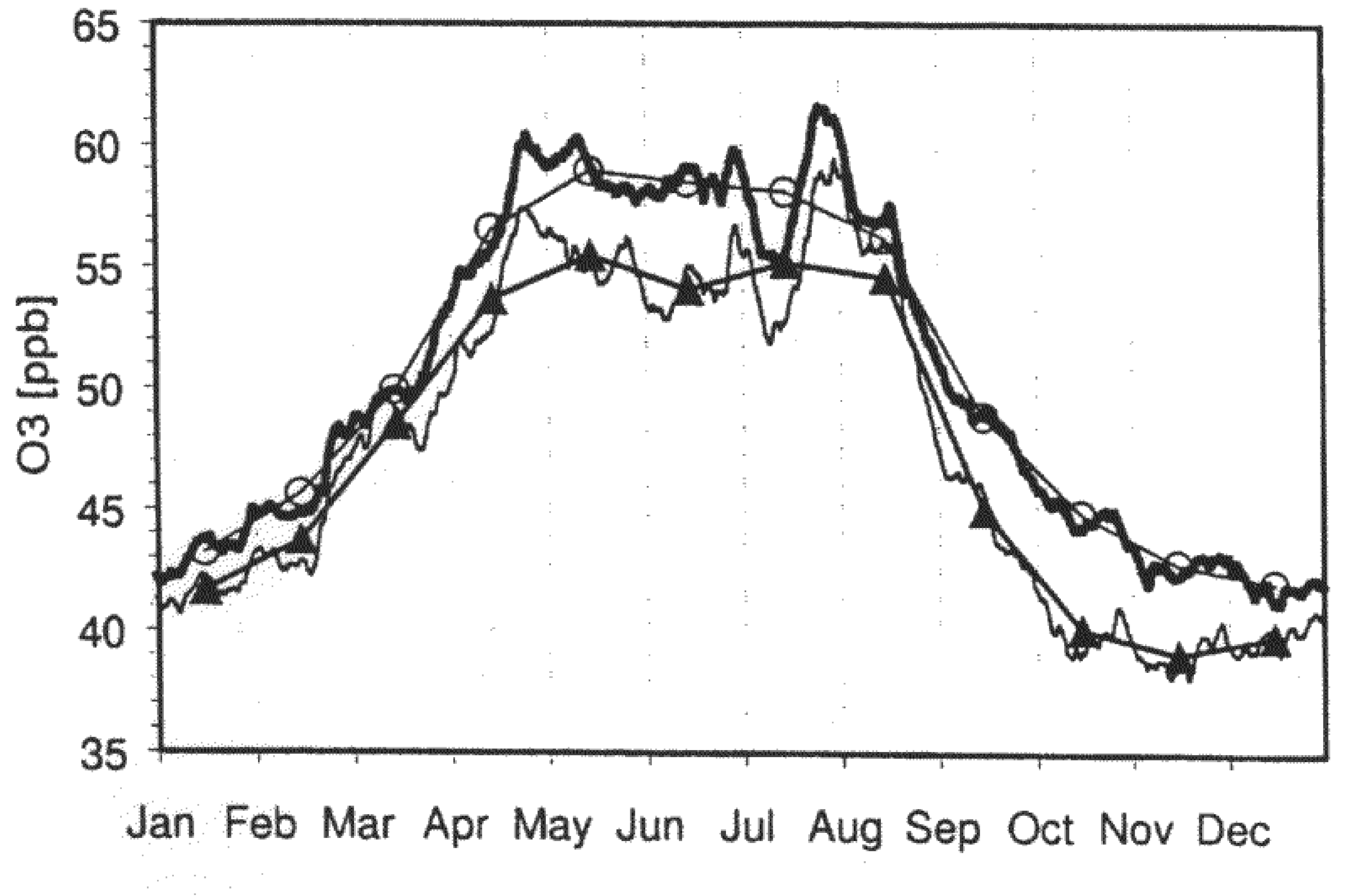

Figure 3. Average seasonal cycle of O_3 (1990-2001) in terms of monthly means and 11-day moving medians for Zugspitze (upper trace) and Wank (lower trace).

Seasonal cycles were determined for a number of atmospheric conditions. In particular data selected by the CO trace gas filter reflect ozone degradation in polluted air in winter and summertime photochemical production (Fig. 4, left). The 'unpolluted' cycle in Figure 4 is regarded as an estimate of background O_3 over Central Europe. When comparing 'polluted' with 'unpolluted' conditions, the effects of O_3 surplus or deficit during different seasons tend to cancel each other partially on an annual scale (Fig. 4, right). However, the quantitative results are dependent on the selected cut-off levels. With the CO filters 1 and 2 (as defined above) the annual averages and sums (in ppb) over the ozone differences of Figure 4 amount to 1.1 and 0.7 (avg., filters 1 and 2) as well as 12.7 and 8.9 (sum, filters 1 and 2, respectively). Seasonal cycles calculated for conditions such as 'dry air' (RH < 60%) or 'fine weather' (visibility > 60 km) have yielded monthly O_3 values that were about 1 – 2 ppb above the all-data level throughout the year. When stratospheric/upper tropospheric influence was selected, the monthly values were roughly 5 ppb higher than the average, with an annual profile of similar shape.

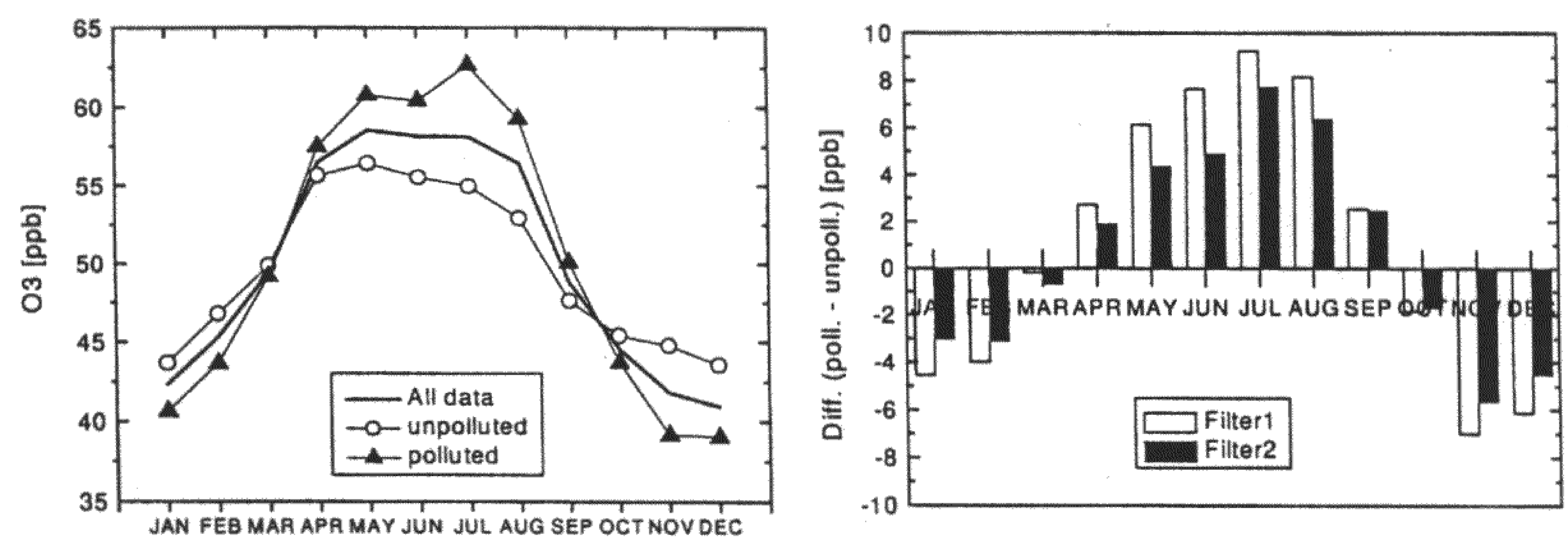

Figure 4. Left panel: Average seasonal cycle of O_3 (Zugspitze, 1990-2000) for 'all data' as well as 'polluted' and 'unpolluted' conditions based on CO filter 2 (for definition see text). Right panel: Difference between the ozone cycles for polluted and unpolluted conditions as obtained with two different cut-off levels (CO filters 1 and 2).

Conclusions

While summertime photochemical O_3 production has been of major importance for the ozone increase during the first part of the Zugspitze and Wank time-series, the highest growth rates of the second part were mainly related to reduced O_3 loss during the winter months. In particular for the Wank site (1780 m), much of the temporal change of the O_3 record is in agreement with a reduction of pollutants. For clean air at Zugspitze the trend is higher than the one determined for unfiltered data, while near-zero growth is indicated for polluted air (CO data filter, 1990-2000). Trend estimates obtained with meteorological data filters (mainly RH and visibility) support the assumption that changes in the advection patterns might have contributed to the Zugspitze O_3 trend over the last decade. Ozone concentrations selected by the stratospheric criterion (using ^7Be and RH) have yielded a rather high growth rate, which, however, is only based on a small monthly data coverage. High and low percentiles on an annual basis, reflecting (roughly) O_3 production in summer and O_3 loss in winter, respectively, show the highest trend for the lower percentiles, thus confirming the contribution of the wintertime concentrations.

A representative seasonal cycle for O_3 of the last decade was determined as well as cycles related to different meteorological and air-chemical conditions. Based on the filtered O_3 data and associated data coverage, the relative contributions of specific conditions to the average ozone concentration could be estimated. In particular, the ozone contribution of stratospheric/upper tropospheric air on an annual scale was calculated to be about 9%.

Increased temporal resolution for the determination of growth rates and seasonal cycles (e.g., 11-day averages) yields valuable additional information. Moreover, such a time scale is well suited for comparisons of major short-term variations between mountain stations in Central Europe. The strong interannual variability of O_3 concentrations observed particularly for spring and summer is related to variations of the duration of different meteorological conditions, but partly is simply due to small time shifts in their occurrence, as seen from the seasonal variations at 11-day temporal resolution. Due to the pronounced year-to-year variability, only after more than ten years of data is the shape of the seasonal cycle largely robust against the addition of another year's data.

Acknowledgements

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