# Trends of ozone and meteorological parameters at high alpine sites

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#### **Abstract**

Climate change and changes in the large scale chemical composition of the atmosphere influence thebiodiversity in and the appearance of protected areas even at remote sites. Data sets from Global Atmosphere Watch (GAW) stations are most appropriate to study background air pollution and meteorological trends. For the alpine sites Sonnblick, Jungfraujoch and Hohenpeißenberg we found increasing ozone concentrations until≈ 2003 especially in winter, whereas in summer there is a small decrease (period 1993-2011). The ozone increase in winter is favoured by changes in the air flow regimes and − to some degree − in an increasing sunshine duration, whereas in summer there are notrends in those meteorological parameters that are relevant for ozone.

# **Keywords**

Climate change, atmospheric trace gases

### Introduction

The alpine sites Sonnblick (3.105 m), Zugspitze (2.962/2.670 m), Hohenpeißenberg (985 m) and Jungfraujoch (3.580 m) contribute to the Global Atmosphere Watch Programme (GAW) of the World Meteorological Organization (WMO). The aim of GAW is to document the large scale chemical composition of the atmosphere and its changes. The analysis of the trace gas concentration trends between 1993 and 2011 shows an increasing trend of ozone until  $\approx$  1999 to 2003, followed by a small decrease afterwards (GILGE et al. 2011). The increase is strongest in winter and spring, whereas during summer, a small decrease is found. On the other hand, the ozone precursors decrease during the whole investigation period, so that the ozone trend cannot be caused by the precursors alone (GILGE et al. 2010).

In this paper we compare the ozone trends at Sonnblick, Jungfraujoch and Zugspitze with trends in the relevant meteorological parameters: Vertical temperature gradient, sunshine duration and air flow regimes for the period from 19930101 to 20111231 exemplified for winter (December to February) and summer (June to August). The analysis of the Zugspitze data is not yet completed.

### **Methods**

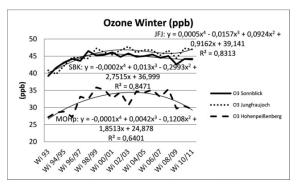
Vertical temperature gradients have been calculated for the valley atmosphere using the temperature data from Rauris (19,5 km north of Sonnblick, 934 m) and Sonnblick (3.105 m); for elevations above Sonnblick we used the temperature data from selected height levels (3.000 m, 5.000 m, 10.000 m and 15.000 m) of the radiosonde soundings at Munich and Vienna. Trends of monthly mean temperatures have been cross-checked with the HISTALP data set (<a href="http://www.zamg.ac.at/histalp/">http://www.zamg.ac.at/histalp/</a>) showing good agreement and a temperature increase for all seasons except winter, wherea temperature decrease is found especially at higher elevations.



Figure 1: Regions of origin of air relevant for the ozone concentrations at the alpine GAW stations. Air masses from the continent are rich, Atlantic air masses poor in ozone (KAISER et al. 2007).

The trend of the sunshine duration has been calculated using the measurements at Sonnblick. The trend of monthly mean sunshine duration at Sonnblick is in good agreement with the sunshine trend for the "Greater Alpine Region (GAR)" from the HISTALP data set and is thus representative for the whole region investigated.

To study the trends of the airflow regimes,4 day back-trajectories are used, calculated with the model FLEXTRA (Stohl 1998), based on the ERA-Interim wind fields of the European Centre for Medium Range Weather Forecast (Dee et al. 2011). The ERA-Interim fields are fields are not subject to model updatesand thus most appropriate for trend studies. The horizontal resolution of ERA-Interim is 1°. Arrival time of the trajectories at Sonnblick is each 3 hours, arrival height 100 m above model topography. Thus the air at Sonnblick is traced back each three hours for 4 days to study its origin. Based on Kaiser et al. (2007)7 regions that are relevant for the ozone concentrations at Sonnblick (and at the other alpine GAW stations) have been defined (Fig. 1). The vertical extent of the regions is from ground up to 2.000 m. An additional category is the "free troposphere" at heights above 2.000 m, covering the whole region. For each trajectory the time it spends within such a region is calculated. Trends of the air flow regimes are derived with the help of monthly mean trajectory residence times for the different regions defined in Fig. 1.



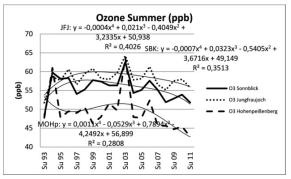
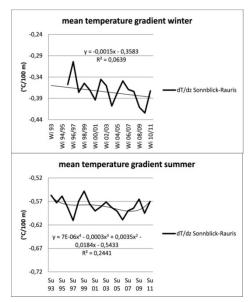


Figure 2: Mean seasonal ozone concentrations at Sonnblick (SBK), Jungfraujoch (JFJ) and Hohenpeißenberg(MOHp) in winter (left) and summer (right) with fitted 4th degreepolynomials.

#### **Results**

Fig. 2 shows seasonal mean ozone concentrations at Sonnblick, Jungfraujoch and Hohenpeißenberg and the respective ozone trends, fitted with a 4th degree polynomial. Consistent with GILGE et al. (2010) the increase of ozone is strongest in winter, whereas in summer the increase is limited to the very beginning of the period and predominantly caused by the low values in summer 1993. Overall there is an increase of ozone in winter, especially at the high elevated sites, but a decrease in summer.

Fig. 3 shows vertical temperature gradients for winter and summer for the valley atmosphere, derived from the temperature data from Rauris and Sonnblick and for the layer above the high alpine sites between 3.000 m and 5.000 m, derived from the radiosonde data from Munich and Vienna. A negative temperature gradient represents temperature decrease with height, a positive gradient temperature increase. Due to its diurnal variation with stable temperature gradients during nighttime (temperature decrease less than -1°C/100 m or even temperature increase with height) and near adiabatic gradients (-1°C/100 m) during the afternoon, monthly mean gradients are stable. Nevertheless, the more negative the mean temperature gradient, the stronger the vertical mixing of the atmosphere.



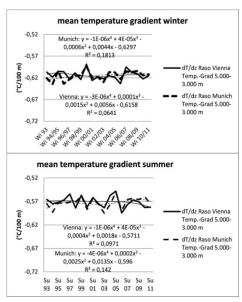
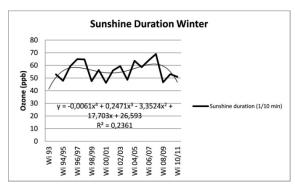


Figure 3: Mean vertical temperature gradient of the valley atmosphere (left) and for the layer between 3.000 m and 5.000 m (right) in winter(top) and in summer (bottom)with a fitted 4th degree polynomial.

Only for the valley atmosphere a relevant trend is found in winter, but much less pronounced in summer, indicating increased vertical mixing of the near ground atmosphere. For the level above the high elevated alpine sites between 3.000 m and 5.000 m, neither the radiosonde data from Munich nor those from Vienna show relevant trends.

Sunshine duration at Sonnblick shows two maxima in winter (from 96/97 to 97/98 and from 06/07 to 07/08) and in summer (94 and 03) as well (Fig. 4). For the whole period there is some increase in winter, but no trend at all in summer. In summer 1993 the sunshine duration is near to its average, the outstanding low ozone values in summer 1993 (Fig. 2) cannot be explained by the sunshine duration.



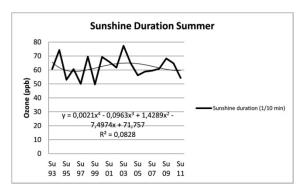
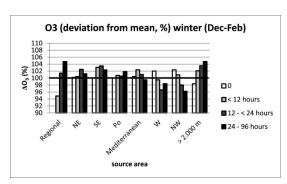


Figure 4: Mean sunshine duration at Sonnblick in winter (left) and summer (right) with a fitted 4th degree polynomial.

The dependence of the ozone concentration at Sonnblick (representative for all sites) from the trajectory residence time in the respective regions (Fig. 1) is shown in Fig. 5. Main sources of elevated ozone concentrations at the alpine GAW stations are air masses from the free troposphere from heights above 2.000 m in winter and air masses from the continent and the Mediterranean in summer whereas Atlantic air masses are relatively poor in ozone over the whole year (see also KAISER et al. 2007): The longer the air stays within these regions, the higher/lower is the ozone concentration. Note that the relations are not always linear.



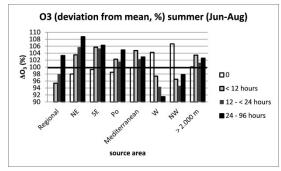


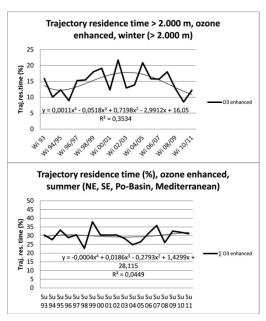
Figure 5: Ozone concentration at Sonnblick (deviation from mean) in dependence of the trajectory residence time (hours) in the respective regions of origin.

The trends of the air flow regimes are shown in Fig. 6. In winter the trend for free tropospheric air masses shows a pronounced maximum near 2003 and air flow from the Atlantic decreases over the whole period. Both processes favour increasing ozone especially until 2003. In summer the trends of the relevant air flow regimes are small.

In 1993 the relevant air flow regimes are close to the average: In winter 1993 air flow from the free troposphere (connected with high ozone concentration) is near to the average, that from the Atlantic (low ozone concentration) is relatively frequent; in summer both air flow regimes connected with enhanced, but also with reduced ozone are close to the average. Thus the outstanding low ozone concentration in 1993 cannot be explained by the airflow regimes.

### **Discussion**

GILGE et al. 2010 assumed that the ozone increase at the alpine GAW stations may be caused by increasing vertical mixing of the atmosphere. Indeed, the valley atmosphere shows a tendency to less stable temperature gradients, i.e. some tendency to increased vertical mixing, especially in winter (Fig. 3). But the correlation between the monthly mean temperature gradient Sonnblick-Rauris and ozone in winteris positive (0.63 at Sonnblick, 0.56 at Jungfraujoch and 0.23 at Hohenpeißenberg) – the more stable the valley atmosphere, the higher the ozone concentration especially at the high elevated sites. This is an effect of large scale anticyclonic weather conditions with relatively high ozone values in the free troposphere above the inversion. On the other hand there is no relevant trend of the temperature gradient at higher elevations, and the correlation with ozone is small (values near to -0.3 for all stations) both in winter and in summer. Thus, the trend found in the temperature gradient of the valley atmosphere would rather contribute to decreasing ozone in winter. The assumption of GILGE et al. (2010) may be important for low situated sites, but must be dismissed for high alpine sites and even for Hohenpeißenberg.



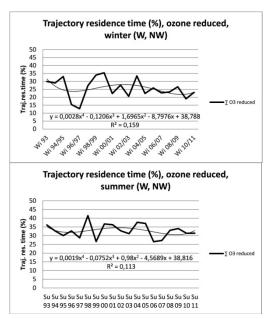


Figure 6: Trajectory residence time for air flow regimes connected with enhanced (left) and reduced ozone concentration (right) at the alpine GAW stations for winter (top) and summer (bottom).

The increase of the sunshine duration in winter (Fig. 4) may contribute to enhanced ozone formation to some degreeeven though photochemical ozone production is small in this season (the correlation is positive at all stations up to 0.5 at Sonnblick). The sunshine peaks in summer 1994 and 2003 are consistent with strong ozone peaks at Hohenpeißenbergwhere the correlation of ozone with the sunshine duration in summer is highest (0.6), whereas the high elevated sites only show ozone peaks in summer 2003. The effects of photochemical ozone production in summer are strongest at Hohenpeißenberg, but even visible at the high elevated sites. The course of the sunshine trend in summer with a maximum in summer 2003 (Fig. 4) is similar to that of ozone especially at Hohenpeißenberg (Fig. 2) indicating some effect of enhanced photochemical ozone production due to the trend of the sunshine duration. But over the whole period of investigation there is no trend of sunshine duration and some decrease of ozone.

The trends of the air flow regimes that are relevant for the ozone concentration at the alpine sites clearly favour an increase of ozone in winter (Fig. 6):The course of the trend for free tropospheric air masses agrees with the course of the ozone trend to a high degree; in addition, air flow from the Atlantic shows a small decrease. Both processes favour increasing ozone. In summer the trends of the relevant air flow regimes are small.

# Conclusion

In this study, the dependence of ozone from the relevant meteorological parameters (vertical mixing of the atmosphere, sunshine duration and air flow regimes) is analysed with the aim to find an explanation for the ozone increase before 2003 which is accompanied with decreasing precursor concentrations. The correlations between these parameters and ozone are relatively small, but one should consider that all these processes go parallel, sometimes enhancing, sometimes compensating each other and the relations are not always linear.

Elevated ozone concentrations at the high alpine sites are connected with large scale anticyclonic conditions with inversions beneath the stations and free tropospheric air above the inversion in winter, enhanced sunshine duration and photochemical ozone production in summer and transport from the continent all over the year. The increase of the sunshine duration and of the transport of air from the continent in all seasons except summerfound in the period from 1993 to 2011favoursan increase of the ozone concentration at the high elevated alpine background sites in winter, spring and autumn. During winter, the trend of transport of ozone-rich air from the free troposphere to the alpine GAW sites is in agreement with the ozone trend, whereas in summer, neither the relevant meteorological parameters, nor the ozone concentrations show significant trends.On the other hand decreasing precursor concentrations (GILGE et al. 2010), but also increasing vertical mixing of the valley atmosphere as found in this study, may contribute to some decrease of ozone as found after  $\approx$  2003 in winter and after 2000 in summer. The estimation of the net effect ofprocesses reducing or increasing the ozone concentration is challenging and will need further investigation.

The outstanding low ozone values in the year 1993 cannot be explained by the meteorological trends and may be a result of the Pinatubo eruption.

# Acknowledgements

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