



Ozone changes at Jungfraujoch (Switzerland) in 1990-2005: Analysis by backward trajectories

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Surface ozone measurements at Jungfraujoch (JFJ) covering the period 1990-2005 are investigated in this study. Increasing trends with the maximum $+0.63 \pm 0.22$ ppb/y in winter and the minimum $+0.4 \pm 0.35$ ppb/y in summer are found. The influences of air masses from various source regions on ozone trends are examined using 10 days backward trajectory analysis. Positive trends in ozone are found for both European planetary boundary layer (PBL) air and background air. The ozone increase in winter in European PBL air is particularly large, most probably in response to the steady decrease in European NO emissions leading to less ozone titration. Summer ozone increase in the air masses advected from the European PBL is small, possibly attributable to the balancing effects of decreasing European ozone precursor emissions and increasing background ozone concentrations. A stronger increase of European PBL air advection is found in the most recent years for all seasons particularly in winter, implying that the exposure time of JFJ site to the free troposphere is decreasing. A first analysis on the variations of PBL height suggests that deepening of PBL height might partly be the reason of more exposure of JFJ to European PBL air. Furthermore, individual source regions of background air are identified using a series criteria applied to trajectories. Ozone increase in the stratospheric intruded air and free tropospheric air are larger than average, indicating their large impact on the overall positive trends. Simple linear trend analysis is applied to ozone measurement data related to the individual source regions, respectively. Strikingly, ozone from all source regions shows an increasing trend throughout 1990-2005 with a similar magnitude. Investigation on the frequency of air mass from the individual source regions shows that European PBL air and free tropospheric air are the two dominating air flow regimes, and the increase on European PBL air is balanced mainly by the free tropospheric air, whose frequency shows an evident decline from about 60% in the earlier 1990s to about 40% in the recent years. It is also found that effect of particular years (El Nino, Mount Pinatubo eruption) do not strongly affect calculated trends. The length of 16 years of ozone measurements is too short for more in depth analysis of potential causes of the trends. The most dominant part of ozone in the air at JFJ originates from the free troposphere, which cannot be traced to any particular origin within the trajectory length of 10 days, therefore changes e.g., in global methane emissions cannot be assessed by the used method, which is also limited by the lack of description of mixing of air masses.

The same method of source regions' classification was applied to ozonesonde measurements of two European ozonesonde stations (Hohenpeissenberg and Payerne, using Brewer Mast (BM) sondes). With respect to the dominating air classes, a large discrepancy is found in the ozone evolution between the two ozonesonde measurements and the surface ozone measurement at JFJ. This suggest that the data quality problem with respect to ozone measurements in the troposphere using Brewer Master sondes remains though in the lowermost stratosphere, the agreement on the ozone anomalies derived from two ozonesondes stations is convincing.