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Seasonal variability of measured ozone production efficiencies in the lower free troposphere of Central Europe

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Abstract. In this study we present the seasonal variability of ozone production efficiencies (E_N), defined as the net number of ozone molecules produced per molecule of nitrogen oxides (nitrogen oxide (NO) + nitrogen dioxide (NO_2)=NO_x) oxidized to NO_z (total reactive nitrogen (NO_y)–NO_x) determined from field measurements of a seven-year period (1998–2004) at the Swiss high-alpine research station Jungfraujoch (JFJ), 3580 m a.s.l. This dataset is a unique long-term data series of nitrogen levels in the free troposphere over Central Europe and hence it offers an excellent opportunity to perform such an analysis and provide further evidence to the photochemical origin of the ozone spring maximum at locations of the northern hemisphere distant from nearby pollution sources. Experimentally derived daily E_N values have been selected for 571 days out of the 2557 days from 1998 to 2004, from which an average ozone production efficiency of 18.8±1.3 molecules of O_3 produced per molecule of NO_x oxidized was calculated. This value indicates the great potential and importance of photochemical ozone production in the free troposphere. The monthly means of experimentally derived daily E_N values show a seasonal variation with lower values from May to August, which can be probably attributed to more efficient vertical transport of polluted air masses from the atmospheric boundary layer up to JFJ. In agreement, theoretically derived monthly E_N values show similar seasonal variation. The ratio NO_y/CO, a parameter to assess the aging process that has occurred in an air parcel, was used as a criterion to disaggregate the 571 selected days between undisturbed and disturbed free tropospheric (FT). The monthly means of experimentally derived E_N values for the undisturbed FT conditions show a distinct seasonal cycle with higher values in the cold season from November to April. The E_N values for undisturbed FT conditions are particularly higher than the respective monthly E_N values for disturbed FT conditions from February to October. It should be noted that the monthly E_N values of March (E_N=35.8) and April (E_N=34.9) are among the highest values throughout the year for undisturbed FT conditions at JFJ. These results highlight the key and possibly the dominant role for photochemistry in the observed build-up of tropospheric ozone in the

winter-spring transition period.

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