

# Supplementary material of “50 years of balloon-borne ozone profile measurements at Uccle, Belgium: short history and contributions in understanding the vertical ozone distribution”

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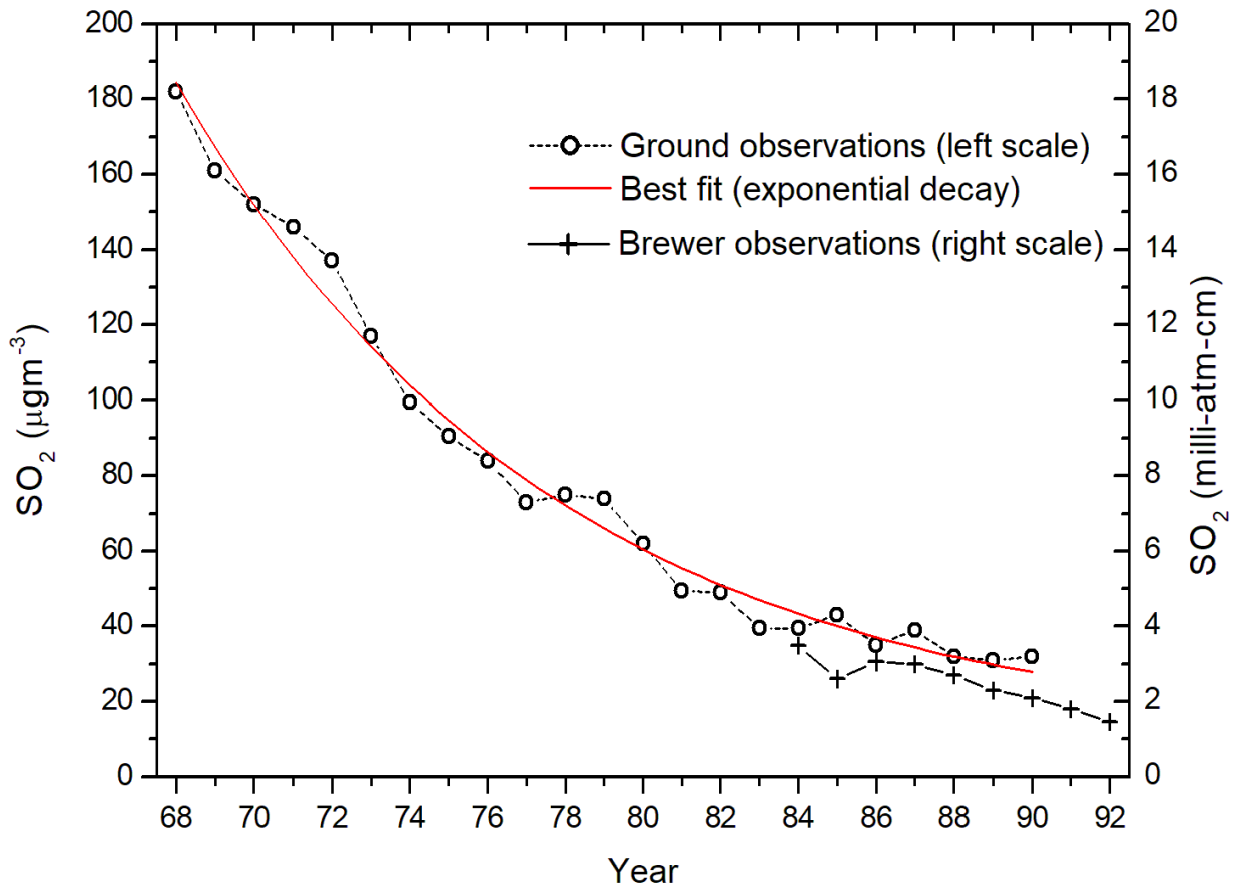
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**Figure S1:** Variation of annual mean values of the SO<sub>2</sub> density in the urban area of Brussels from 1968 to 1992 with the best fitting exponential curve and variation of annual mean values of the reduced SO<sub>2</sub> amount measured with Brewer spectrophotometer 16 at Uccle since 1984 [fig. adapted from De Muer and De Backer, 1993].

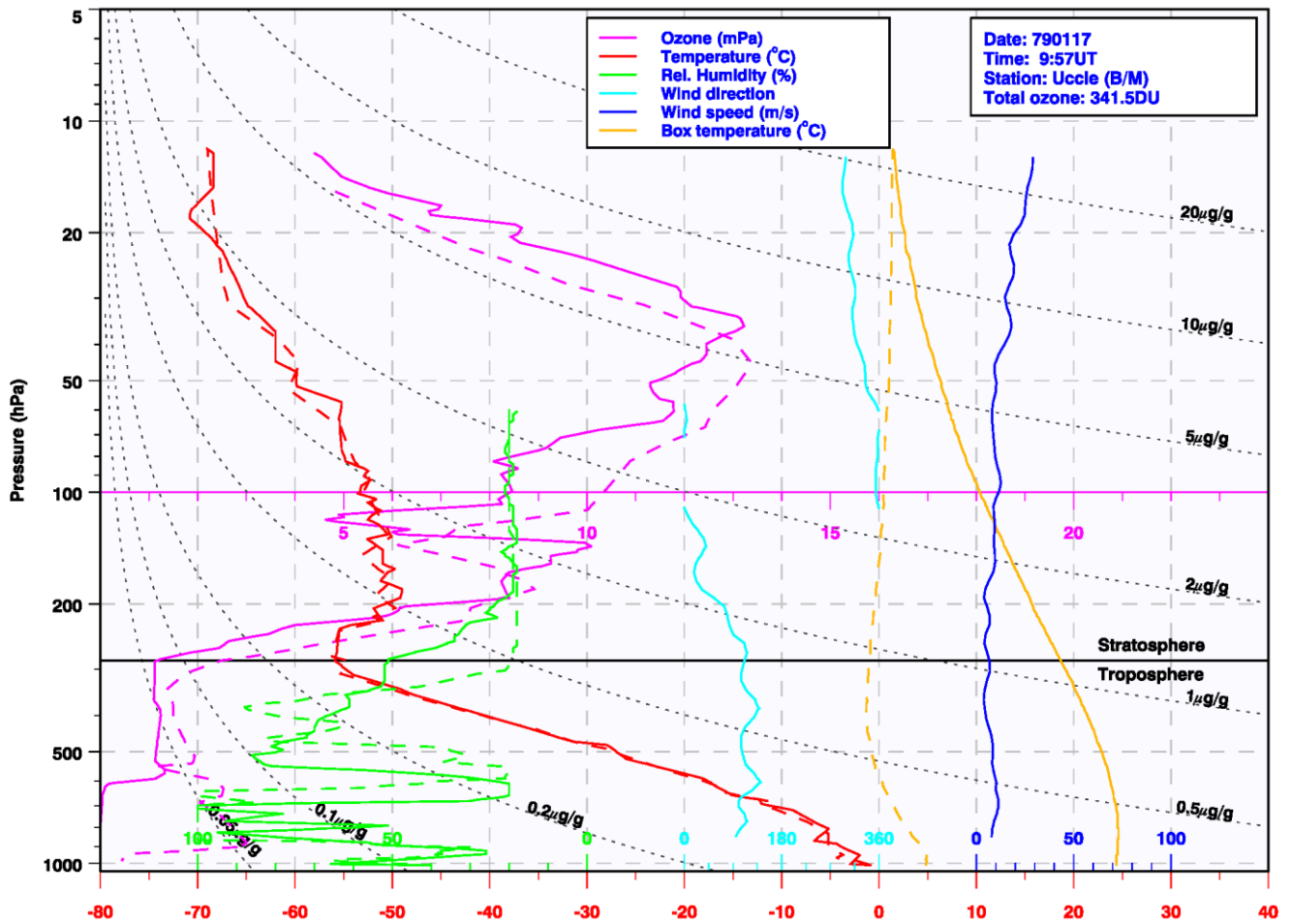


Figure S2: Example of an Uccle ozone profile, on 17 January 1979, in which the effect of the SO<sub>2</sub> interference with the ozone measurements during the ascent of the ozonesonde (full lines) is clearly visible in the boundary layer up to the lower troposphere at about 600 hPa. In the descent ozone profile (dashed lines), the SO<sub>2</sub> interference effect is absent until just before the landing. Please note that the method for deconvolution has not been applied to the ozone profile here.

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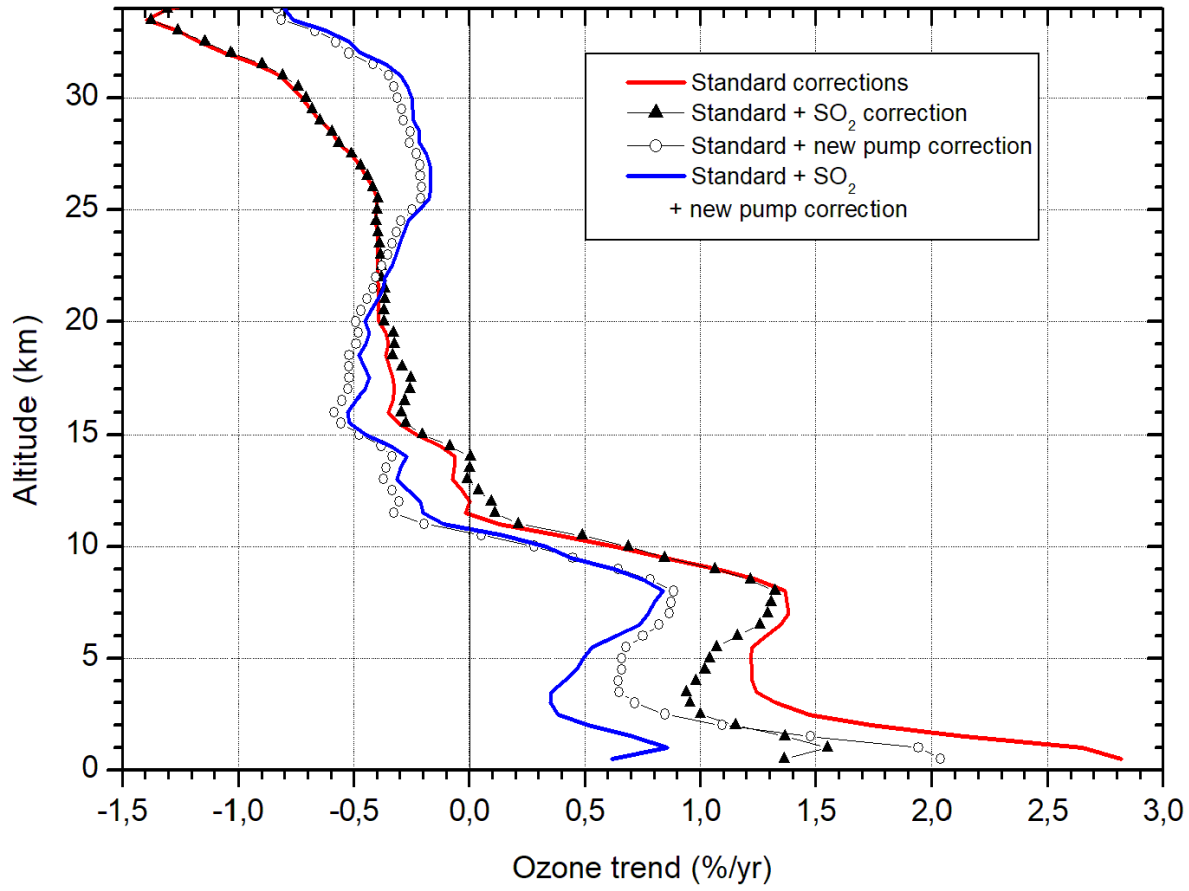
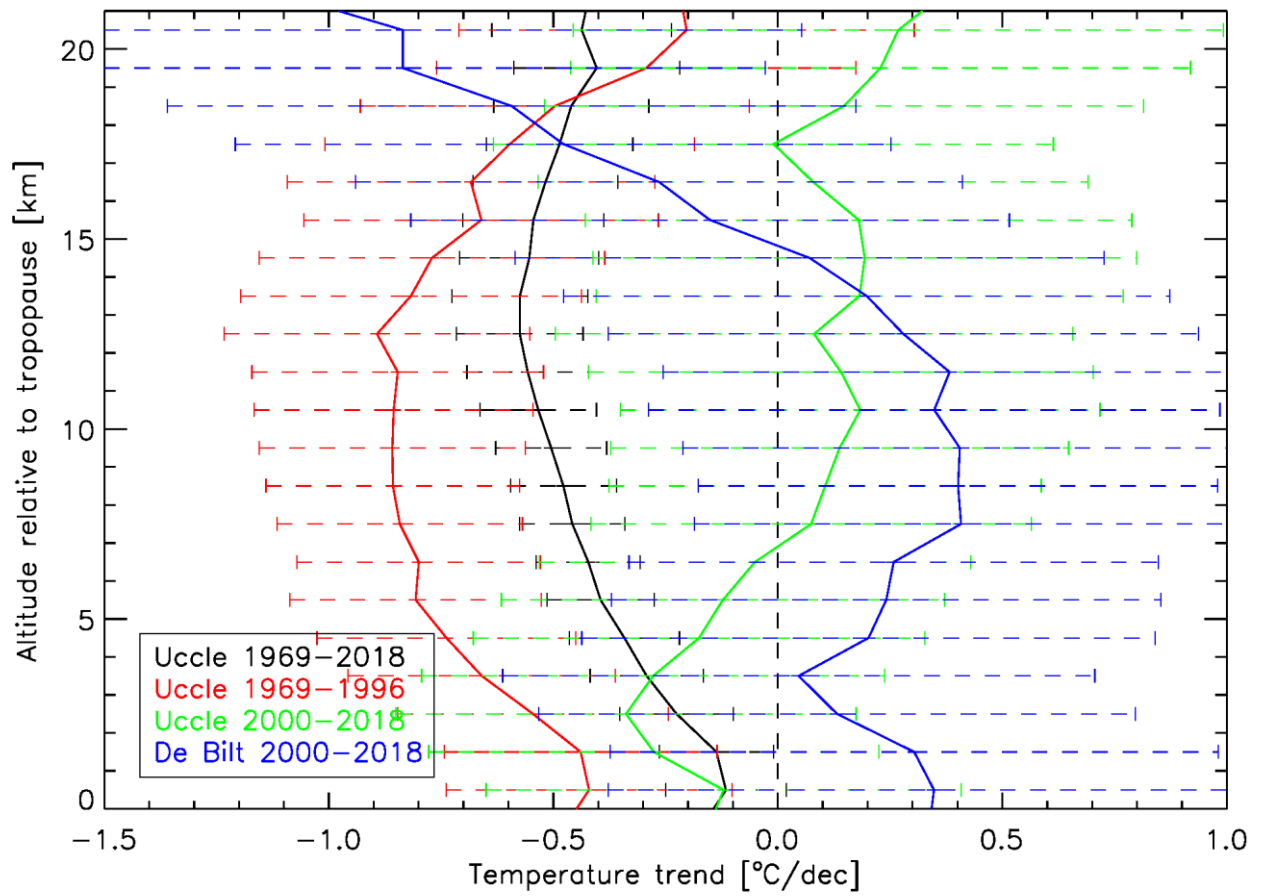
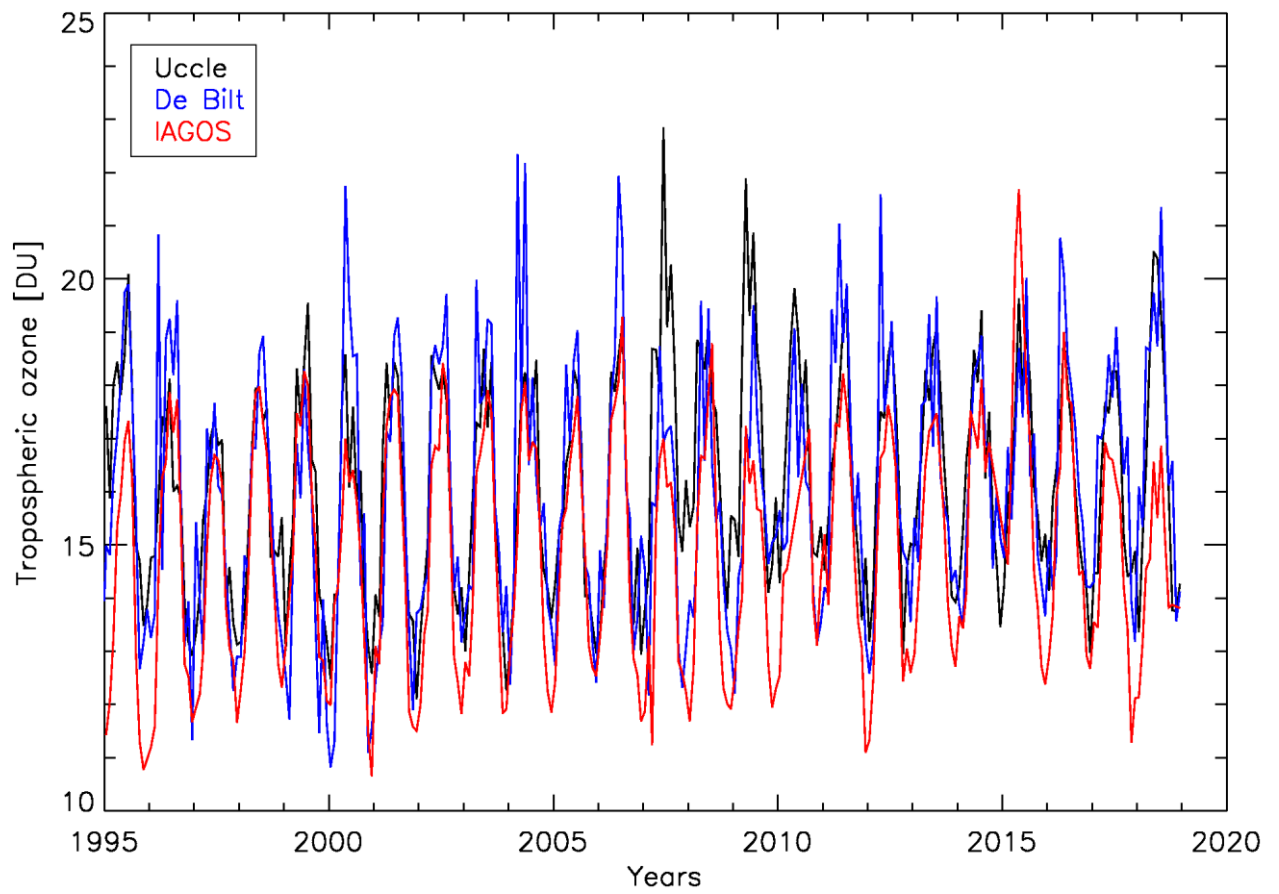


Figure S3: Profiles of calculated ozone trends at Uccle for the period 1969-1996 for different combinations of corrections applied to the BM ozonesonde time series. The “new pump correction” in the legend is described in Sect. 2.2.3.



**Figure S4: Vertical trends of stratospheric temperature trends at Uccle for different periods and at De Bilt (2000-2018). The trends are estimated from a simple linear regression of the monthly anomaly temperature time series, for layers of 1 km height, relative to the tropopause height. The error bars denote the  $2\sigma$  standard errors of the linear regression slope determination.**



**Figure S5: Monthly means of the 3-8 km column tropospheric ozone amounts from the Uccle and the De Bilt ozonesonde profiles and the MOZAIC/IAGOS ascent and descent profiles over Frankfurt airport for the period 1995-2018.**

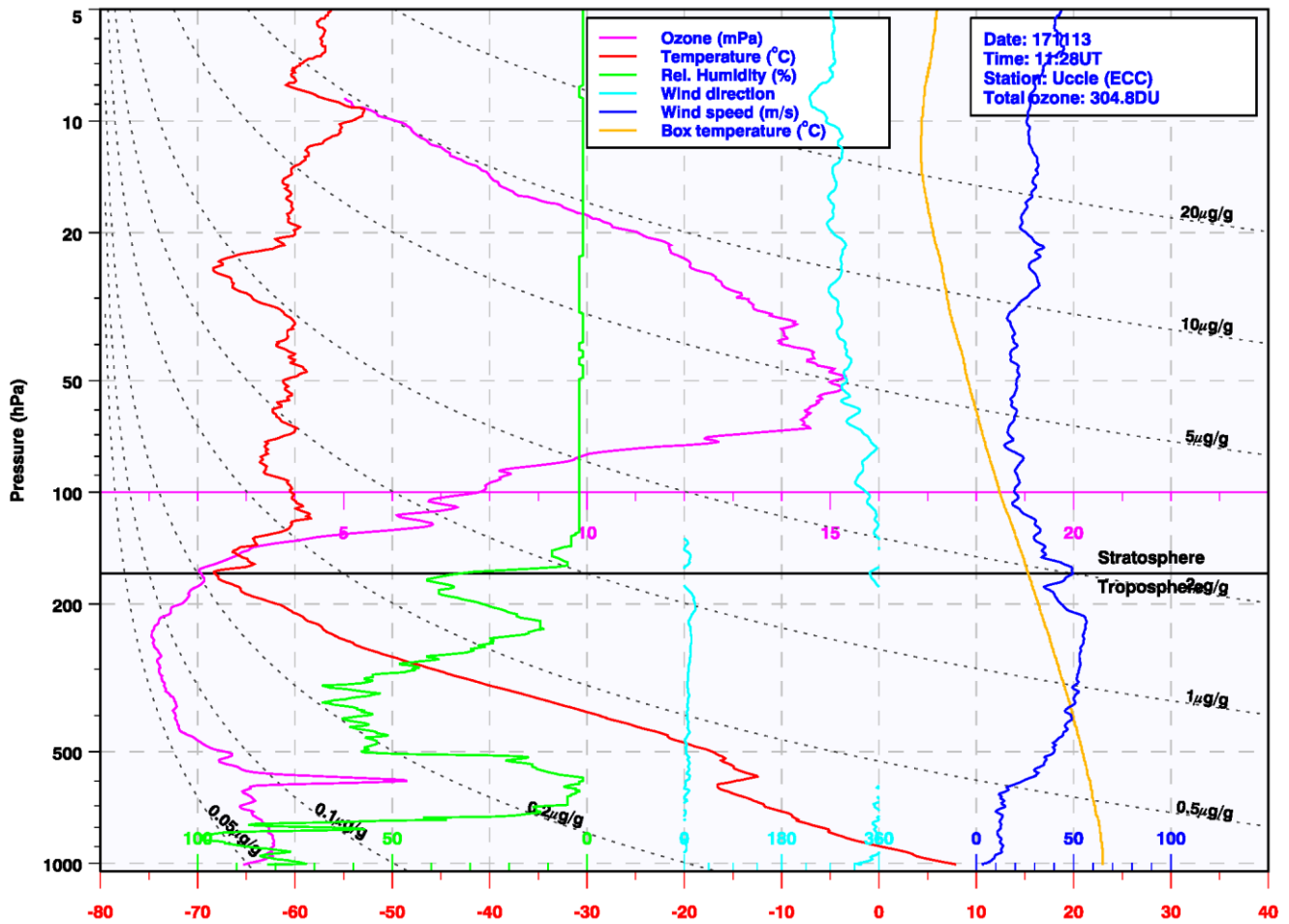


Figure S6: Example of a tropopause fold around 600 hPa in the Uccle ozone sounding on 13 November 2017. This tropopause fold has been identified based on (i) its pronounced ozone maximum (in magenta; exact criterion: ozone mixing ratio enhanced by at least 25% compared to the climatological mean and compared to adjacent minima in the ozone profile), (ii) its very low relative humidity (in green; below 25%), (iii) its high vertical stability (see the thermal inversion in red; vertical potential temperature gradient larger than 11.5 K/100 hPa, its location (iv) in the vicinity of an upper tropospheric jet stream (in blue; wind speed higher than  $20 \text{ m s}^{-1}$ ), and (v) within an upper level front (also in blue; strong vertical wind shear above  $5 \text{ m s}^{-1} \text{ km}^{-1}$  over at least 2 km in the folding region).

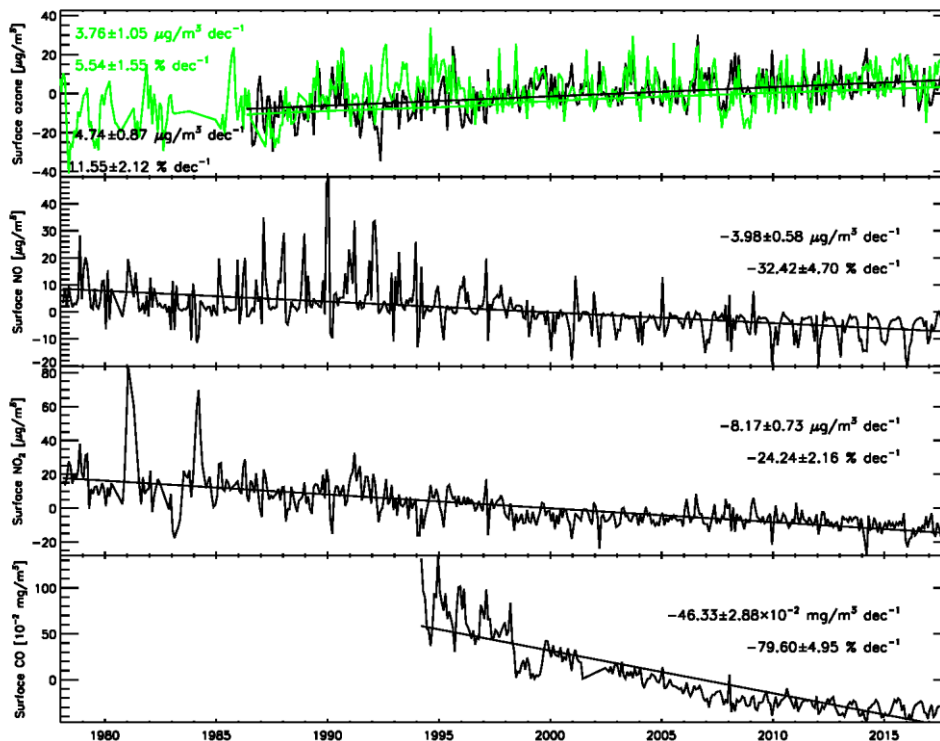


Figure S7: Monthly anomaly time series of Uccle surface ozone (upper panel, black) and mean ozone in the lowest 1 km above Uccle from the ozonesonde launches (upper panel, green) and ozone precursor measurements at Uccle (NO, NO<sub>2</sub>) and Elsenne (CO, 5 km from Uccle). Linear trends are shown, together with the absolute and relative trend estimates, with their 2 $\sigma$  uncertainties.

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### Change in O<sub>3</sub> percentiles Uccle

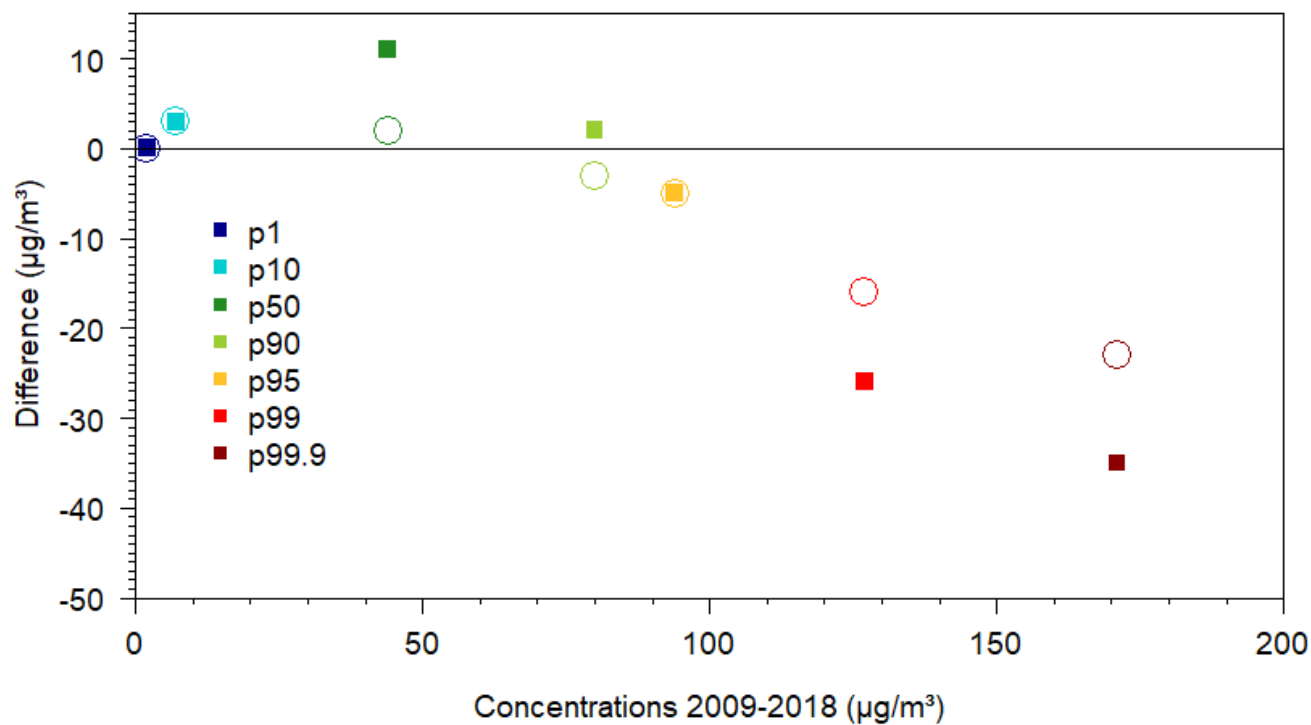
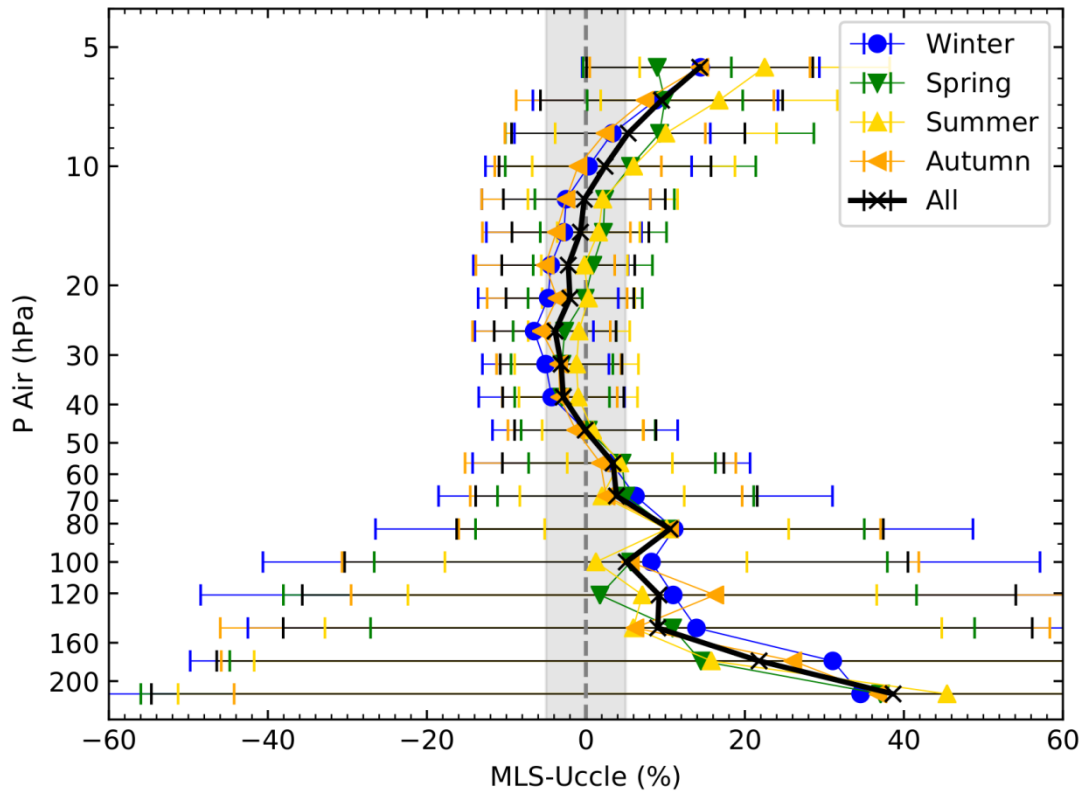


Figure S8: Differences between the surface ozone concentration percentiles at Uccle between the periods 1990-1999 and 2009-2018 (filled squares) and between the periods 2000-2009 and 2009-2018 (open circles), as a function of the 2000-2018 ozone concentrations.

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**Figure S9: Relative ozone profile differences between MLS and Uccle ozonesonde for each season. The different colors correspond to each season and the black line to the overall relative difference. The error bar is only shown for the overall and corresponds to 1 standard deviation.**

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