New version of Fig. 1

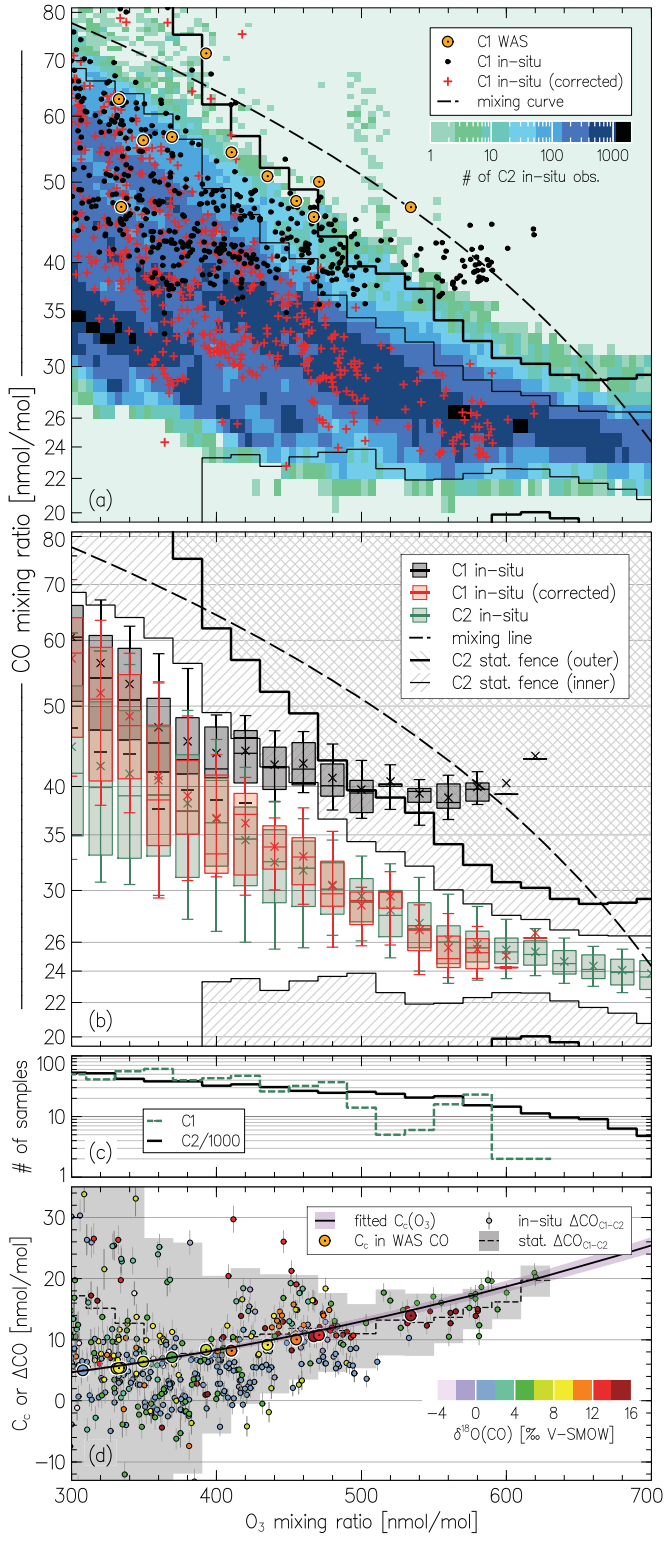

Fig. 1. (a) Distribution of CO mixing ratios as a function of concomitant O_3 mixing ratios measured by CARIBIC in the LMS ([O_3]>300 nmol/mol). The shaded area is the two-dimensional histogram of the C2 measurements (all C2 data obtained until June 2013) counted in 5×1 nmol/mol size [O_3]×[CO] bins, thus darker areas emphasise greater numbers of particular CO–O_3 pairs observed. Small symbols denote the original C1 *in situ* measurements (black) and corrected for the artefacts (red); the C1 WAS analyses (11 of total 408) are shown with large symbols. Thin and thick step-lines demark the inner and outer statistical fences (ranges outside which the data points are considered mild or extreme outliers, see text) of the C2 data, respectively. The dashed curve exemplifies compositions expected from the linear mixing of very different (*e.g.*, tropospheric and stratospheric) end-members. (b) Statistics on CO mixing ratios from C1 and C2 data shown in box-and-whisker diagrams for samples clustered in 20 nmol/mol O_3 bins (whiskers represent 9^{th}/91^{st} percentiles). (c) Sample statistic for each CARIBIC dataset (note the C2 figures scaled down by a factor of 1000). (d) Estimates of the C1 *in situ* CO contamination strength C_c as a function of [O_3] (solid line) obtained by fitting the difference ΔCO between the C2 and C1 *in situ* [CO] (small symbols) in the kinetic framework (see Appendix A, Eq. (A1)). Step line shows the ΔCO for the statistical averages (the shaded area equals the height of the inner statistical fences of the C2 data). Large symbols denote the estimates of C_c in the C1 WAS data (slight variations *vs.* the *in situ* data are due to the sample mixing effects, see Sect. 3). Colour denotes the respective C1 WAS $\delta^{18}O(CO)$ (note that typically 6–7 *in situ* measurements correspond to one WAS sample). Note: The entire C1 CO/O_3 dataset is presented in the Supplementary Material, Fig. S1.

Updated Fig. 2

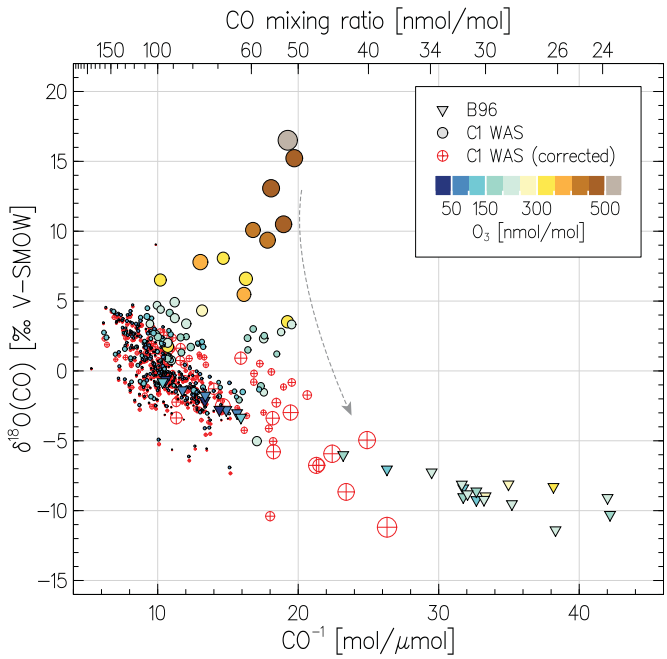

Fig. 2. $^{18}O/^{16}O$ isotope composition of CO as a function of its reciprocal mixing ratio. Triangles present the data from the remote SH UT/LMS obtained by Brenninkmeijer *et al.* (1996) (B96). Colour refers to the concomitantly observed O_3 abundances; note the extremely low [O_3] encountered by B96 in the Antarctic "ozone hole" conditions. Filled and hollow circles denote the original and corrected (as exemplified by the dashed arrow) C1 WAS data, respectively, with the symbol size scaling proportional to the estimated contamination magnitude (see text).

Supplementary Material for the ACP manuscript:

An estimation of the $^{18}O/^{16}O$ ratio of UT/LMS ozone based on artefact CO in air sampled during CARIBIC flights

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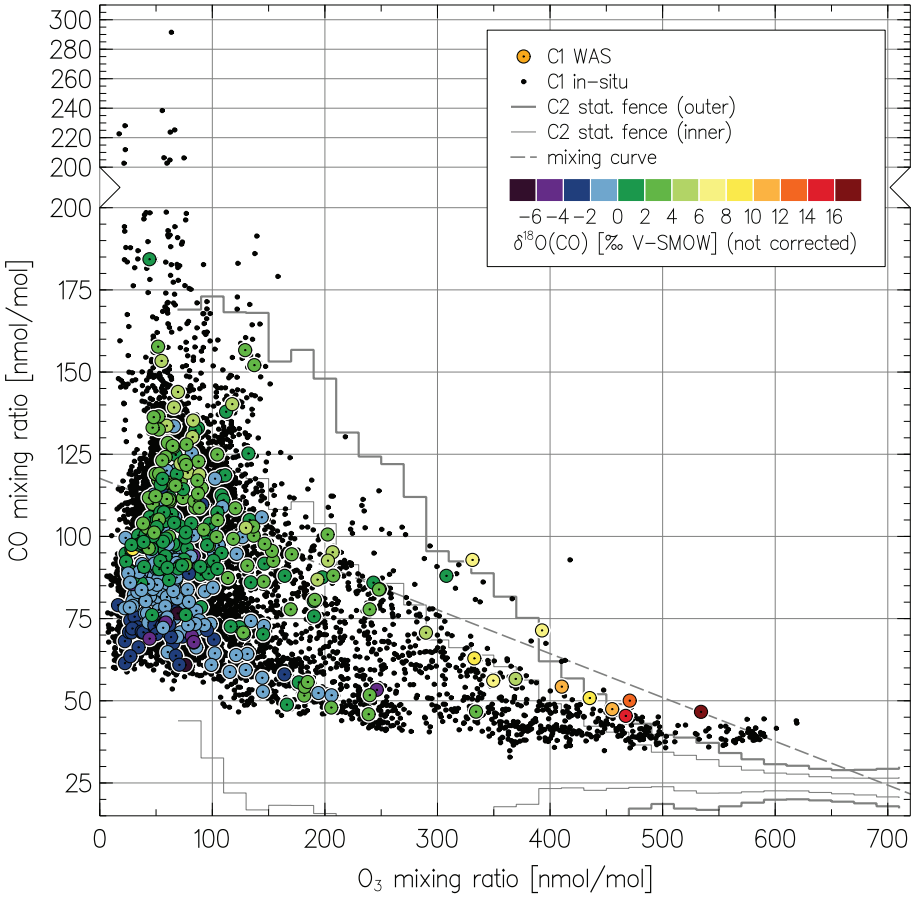

Fig. S1. (*accompanies Fig. 1 of the manuscript*) Carbon monoxide and ozone mixing ratios measured in C1. Small black symbols denote the C1 *in-situ* measurements (n = 12753). The C1 WAS analyses (n = 408) are shown with large symbols; colour denotes the concomitant $\delta^{18}O(CO)$ measurements. Thin and thick step-lines denote the inner and outer statistical fences (ranges outside which the data points are considered mild or extreme outliers) of the C2 data, respectively. The dashed curve exemplifies compositions expected from the linear mixing of very different (*e.g.*, tropospheric and stratospheric) end-members.

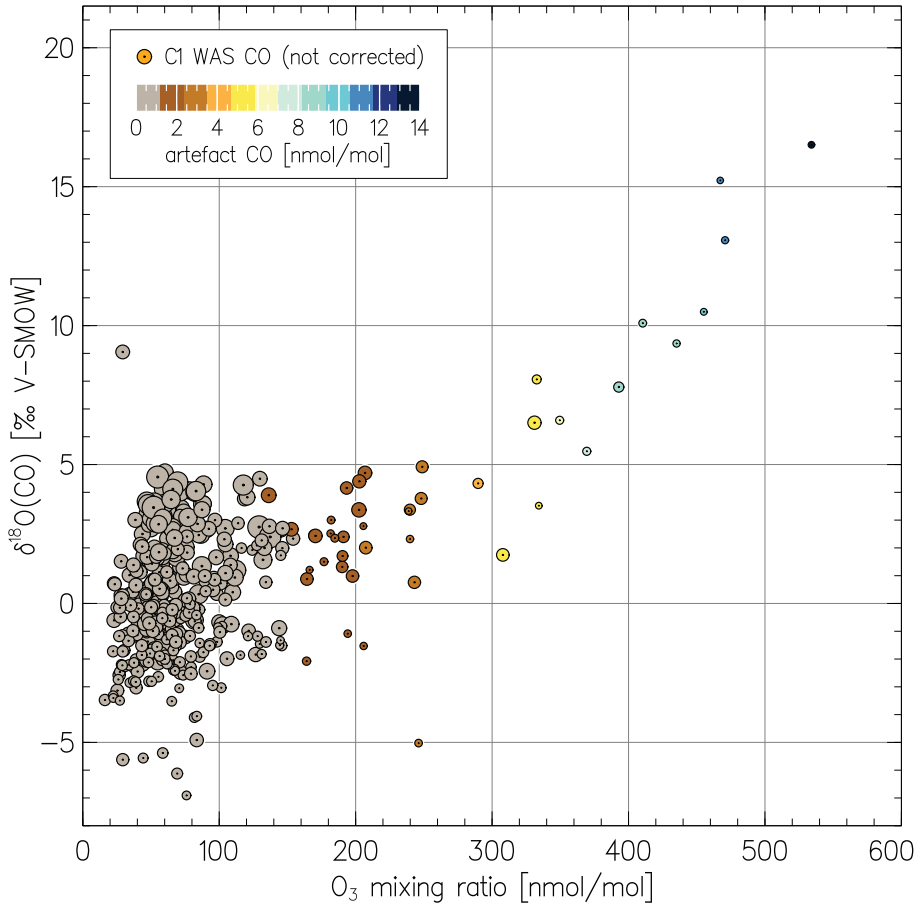

Fig. S2. Measured C1 WAS $\delta^{18}O(CO)$ (not corrected for artefacts) as a function of concomitant O_3 mixing ratio. Symbol colour denotes the artefact CO component (integral C_c per each WAS); symbol size scales proportionally to the WAS CO mixing ratio corrected for artefacts.

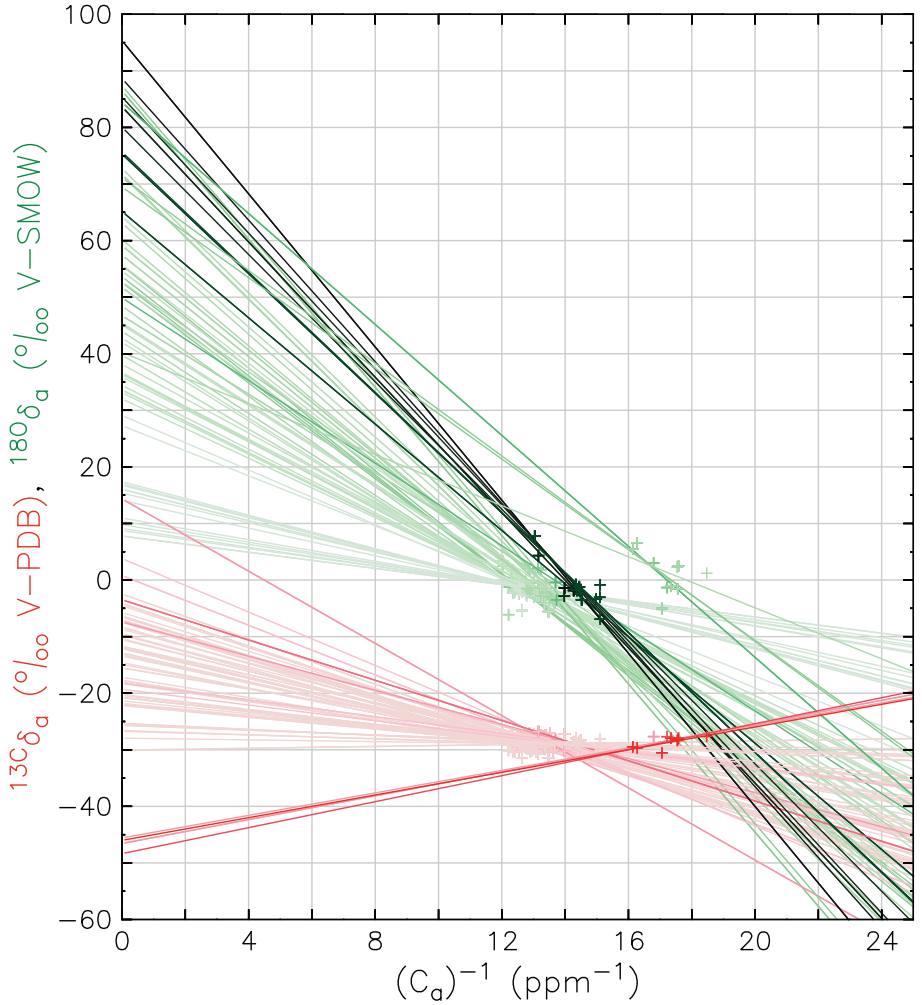

Fig. S3. (*accompanies Fig. 3 of the manuscript*) Keeling plot of the data used in the calculations with the mixing model (MM). The C1 WAS isotope CO measurements are shown with symbols, solid lines denote the linear regressions through the various sets of samples selected by the MM (n = 80 sets are plotted). Colours refer to the $\delta^{13}C$ (red) and $\delta^{18}O$ (green) data, colour intensity indicates the coefficient of determination (R^2) of each regression, respectively. Darker colours denote higher R^2 values, with maxima of 0.92 for $\delta^{18}O$ and 0.54 for $\delta^{13}C$ data, respectively. The inferred contamination signatures ($^i\delta_c$) are found at $(C_a)^{-1} \rightarrow 0$. Regression uncertainties are shown in Fig. 3 of the manuscript. Note that because different subsets of samples contain same data points, some of the symbols are plotted over (*i.e.*, not all symbols contributing to a particular regression case may be seen).

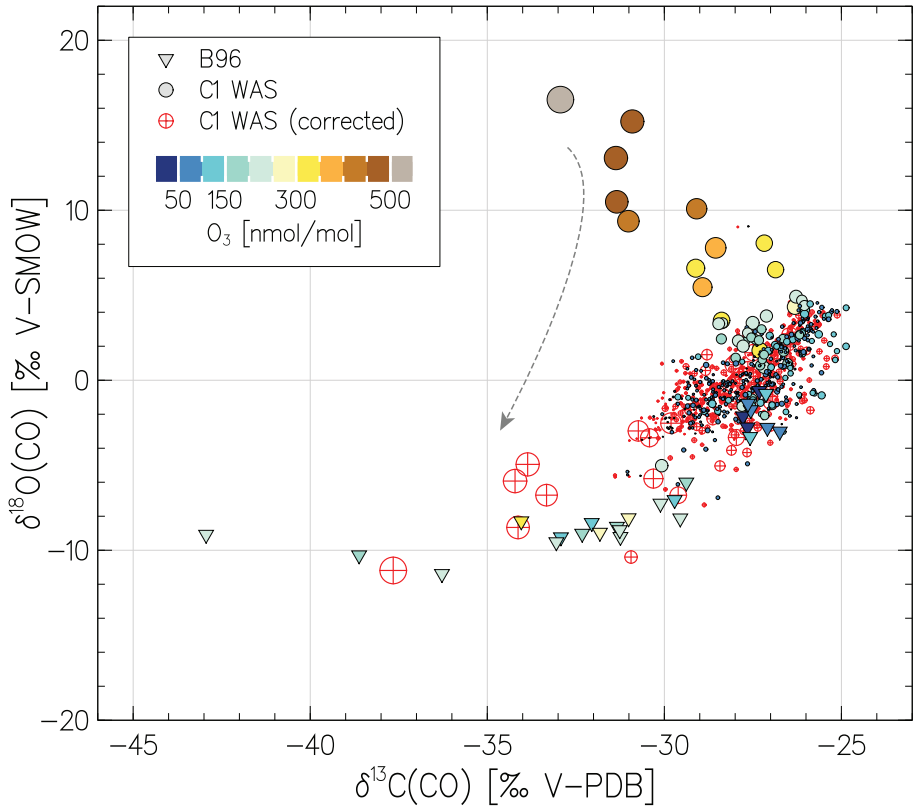

Fig. S4. $^{18}O/^{16}O$ and $^{13}C/^{12}C$ isotope composition of CO measured in C1. Triangles present the data from the remote SH UT/LMS obtained by Brenninkmeijer *et al.* (1996) (B96). Colour refers to the concomitantly observed O_3 abundances; note the extremely low [O_3] encountered by B96 in the Antarctic ozone-hole conditions. Filled and hollow circles denote the original and corrected (as exemplified by the dashed arrow) C1 WAS data, respectively, with the symbol size scaling proportional to the estimated contamination magnitude (see the manuscript for details).

References

Brenninkmeijer, C. A. M., Müller, R., Crutzen, P. J., Lowe, D. C., Manning, M. R., Sparks, R. J., and van Velthoven, P. F. J.: A large ^{13}CO deficit in the lower Antarctic stratosphere due to "Ozone Hole" Chemistry: Part I, Observations, *Geophys. Res. Lett.*, **23**, 2125–2128, doi: 10.1029/96gl01471, 1996.