S1 Retrieval

Table S1. NO_2 and NO retrieval parameters including interfering species, a priori profiles and line lists used for retrieval with PROFFIT96 v2022 code.

	NO_2 (Garmisch)	NO_2 (Zugspitze)	NO (Zugspitze)
spectral micro window	2914.30 – 2914.85 cm^{-1}		1899.90 – 1900.10 cm^{-1}
species retrieved via vmr profile inversion	NO_2 (mean DOFS = 1.48), CH_4 (mean DOFS = 2.78)	NO_2 (mean DOFS = 1.38), CH_4 (mean DOFS = 2.68)	NO (mean DOFS = 2.14), CO_2 (mean DOFS = 1.48)
species retrieved via vmr profile scaling	H_2O, O_3, H_2CO, OCS, C_2H_6		O_3
species included in the forward calculation (not retrieved)	-		H_2O, N_2O
spectroscopy	ATMOS version 20200512		HITRAN version 2020
vertical profile resolution	47 levels (0.745 km to 120 km)		43 levels (2.964 km to 120 km)
a priori profile NO_2 / NO	WACCM V6		
a priori profile CH_4 / CO_2	gfit daily		
a priori profile other interfering species	gfit daily		
T,p profiles	National Centers for Environmental Prediction (NCEP)		
NO_2 / NO regularization	Tikhonov L_1		
CH_4 / CO_2 regularization	Tikhonov L_1		
fit of solar spectrum	yes		
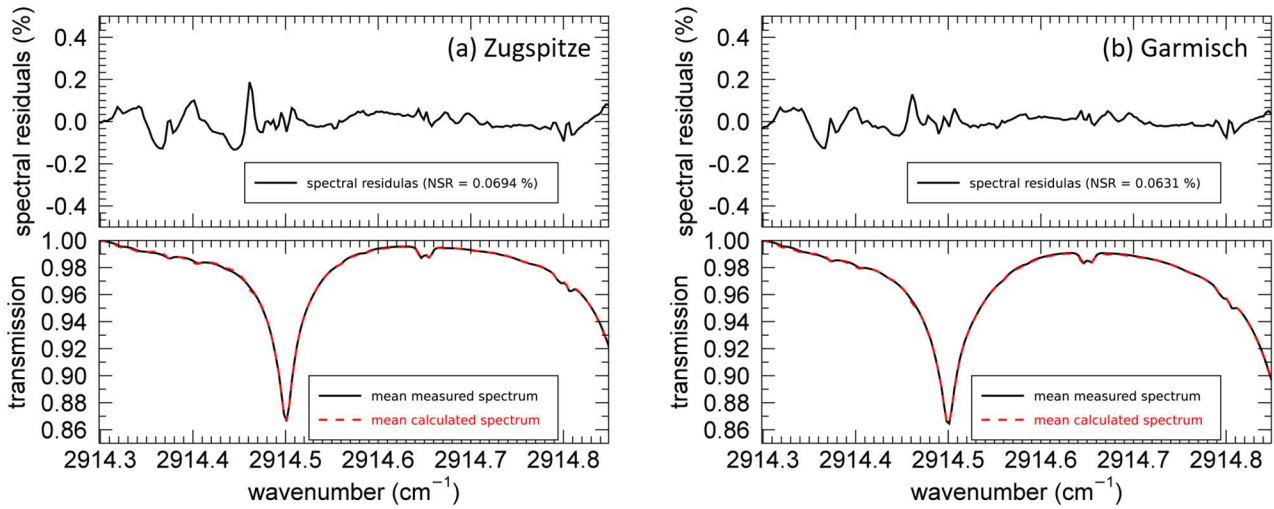
Figure S1. Top row: resulting mean spectral residuals in dependency of the wavenumber of the retrieval of NO_2 at Zugspitze (a) and Garmisch (b). Additionally, the noise-to-signal ratio (NSR) of the mean spectral residuals is given in the legend. Bottom row: mean measured (black lines) and calculated (red lines) spectra in dependency of the wavenumber in the given micro-window of the retrieval of NO_2 at Zugspitze (a) and Garmisch (b).
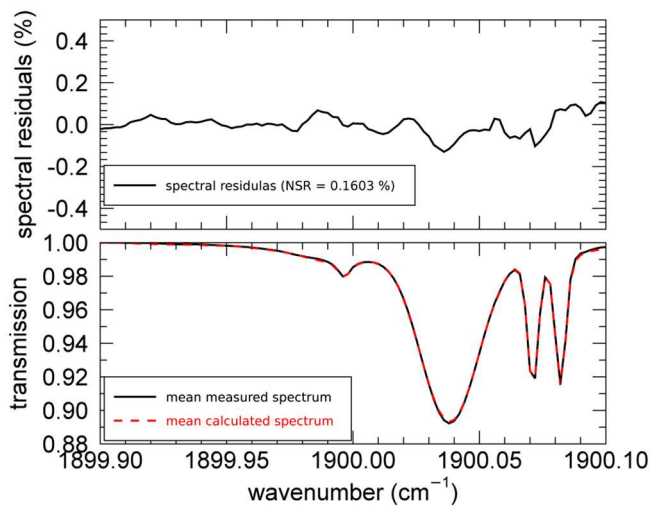
Figure S2. Top row: resulting mean spectral residuals in dependency of the wavenumber of the retrieval of NO at Zugspitze. the noise-to-signal ratio (NSR) of the mean spectral residuals is given in the legend. Bottom row: mean measured (black lines) and calculated (red lines) spectra in dependency of the wavenumber in the given micro-window of the retrieval of NO.

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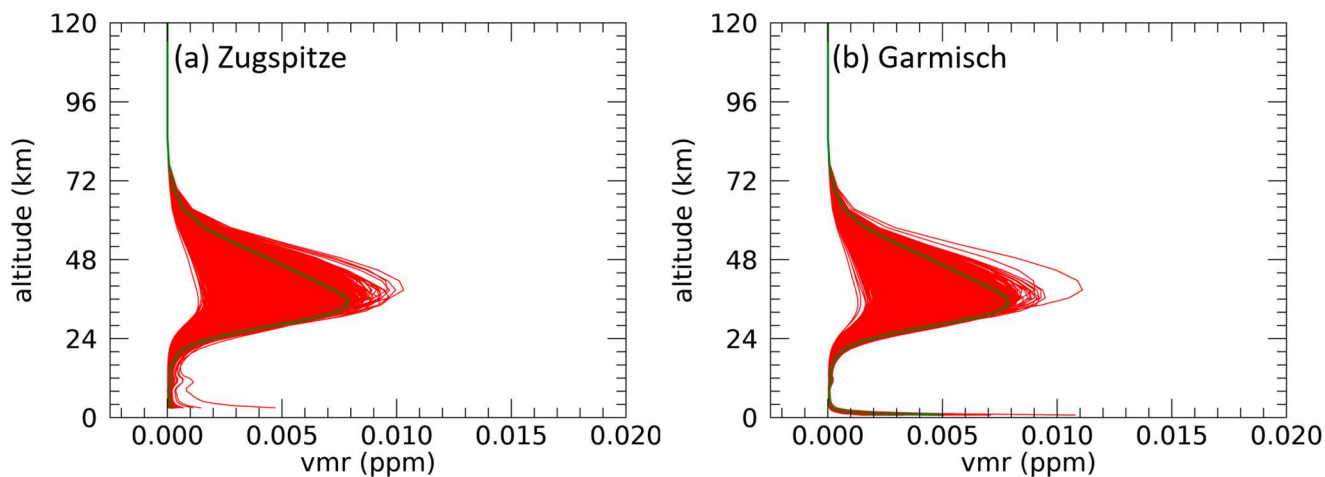
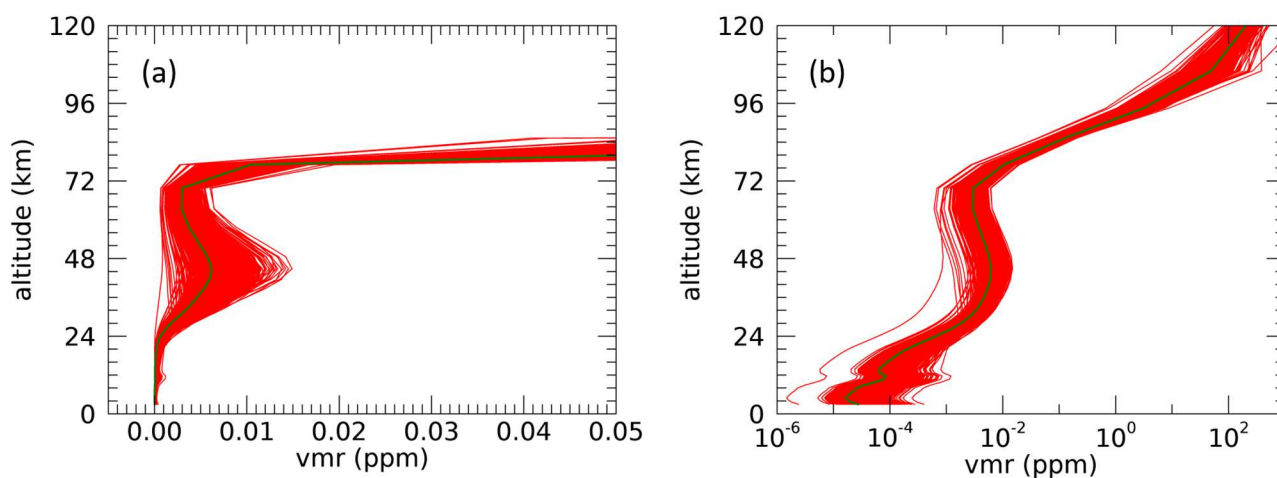


Figure S3. Retrieved NO_2 vmr profiles from a) 1995 to 2022 above Zugspitze and b) 2004 to 2022 above Garmisch (after quality control; red lines). Additionally, the a priori profile (green line) is shown.

20 **Figure S4.** All retrieved NO profile families from 1995 to 2022 at Zugspitze as vmr profile against altitude after the quality control (red lines, see text for details). Additionally, the used a priori profile (green line) is shown; a) linear *x*-axis, b) logarithmic *x*-axis.

S2 NOₓ columns and pollution correction

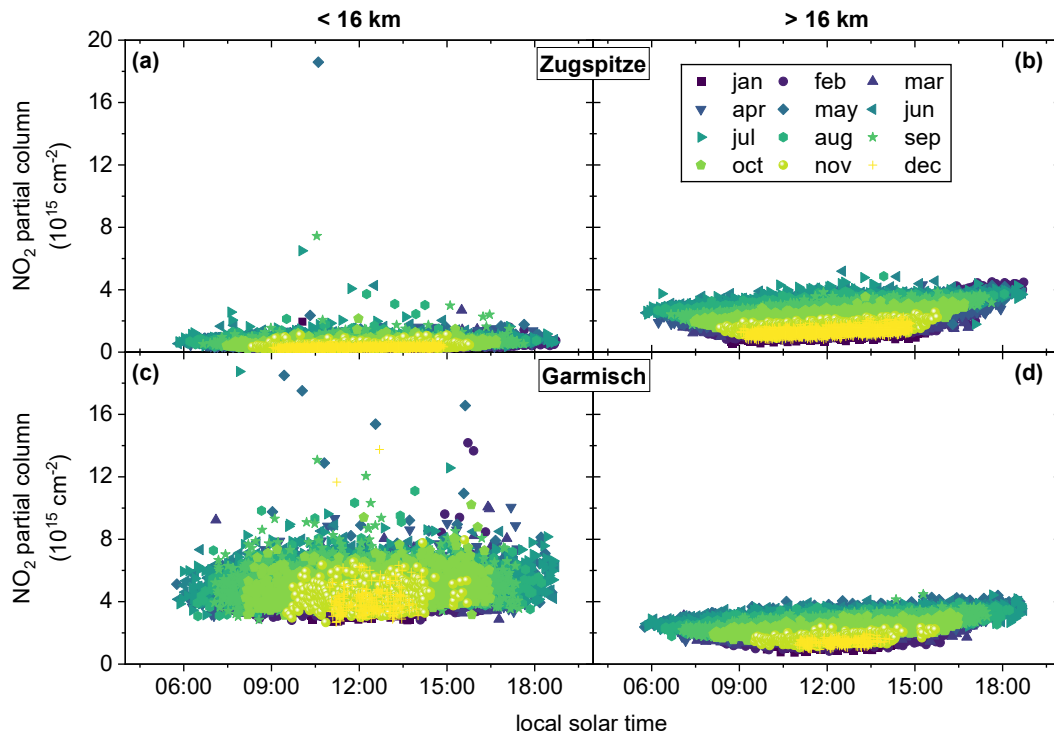



Figure S5. Retrieved NO₂ partial columns for every month below (left) and above (right) 16 km altitude above Zugspitze (top row) and Garmisch (bottom row) in dependency of local solar time (blue to yellow symbols from January to December, see legend).

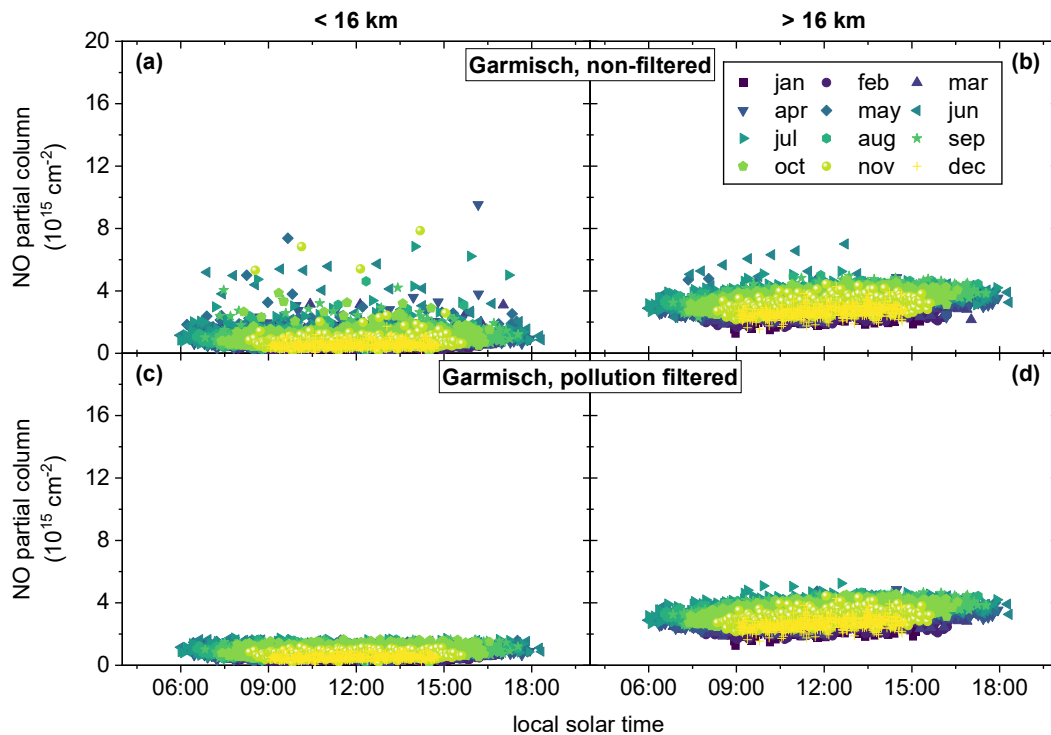



Figure S6. Retrieved Garmisch NO partial columns (top row) and pollution corrected NO partial columns (bottom row, see text for details) for every month below (left) and above (right) 16 km altitude in dependency of the local solar time (blue to yellow symbols from January to December, see legend).

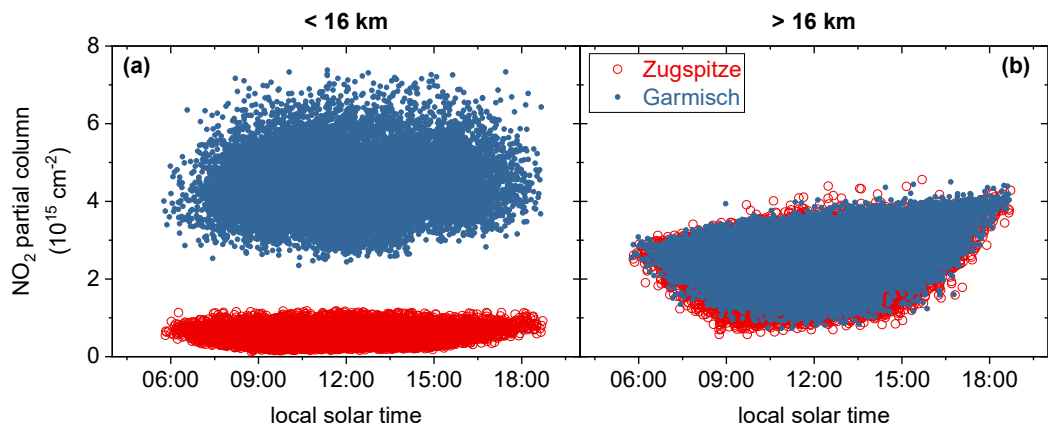



Figure S7. Retrieved pollution filtered NO$_2$ partial columns below (a) and above (b) 16 km altitude measured at Zugspitze (red open symbols) and at Garmisch (blue filled symbols) in dependence of the local solar time.