## S1 Harmonized datasets

The limb harmonized dataset is provided to facilitate the validation and intercomparison efforts within the LOLIPOP project and to provide potential users with a low(er)-threshold comprehensive long-lived greenhouse gases (excluding methane and carbon dioxide) dataset. A harmonized data structure was designed and implemented for these data that built from earlier CCI projects using limb observations.

Each of the satellite datasets are provided in different formats by the instrument teams; they have different file types, time periods per file, vertical grids, variables included, and filtering methods. The harmonized dataset removes these complications so that measurements from each instrument are ready to compare without further modification by the user.

The harmonization methodology was based on that of two ESA CCI projects: ozone_CCI (HARMOZ; Sofieva et al., 2013) and WV_CCI (WAVAS). First, the data from each instrument were filtered following the recommendations of the instrument teams (except HIRDLS, which is provided already filtered). The filtering methods are included in the harmonized files' global attributes.

Once filtered, the vertical grids were harmonized by interpolating each of the profiles onto both a common altitude grid (6-93 km) and a common pressure grid (300-0.0001 hPa). The grid spacings for each dataset are described in Sofieva et al. (2013). These datasets are provided separately as ALT (common altitude) and PRS (common pressure) files, respectively. For the harmonized files on a common altitude grid, the log pressure values for each individual profile were linearly interpolated onto this altitude grid and the interpolated values were reported as pressure in the harmonized file. For the harmonized files on a common pressure grid, the altitude values for each individual profile were linearly interpolated onto this pressure grid. Interpolations onto the pressure grid were done in log pressure space. The altitudes corresponding to each MLS measurement were taken from the Jet Propulsion Laboratory Derived Meteorological Products (Manney et al., 2007). The estimated uncertainties and the corresponding atmospheric temperature for each measurement were also interpolated onto the common grids. For MIPAS-ESA and MLS, the vertical resolution was interpolated as well.

The harmonized files have 12 "mandatory variables" that are common to all five instruments. The CCI Harmonized Ozone (HARMOZ) files were used as a guide for determining these variables. These mandatory variables are outlined in Table S1.

**Table S1.** Mandatory variables in the LOLIPOP harmonized netCDF files. $N_{\text{lev}}$ denotes the number of vertical levels (altitude or pressure) and $N_{\text{prof}}$ denotes the number of profiles.

| Variable [unit] | Dimensions | Description |
|---|---|---|
| time [days since 1900-01-01 00:00:00] | $N_{\text{prof}} \times 1$ | The parameter to index the profiles. |
| air_pressure [hPa] | $N_{\text{prof}} \times 1$ (PRS)<br>$N_{\text{prof}} \times N_{\text{lev}}$ (ALT) | The retrieved air pressure. The vertical coordinates for PRS files and an interpolated value for ALT files. |
| altitude [km] | $N_{\text{prof}} \times N_{\text{lev}}$ (PRS)<br>$N_{\text{prof}} \times 1$ (ALT) | The geometric altitude. An interpolated value for PRS files and the vertical coordinate for ALT files. |
| latitude [deg north] | $N_{\text{prof}} \times 1$ | Latitude of each profile, from $-90°$ to $90°$. |
| longitude [deg east] | $N_{\text{prof}} \times 1$ | Longitude of each profile, from $-180°$ to $180°$. |
| local_solar_time [h] | $N_{\text{prof}} \times 1$ | Local solar time for each profile, calculated for LOLIPOP for ACE-FTS. |
| solar_zenith_angle [deg] | $N_{\text{prof}} \times 1$ | Solar zenith angle for each profile, calculated for LOLIPOP for ACE-FTS. |
| [gas] [ppv] | $N_{\text{prof}} \times N_{\text{lev}}$ | The VMR interpolated onto the common grid. Invalid measurements have a fill value of -999. |
| [gas]_error [ppv] | $N_{\text{prof}} \times N_{\text{lev}}$ | The reported uncertainty of the VMR interpolated onto the common grid. Invalid measurements have a fill value of -999. |
| vertical_resolution [km] | $N_{\text{prof}} \times N_{\text{lev}}$ | The estimated vertical resolution at each measurement location. |
| air_temperature [K] | $N_{\text{prof}} \times N_{\text{lev}}$ | The retrieved air temperature at each measurement location, interpolated onto the common grid. |
| quality_flag | $N_{\text{prof}} \times N_{\text{lev}}$ | LOLIPOP quality flag. Currently all set to 0=good. |

Some instruments have additional variables that could be useful to a harmonized dataset user, so these were also saved. These are mostly related to the orbit number or quality of the retrieval, and all have dimensions of $N_{\text{prof}} \times 1$. The instrument-specific variables are listed in Table S2.

**Table S2.** Additional variables provided for each dataset.

| Instrument | Additional variables |
|---|---|
| ACE-FTS | beta_angle, sunset_sunrise, orbit_number |
| HIRDLS | normalized_chi2, orbit_ascending_flag, orbit_number, profile_id |
| MIPAS (ESA) | geo_id, measurement_mode, dof |
| MIPAS (IMK-IAA) | geo_id, baseline_version, orbit_number, chi2, dof, rms |
| MLS | chunk_number |
| SMR | N/A |

## S2 Additional figures

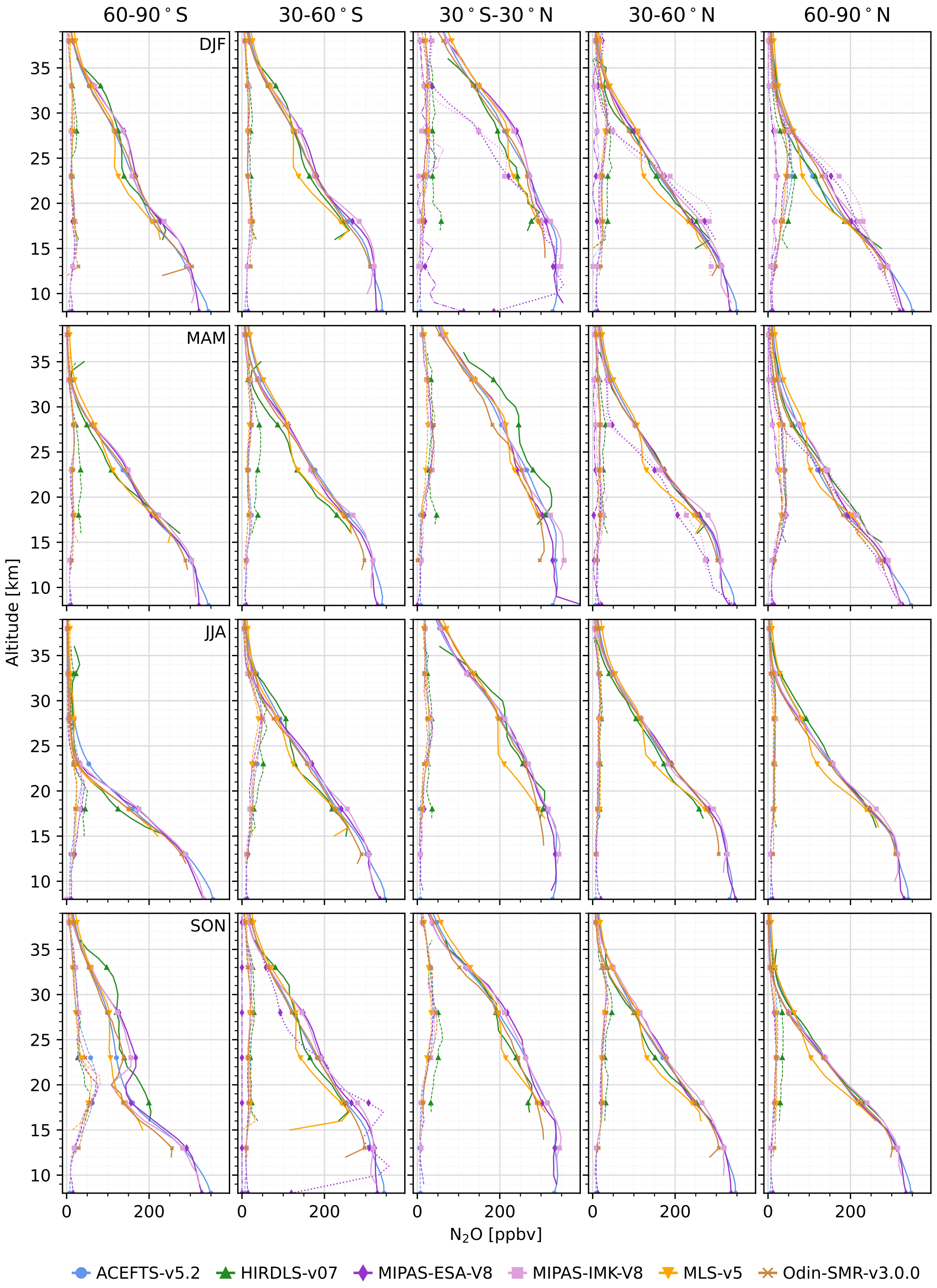


**Figure S1.** Median VMR (solid, except MIPAS full-resolution period, dotted) and median absolute deviation of the VMR (dashed, except MIPAS full-resolution period, dash-dotted) for each instrument, within each latitude band (column) for each season (row). For ACE-FTS, all profiles were included in the median. For other instruments, only profiles coincident with an ACE-FTS profile were included. The MIPAS datasets are split into the full-resolution (dotted/dash-dotted) and reduced-resolution (solid/dashed) periods.

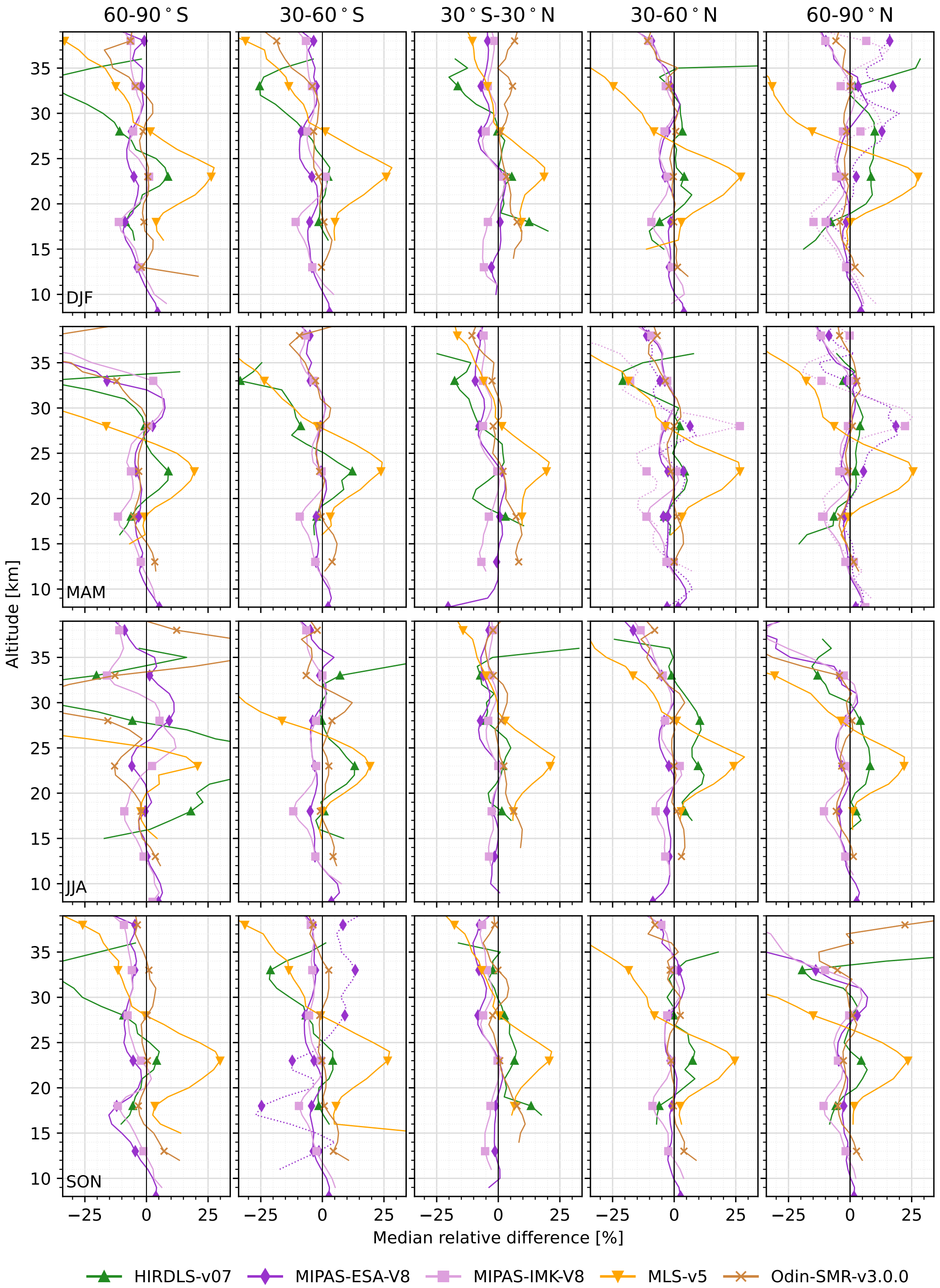


**Figure S2.** Median relative difference between HIRDLS and coincident measurements from each other instrument, within each latitude band (column) for each season (row). The MIPAS datasets are split into the full-resolution (dotted) and reduced-resolution (solid) periods.

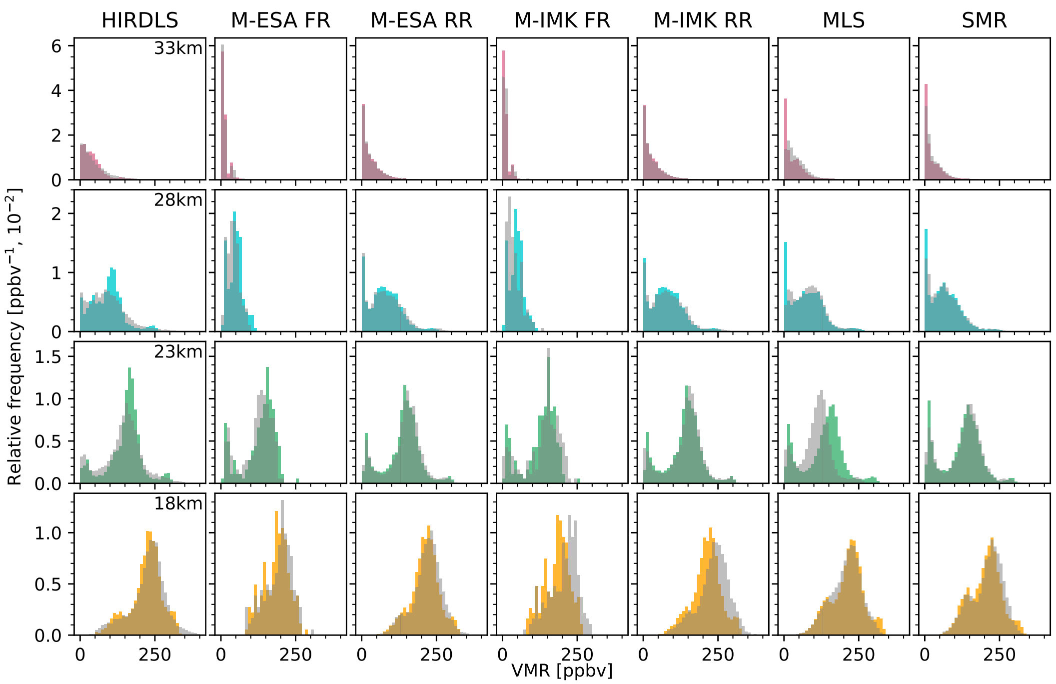


**Figure S3.** PDFs of ACE-FTS (coloured) $N_2O$ measurements that are coincident with each other dataset (grey).

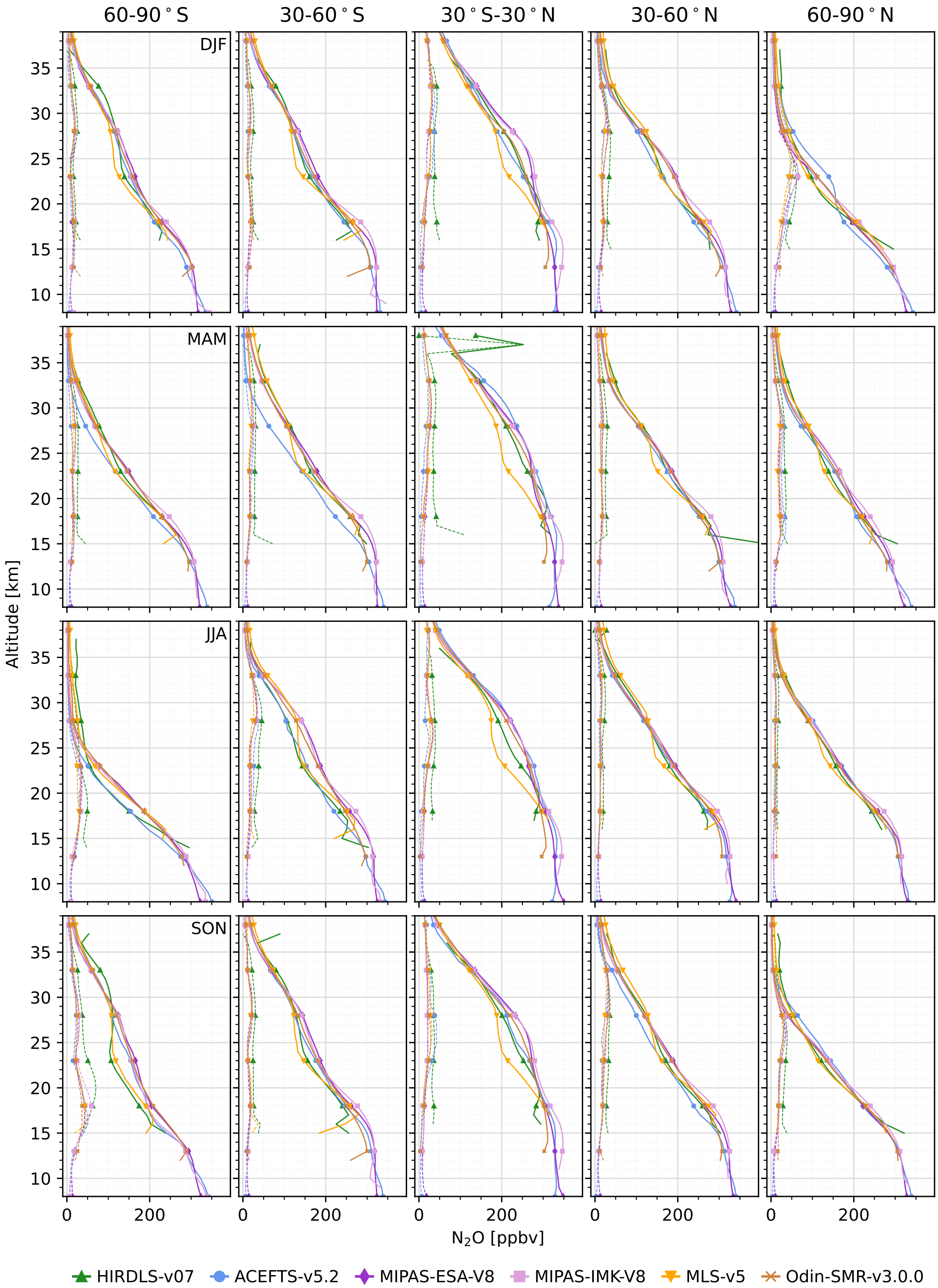


**Figure S4.** Median VMR (solid, except MIPAS full-resolution period, dotted) and median absolute deviation of the VMR (dashed) for each instrument, within each latitude band (column) for each season (row). For HIRDLS, all profiles were included in the median. For other instruments, only profiles coincident with a HIRDLS profile were included.

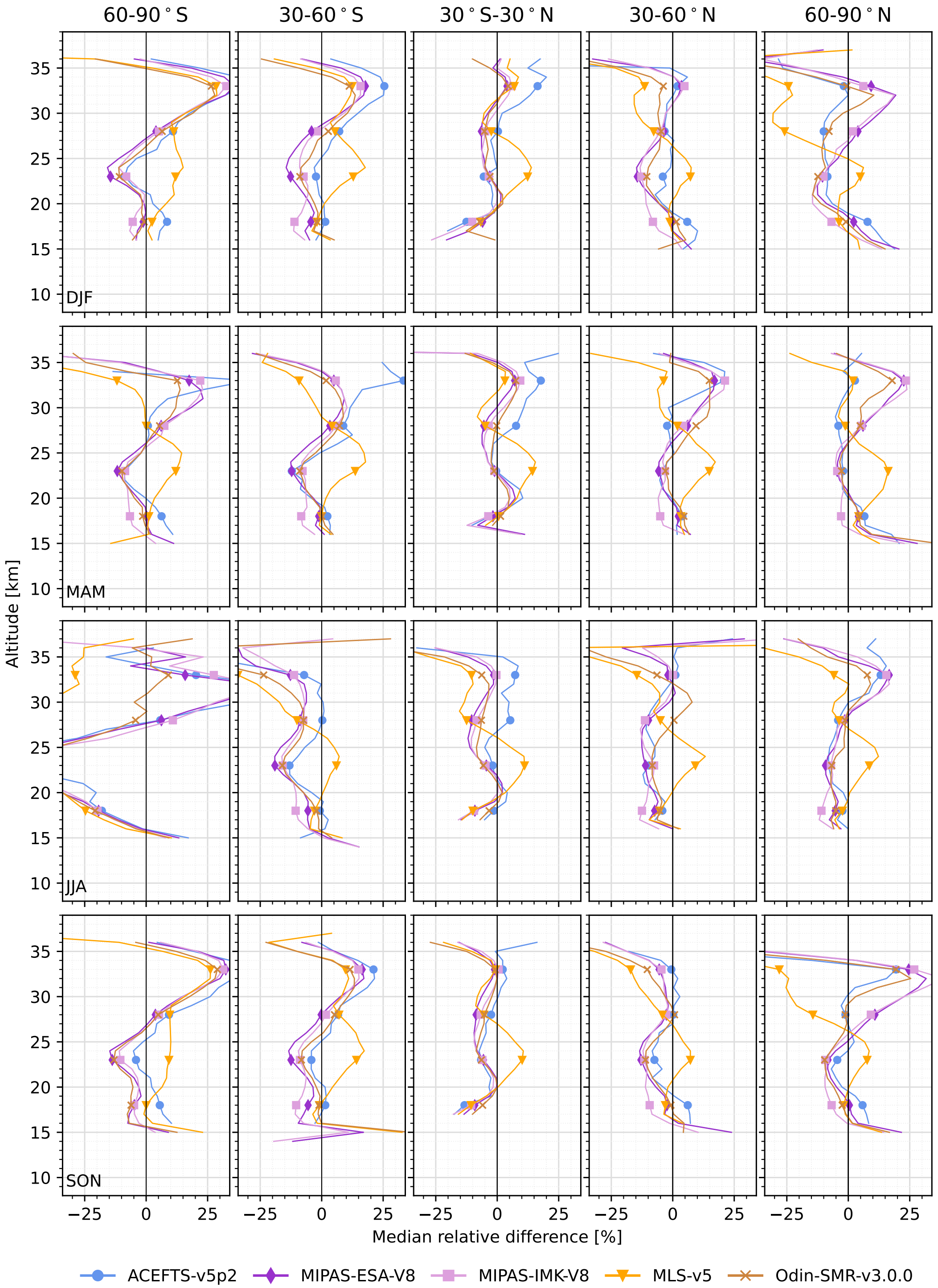


**Figure S5.** Median relative difference between HIRDLS and coincident measurements from each other instrument, within each latitude band (column) for each season (row).

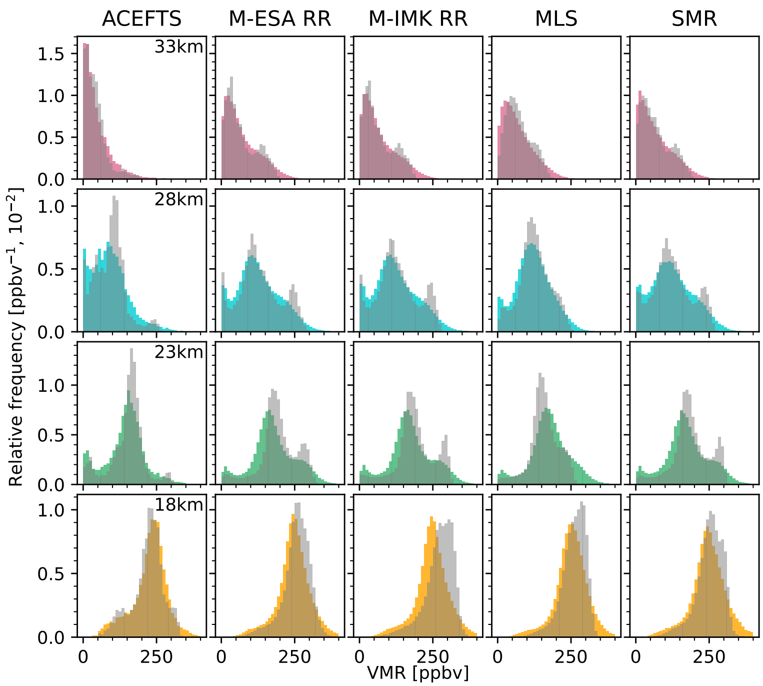


**Figure S6.** PDFs of HIRDLS (coloured) $N_2O$ measurements that are coincident with each other dataset (grey).

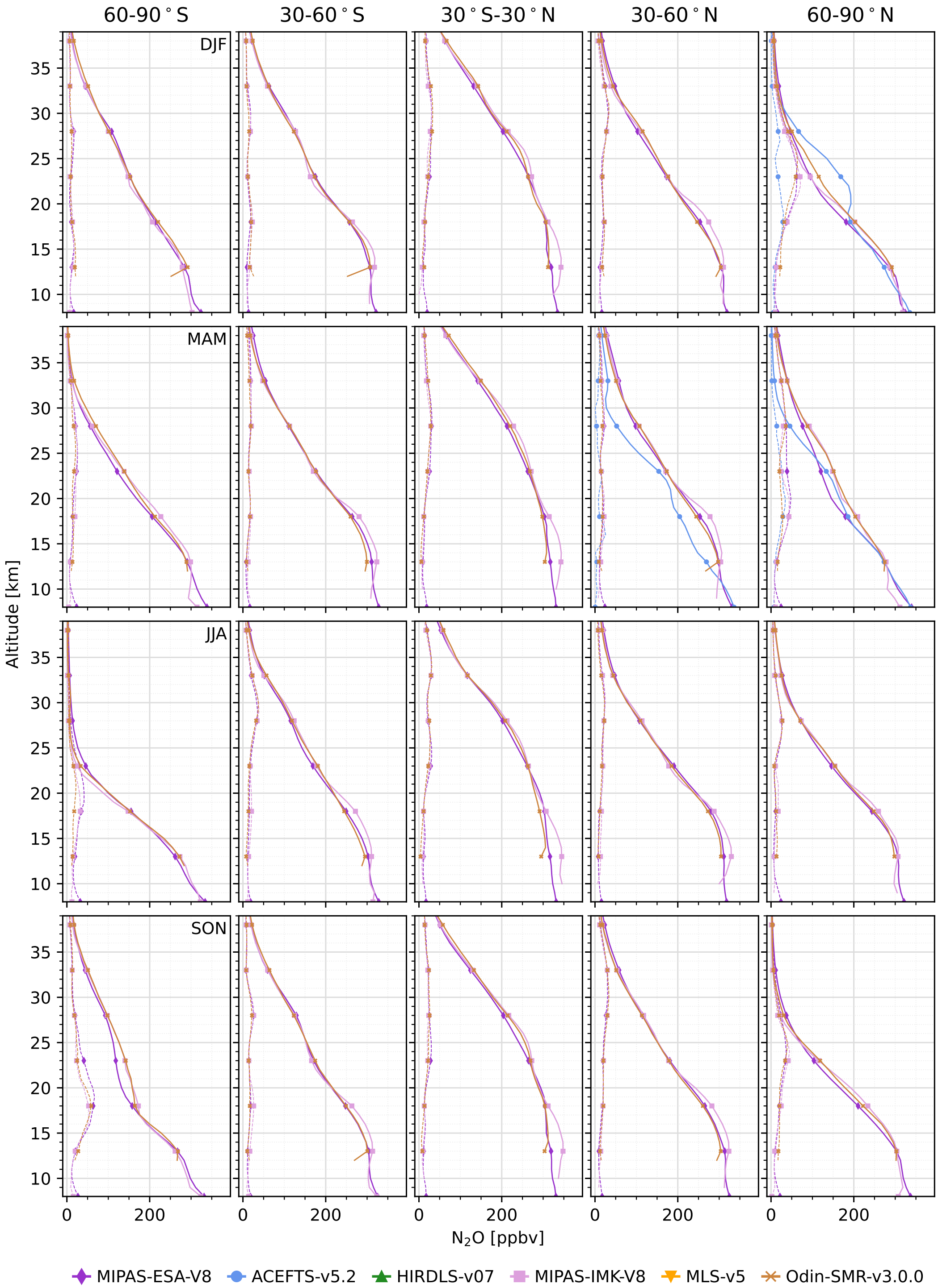


**Figure S7.** Median VMR (solid) and median absolute deviation of the VMR (dashed) for each instrument, within each latitude band (column) for each season (row). For MIPAS-ESA FR, all profiles were included in the median. For other instruments, only profiles coincident with a MIPAS-ESA FR profile were included.

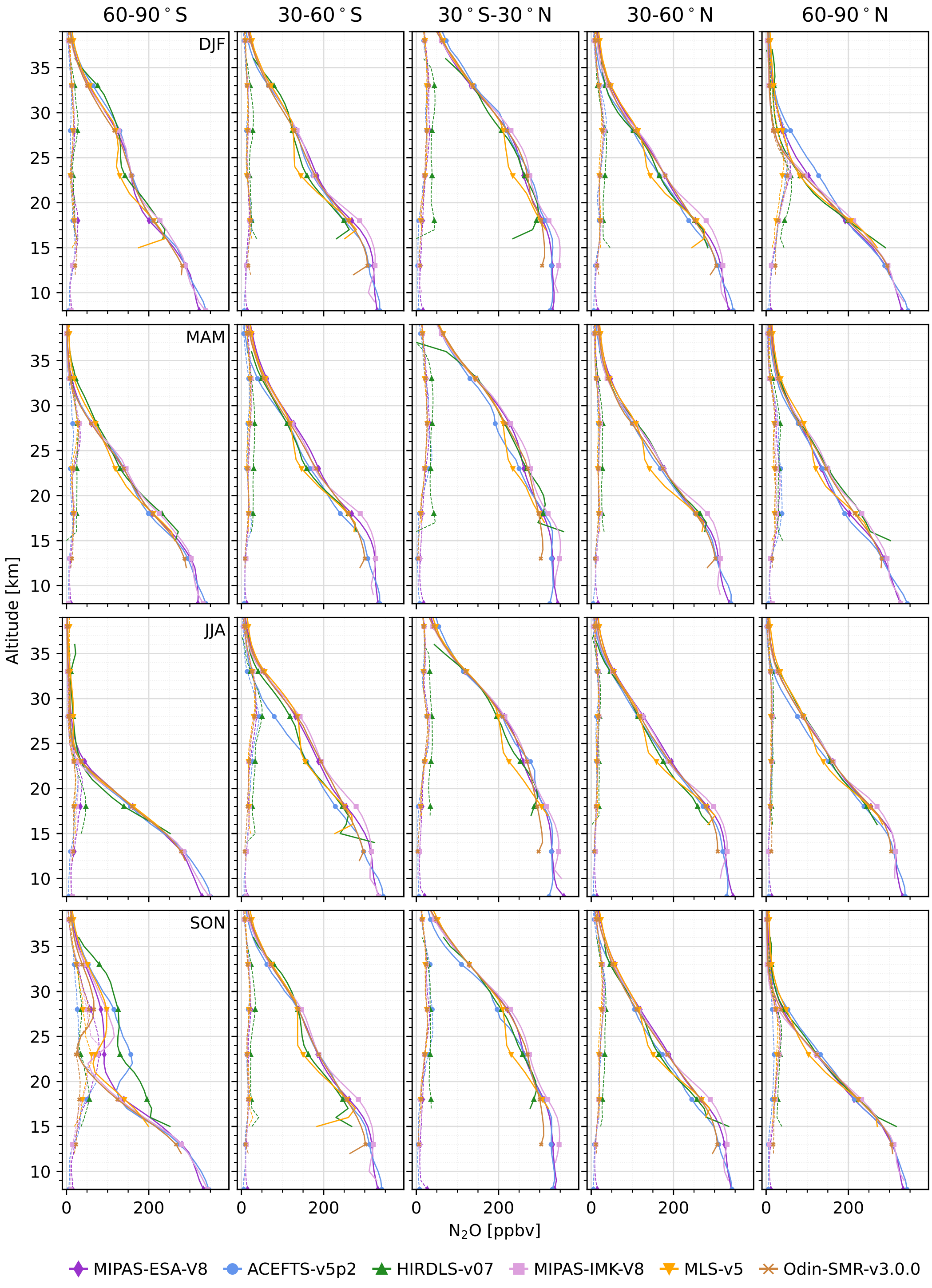


**Figure S8.** Median VMR (solid) and median absolute deviation of the VMR (dashed) for each instrument, within each latitude band (column) for each season (row). For MIPAS-ESA RR, all profiles were included in the median. For other instruments, only profiles coincident with a MIPAS-ESA RR profile were included.

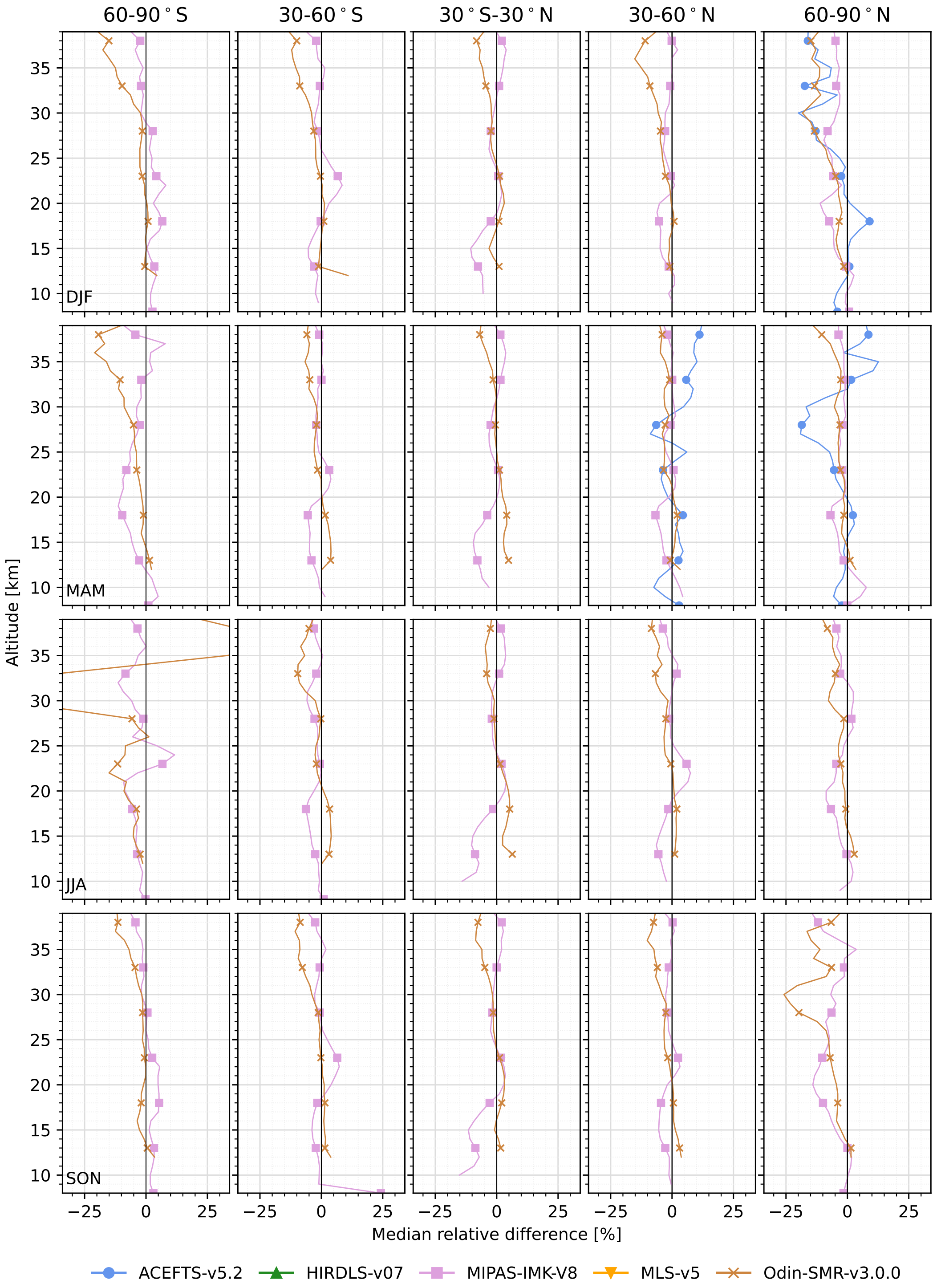


**Figure S9.** Median relative difference between MIPAS-ESA FR and coincident measurements from each other instrument, within each latitude band (column) for each season (row).

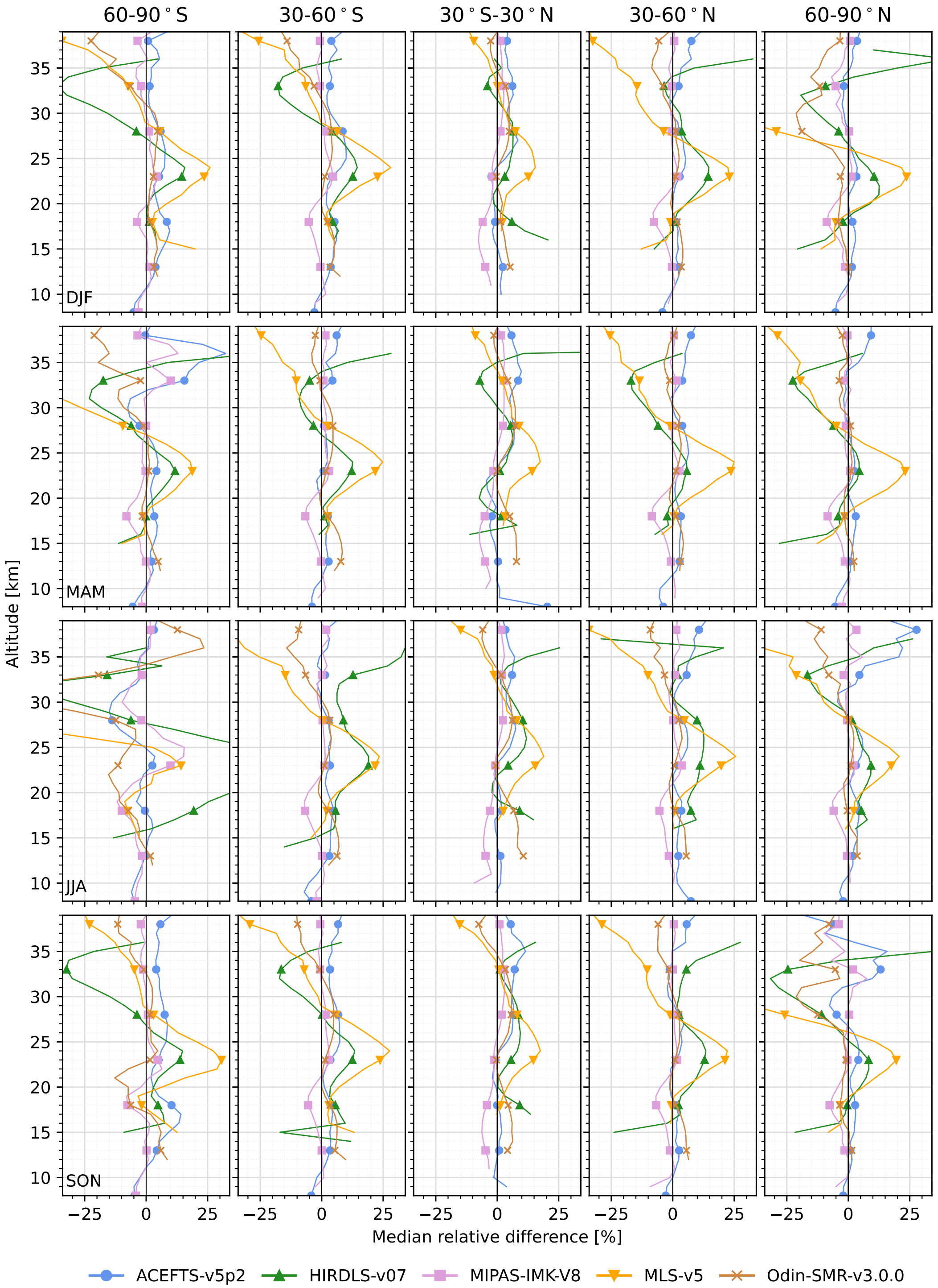


**Figure S10.** Median relative difference between MIPAS-ESA RR and coincident measurements from each other instrument, within each latitude band (column) for each season (row).

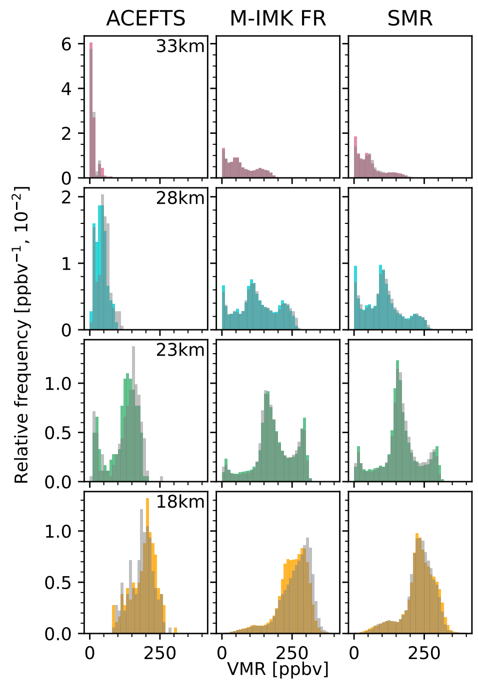


**Figure S11.** PDFs of MIPAS-ESA FR (coloured) $N_2O$ measurements that are coincident with each other dataset (grey).

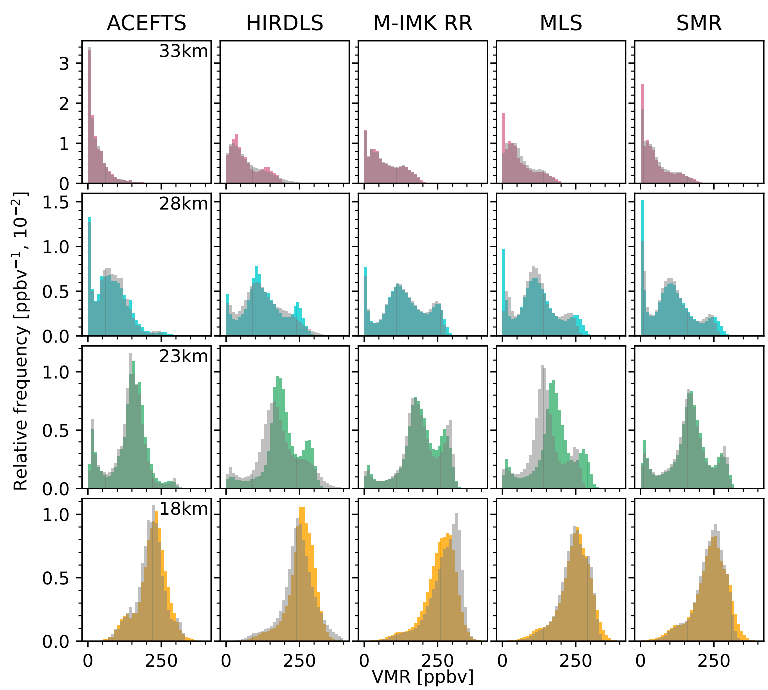


**Figure S12.** PDFs of MIPAS-ESA RR (coloured) $N_2O$ measurements that are coincident with each other dataset (grey).

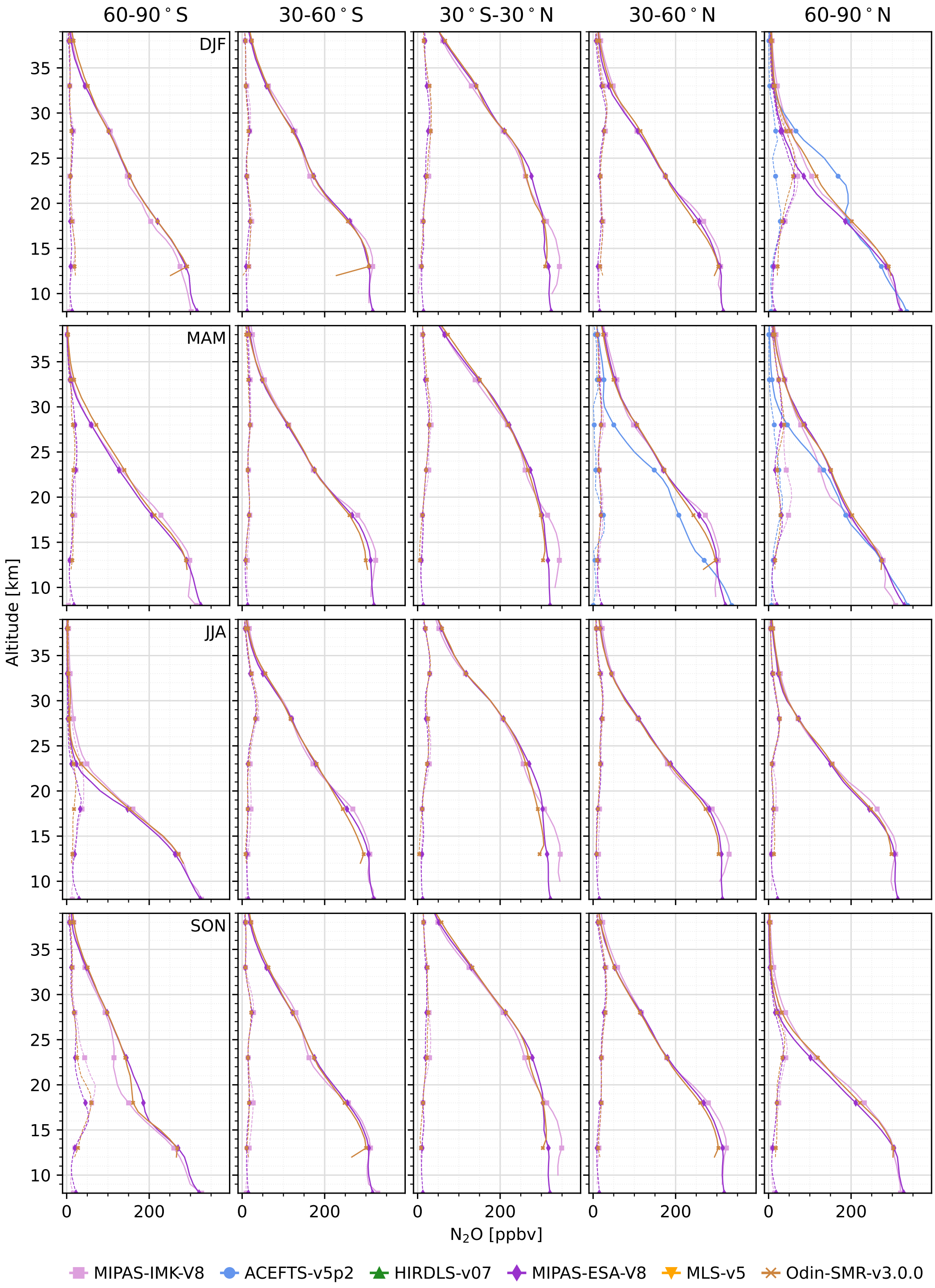


**Figure S13.** Median VMR (solid) and median absolute deviation of the VMR (dashed) for each instrument, within each latitude band (column) for each season (row). For MIPAS-IMK FR, all profiles were included in the median. For other instruments, only profiles coincident with a MIPAS-IMK FR profile were included.

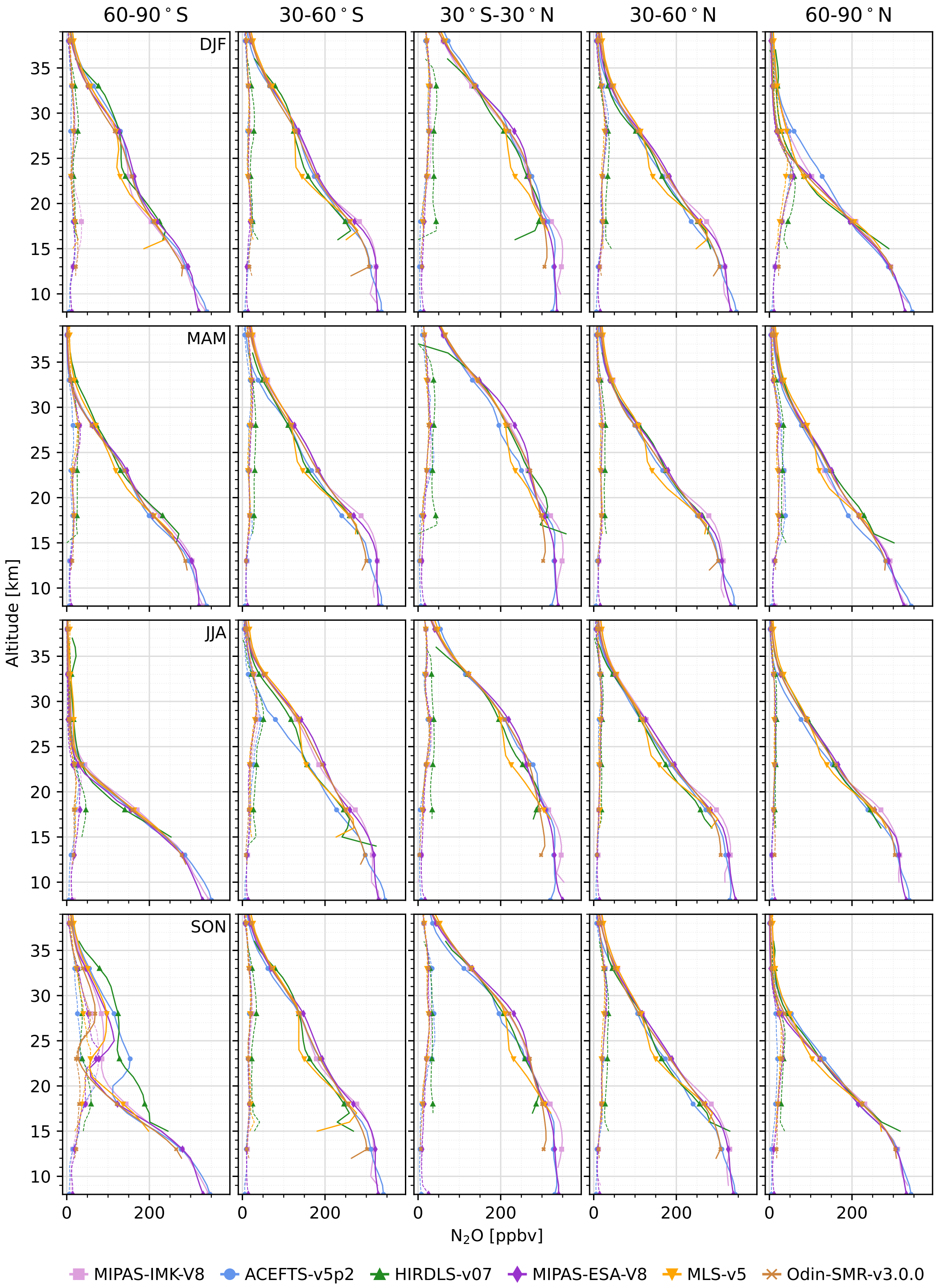


**Figure S14.** Median VMR (solid) and median absolute deviation of the VMR (dashed) for each instrument, within each latitude band (column) for each season (row). For MIPAS-IMK RR, all profiles were included in the median. For other instruments, only profiles coincident with a MIPAS-IMK RR profile were included.

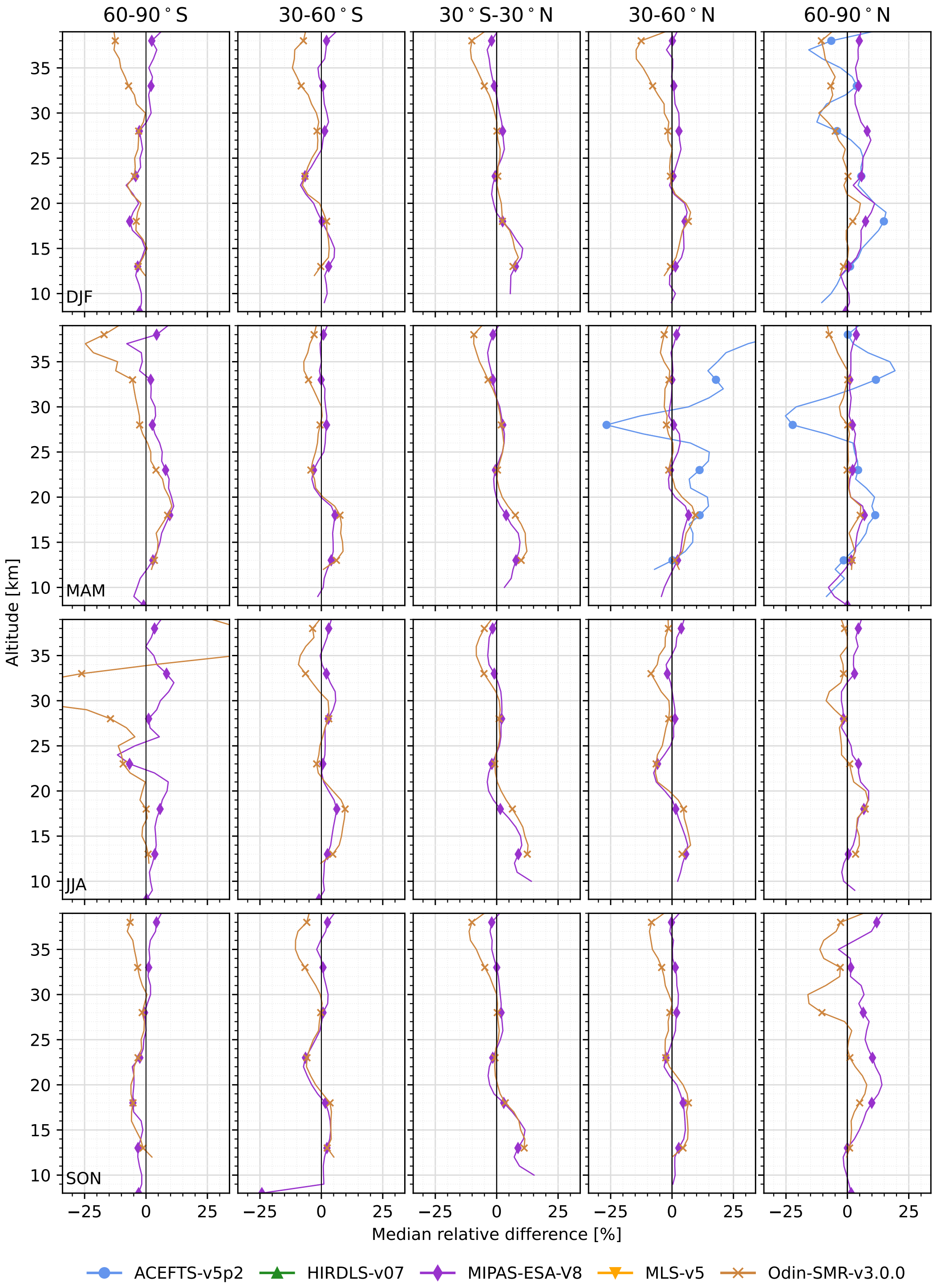


**Figure S15.** Median relative difference between MIPAS-IMK FR and coincident measurements from each other instrument, within each latitude band (column) for each season (row).

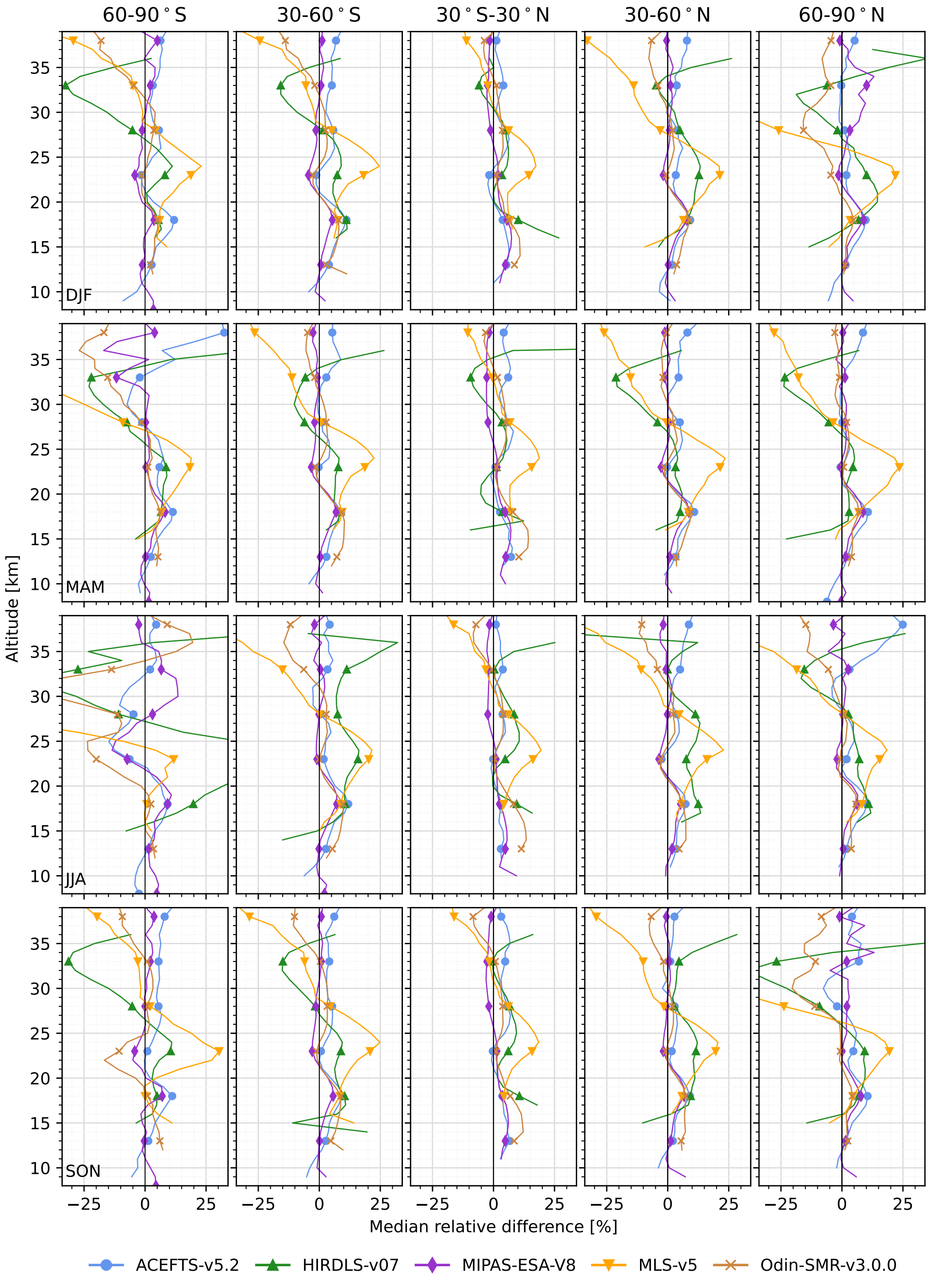


**Figure S16.** Median relative difference between MIPAS-IMK RR and coincident measurements from each other instrument, within each latitude band (column) for each season (row).

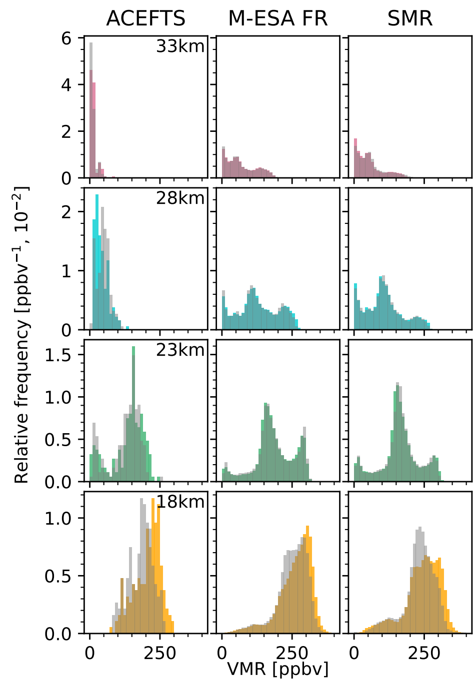


**Figure S17.** PDFs of MIPAS-IMK FR (coloured) $N_2O$ measurements that are coincident with each other dataset (grey).

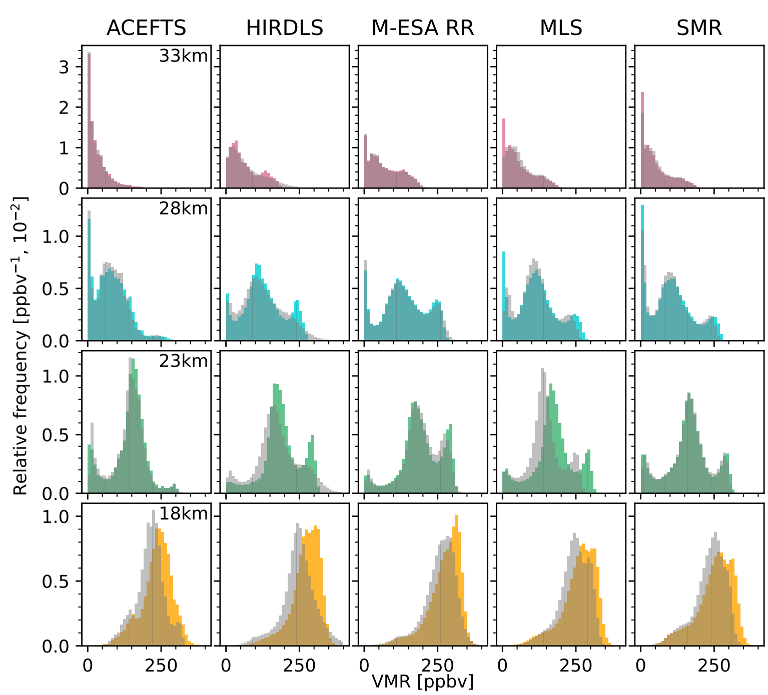


**Figure S18.** PDFs of MIPAS-IMK RR (coloured) $N_2O$ measurements that are coincident with each other dataset (grey).

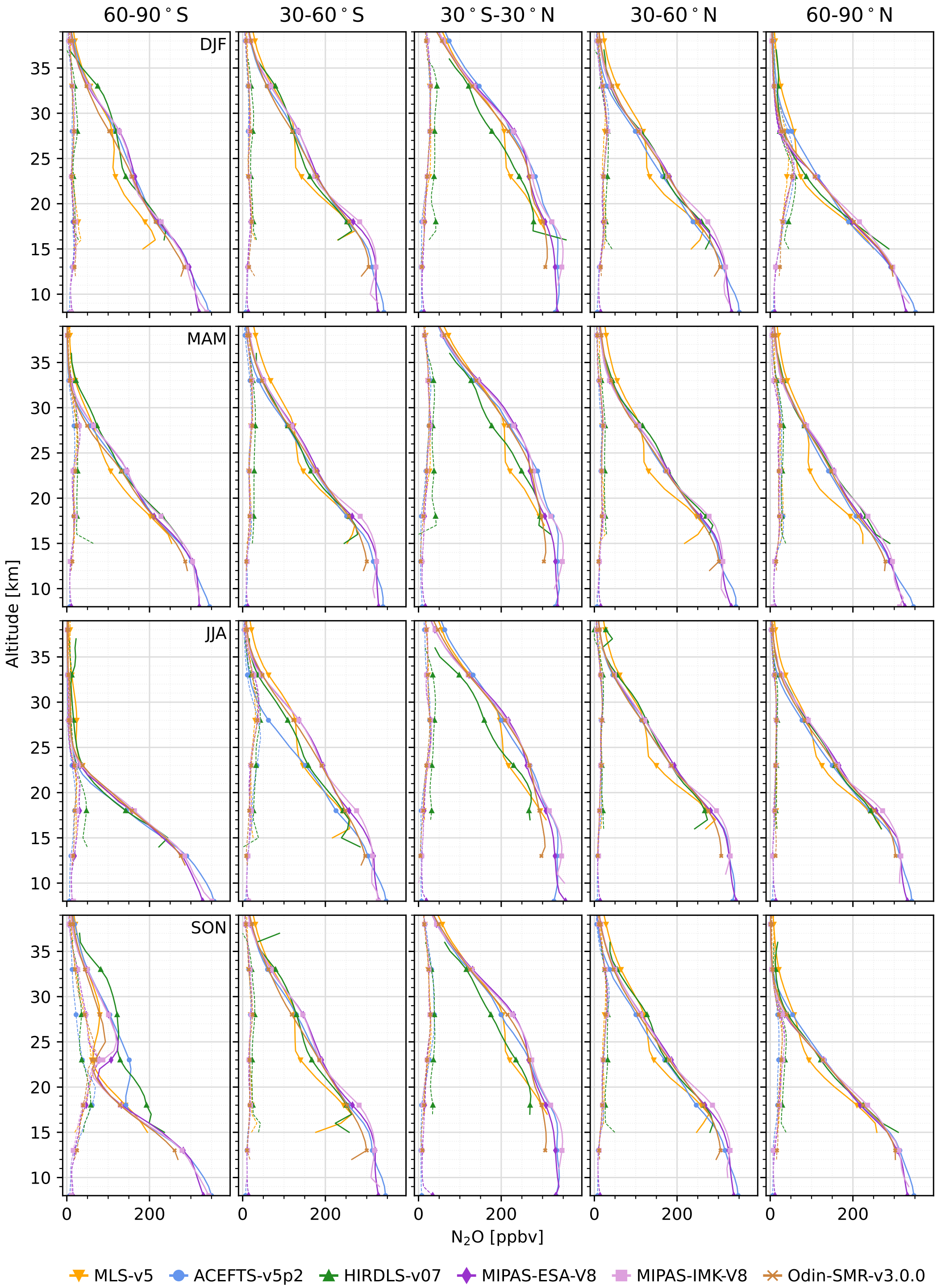


**Figure S19.** Median VMR (solid) and median absolute deviation of the VMR (dashed) for each instrument, within each latitude band (column) for each season (row). For MLS, all profiles were included in the median. For other instruments, only profiles coincident with an MLS profile were included.

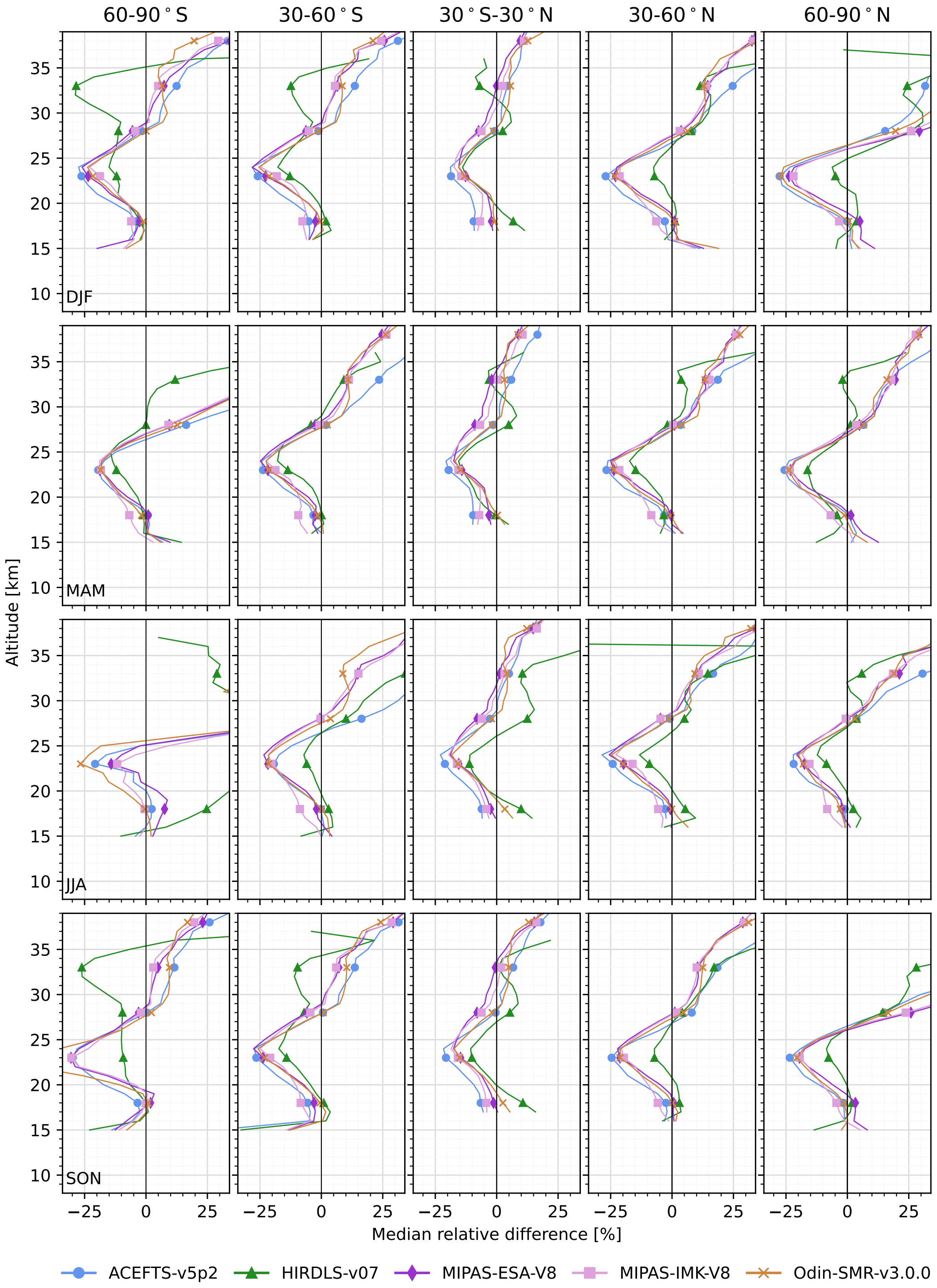


**Figure S20.** Median relative difference between MLS and coincident measurements from each other instrument, within each latitude band (column) for each season (row).

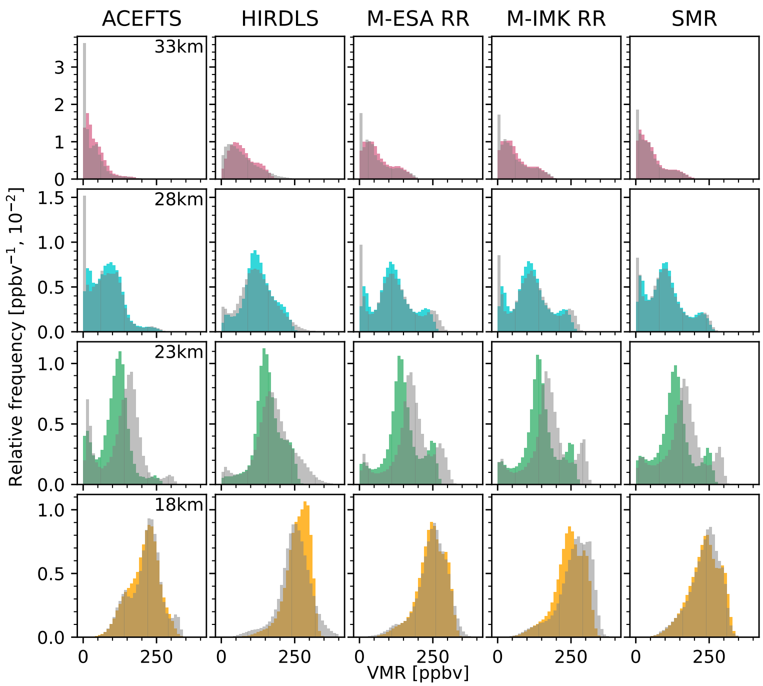


**Figure S21.** PDFs of MLS (coloured) $N_2O$ measurements that are coincident with each other dataset (grey).