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Tropospheric nitrogen dioxide column retrieval based on ground-based zenith-sky DOAS observations

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Nitrogen dioxide (NO_2) is one of the most important chemically active trace gases in the troposphere. Listed as primary pollutant, it is also a key precursor in the formation of tropospheric ozone, aerosols, and acid rain, and can contribute locally to radiative forcing. The long-term monitoring of this species is therefore of great relevance.

Here we present a new method to retrieve tropospheric NO_2 vertical column amounts from ground-based zenith-sky measurements of scattered sunlight. It is based on a four-step approach consisting of (1) the DOAS analysis of zenith radiance spectra using a fixed reference spectrum corresponding to low tropospheric NO_2 content, (2) the determination of the residual amount in the reference spectrum using a Langley-plot-type method, (3) the removal of the stratospheric content from the daytime total slant column using stratospheric vertical columns measured at twilight and simulated stratospheric NO_2 diurnal variation, (4) estimation of the tropospheric vertical columns by dividing the resulting tropospheric slant columns by appropriate air mass factors.

The retrieval algorithm is tested on a 2 month dataset acquired from June to July 2009 by the BIRA MAX-DOAS instrument in the framework of the Cabauw (51.97° N, 4.93° E) Intercomparison campaign for Nitrogen Dioxide measuring Instruments (CINDI). The tropospheric vertical column amounts derived from zenith-sky observations are compared to the vertical columns retrieved from the off-axis and direct-sun measurements of the same MAX-DOAS instrument as well as to data of a co-located SAOZ (Système d'Analyse par Observations Zénithales) spectrometer operated by LATMOS. First results show a good agreement between the different data sets with correlation coefficients and slopes close to or larger than 0.85. We observe that the main error sources arise from the uncertainties in the determination of the residual NO_2 amount in the reference spectrum, the stratospheric NO_2 abundance and its diurnal variation, and the tropospheric and stratospheric NO_2 air mass factors.

Although zenith-sky measurements have been commonly used over the last decades for the monitoring of the stratosphere, this study demonstrates that these observations are also suitable for the retrieval of tropospheric NO_2 column amounts, offering therefore new perspectives for the exploitation of zenith-sky UV-Vis measurements over the NDACC (Network for the Detection of Atmospheric Composition Change) stations.