Tropospheric NO_2 and HCHO derived from dual-scan MAX-DOAS measurements in Uccle (Belgium) and application to S5P/TROPOMI validation

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Motivation

Study of tropospheric nitrogen dioxide (NO_2):

- *Mainly emitted by anthropogenic activities*
- *Participation in tropospheric ozone formation*

and formaldehyde (HCHO)

- *Intermediate product in the oxidation of most volatile organic compounds (VOCs)*
- *Tracer of VOCs*

Focusing on Brussels area:

- *NO_2 concentrations among the highest in Europe as observed by in-situ stations and satellite instruments*
- *HCHO concentrations have never been presented for such a big time period (March 2018 –December 2020)*

Instrumentation

A. Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) instrument

→ Measures continuously in both UV and Vis wavelength ranges in dual-scan configuration
→ Dual-scan configuration (Fig. 1): One vertical scanning towards the blue azimuthal direction (the so-called main azimuthal direction in 9 elevation angles) and 9 azimuthal measurements at one elevation angle (2 degrees)
→ Capable of determining the vertical and horizontal distribution of trace gases

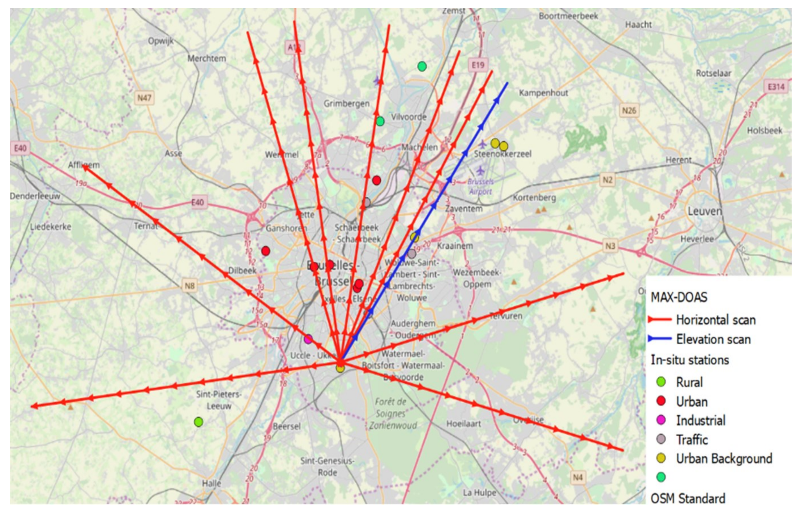

Fig.2: The MAX-DOAS instrument in Brussels.

Fig.1: The dual-scan experimental set-up of the BIRA-IASB MAX-DOAS instrument. The colored dots show the locations of the in-situ stations in Brussels.

B. TROPOMI instrument

- UV-Vis-NIR-SWIR spectrometer
- Atmospheric composition measurements with high spatio-temporal resolution ***(ground pixel of 3.5 x 7 km² and 3.5 x 5.5 km² since 6 August 2019)*** *related to air quality, climate forcing, ozone and UV radiation*
- ***Daily global coverage***
- ***Data continuity*** *between Envisat Satellite and NASA's Aura mission and the launch of Sentinel-5 (period between 2017 and 2023)*

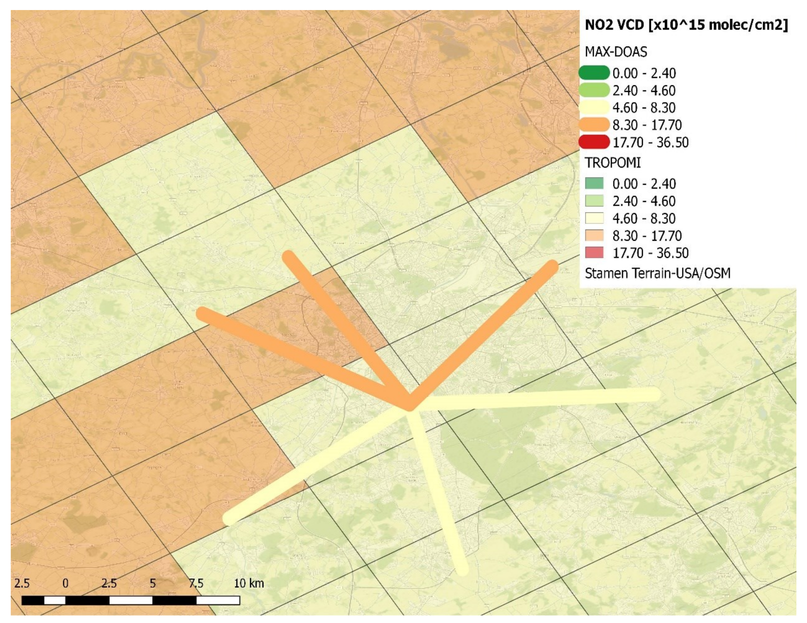

Fig.3: Tropospheric NO_2 columns derived from the TROPOMI and the MAX-DOAS instrument on 06 June 2018 near the measurement site in Uccle (overlaid onto OSM Standard layer).

References

- Friedrich, M. M., Rivera, C., Stremme, W., Ojeda, Z., Arellano, J., Bezanilla, A., García-Reynoso, J. A., and Grutter, M.: NO_2 vertical profiles and column densities from MAX-DOAS measurements in Mexico City, Atmos. Meas. Tech., 12, 2545–2565, https://doi.org/10.5194/amt-12-2545-2019, 2019.
- Sinreich, R., et al. "Parameterizing radiative transfer to convert MAX-DOAS dSCDs into near-surface box-averaged mixing ratios." *Atmospheric Measurement Techniques* 6.6 (2013): 1521-1532.
- Kreher, K., Van Roozendael, M., Hendrick, F., Apituley, A., Dimitropoulou, E., Frieß, U., ... & Anguas, M. (2019). Intercomparison of NO2, O4, O3 and HCHO slant column measurements by MAX-DOAS and zenith-sky UV-Visible spectrometers during the CINDI-2 campaign.

MAX-DOAS: Measurements – Retrieval Strategy

1. DOAS Analysis

- *Measured spectra to differential slant column density (DSCD)*
  → QDOAS spectral fitting software
- *Fitting settings in the UV and in the VIS*
  → Same as during CINDI-2 Inter comparison campaign (Kreher et al., 2020)
- *Measurement dataset → March 2018 – December 2019*

2. OEM-based profile retrieval

- *Main azimuthal direction – Vertical scanning*
- *Application of the MMF inversion algorithm (Friedrich et al., 2019)*
- *Use of quality-checked profile retrievals*
- *Cloud filter based on the measurements of a co-located pyrometer*

3. Dual-scan retrieval strategy

- *OEM-based profile retrieval cannot be applied in the other azimuthal directions (Fig. 1 → red lines)*
- *A parameterization technique proposed by Sinreich et al. (2013) is applied to the dual-scan MAX-DOAS measurements*
- *Retrieval of NO_2 and HCHO near-surface volume mixing ratios (VMR) and vertical column densities (VCD) in all the azimuthal directions (Dimitropoulou et al., AMTD, 2020)*
- *An important variable is the horizontal sensitivity for the NO_2 → until which distance from the instrument the measurements are representative for the NO_2 field*

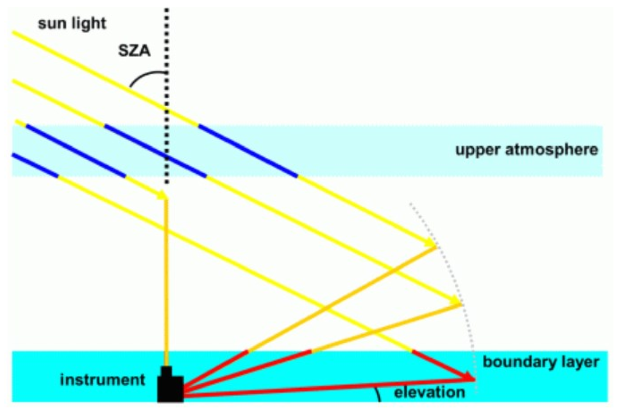

Fig.4: MAX-DOAS technique for tropospheric measurements (© University of Bremen IUP DOAS).

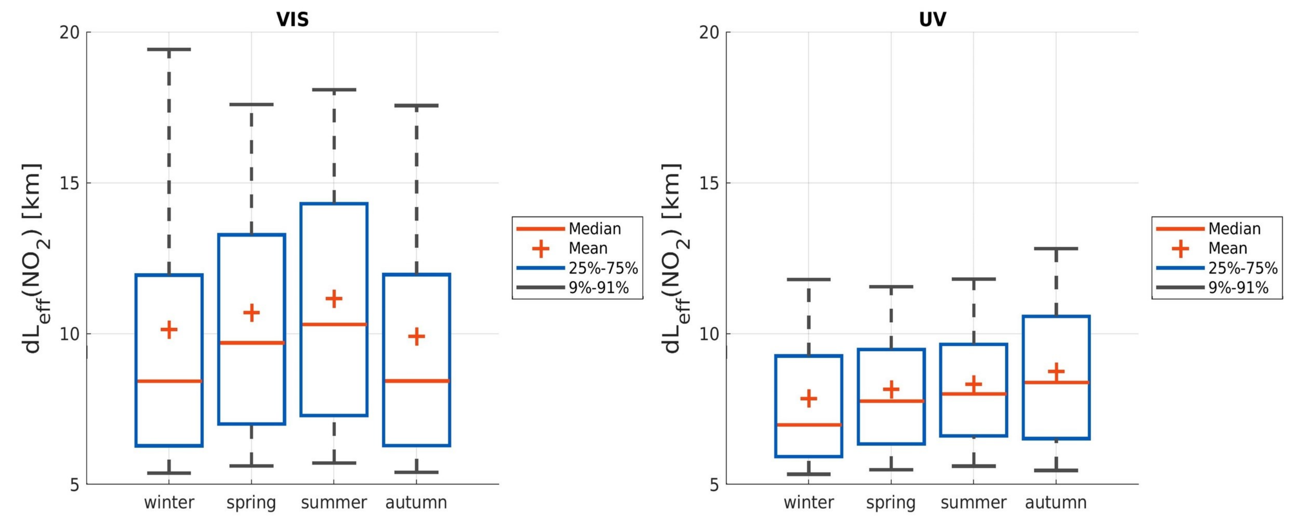

Fig.5: Box and whisker plots representing the seasonal horizontal sensitivity as derived from all the azimuthal viewing directions for the Vis and UV spectral ranges.

Seasonal variation of NO_2 and HCHO

- NO_2 and HCHO near-surface VMRs and VCDs as retrieved in the main azimuthal direction
- Clear seasonal cycle for both trace gases
- Maximum concentrations: NO_2 during cold months and HCHO during warm months, as expected

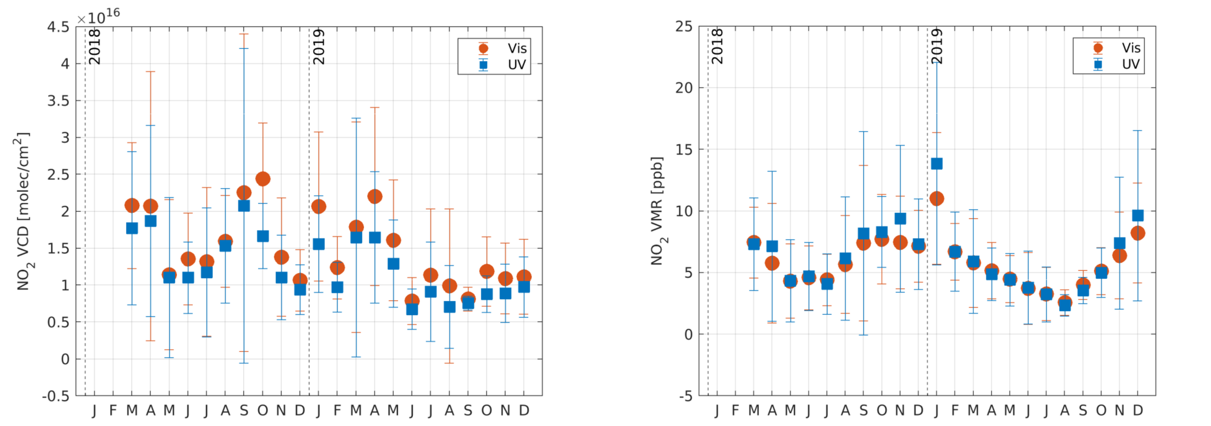

Fig.6: Monthly NO_2 (left panel) VCD and (right panel) VMR means covering two years of MAX-DOAS measurements.

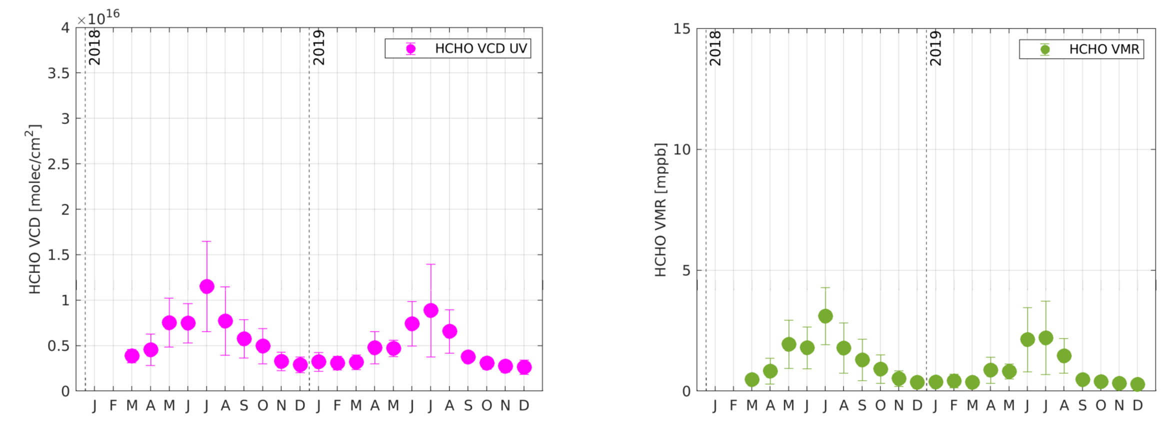

Fig.7: Monthly HCHO (left panel) VCD and (right panel) VMR means covering two years of MAX-DOAS measurements.

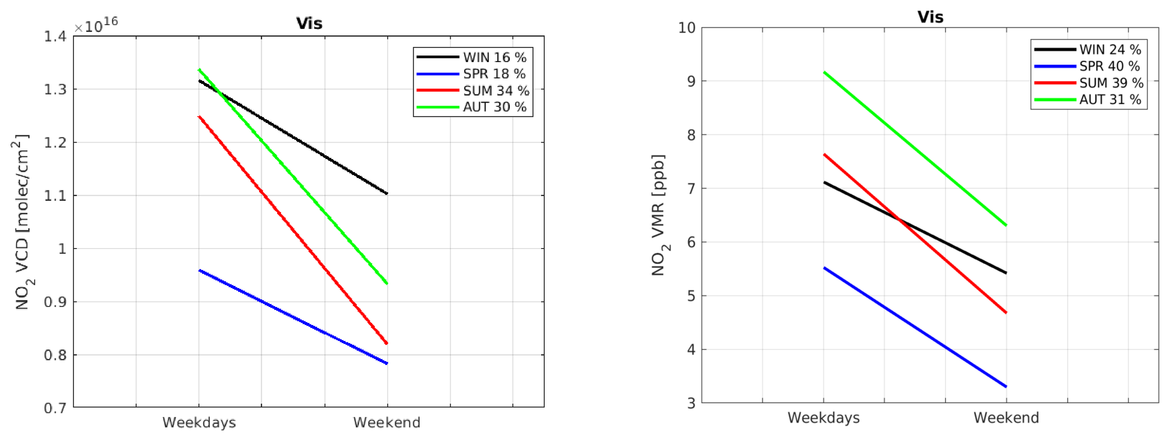

Fig.8: Seasonal NO_2 (left panel) VCD and (right panel) VMR mean values during weekdays and weekends

→ Clear traffic contribution during all seasons (28 – 40 %)

Dual- scan seasonal variation of NO_2 and HCHO

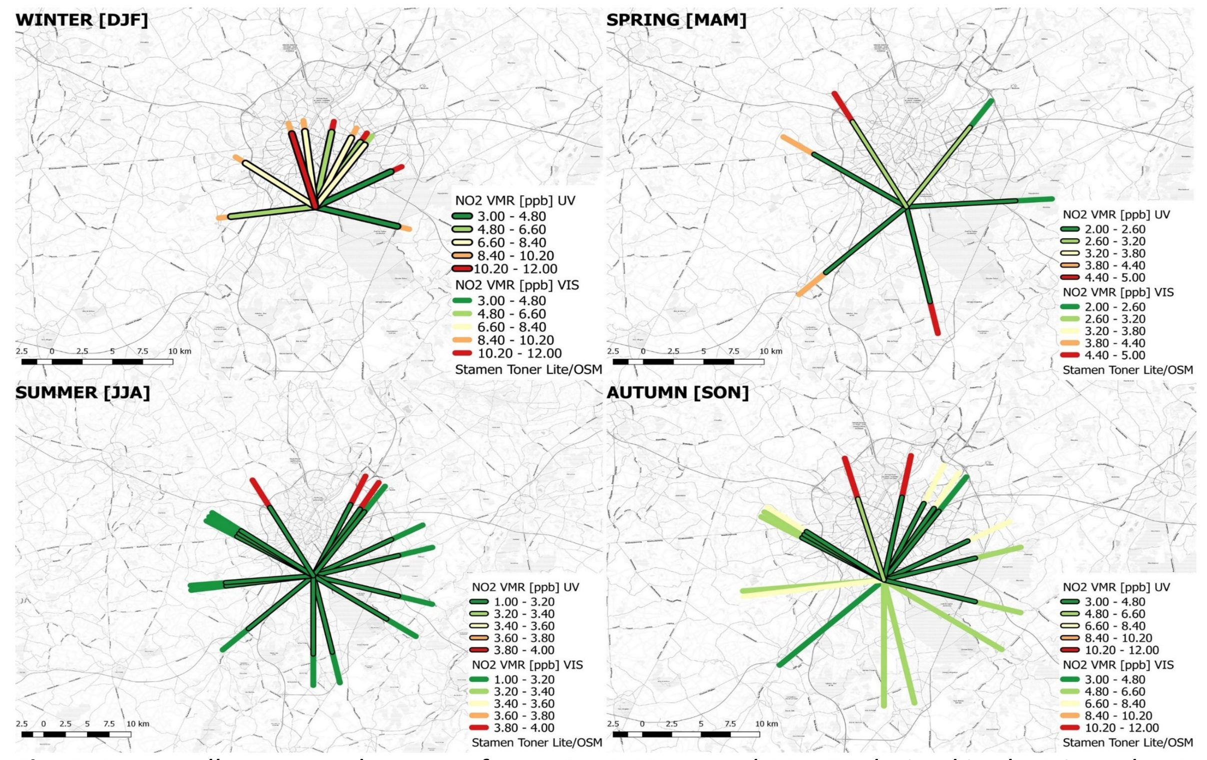

Fig. 9: Seasonally-averaged near-surface NO_2 VMR around 11 UTC derived in the Vis and UV wavelength ranges. Lines with black borders represent the UV VMRs, and lines without black borders show the Vis VMRs. The length of each line represents the seasonally-averaged horizontal sensitivity. Different color scales are used per season.

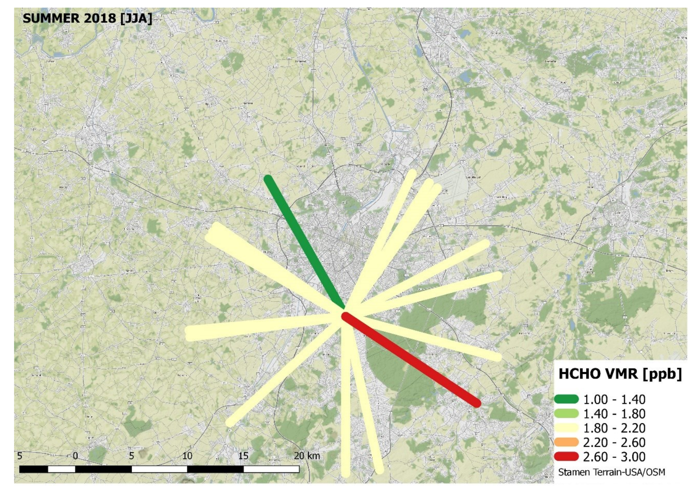

Fig. 10: Near-surface HCHO VMR around 11 UTC as derived during summer 2019.

→Higher NO_2 emissions in the North, where the city center, airport, and congested motorway are located.
→Higher HCHO emissions in the South East: largest forested area in Brussels.

NO_2 and HCHO TROPOMI validation

- *Dual-scan MAX-DOAS tropospheric* NO_2 and HCHO measurements in every MAX-DOAS azimuthal direction are compared with a weighted average of TROPOMI columns as measured in coincident pixels with the weighting being determined by the MAX-DOAS horizontal sensitivity segment crossing every pixel.

→ The correlation coefficient is good but the TROPOMI values are systematically lower than the MAX-DOAS measurements (slope values around 0.4-0.7 and 0.7 for NO_2 and HCHO, respectively).

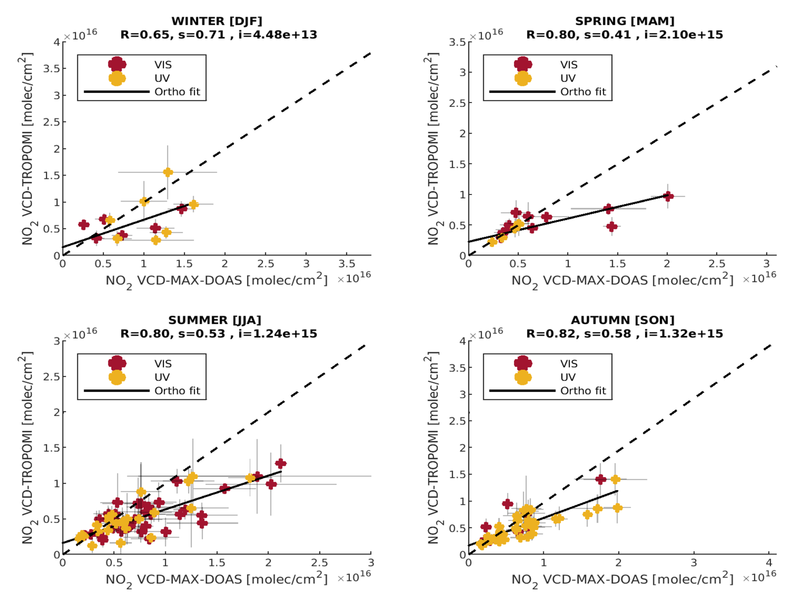

Fig. 11: Seasonal scatter plots between the tropospheric NO_2 columns derived from the dual-scan MAX-DOAS observations and the TROPOMI collocated pixels.

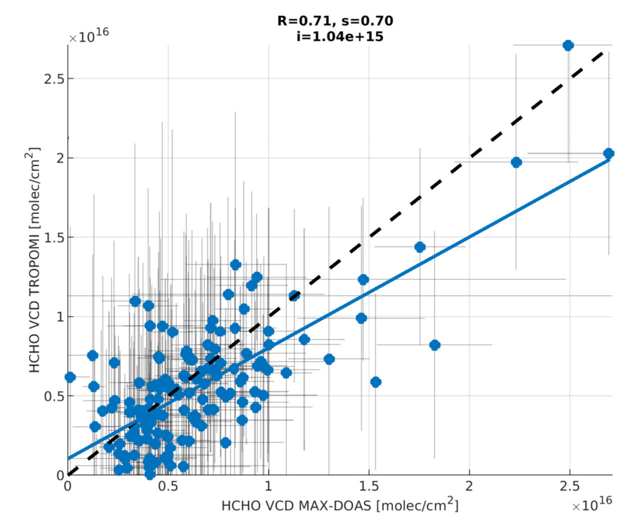

Fig. 12: Scatter plot between the tropospheric HCHO columns derived from the dual-scan MAX-DOAS observations and the TROPOMI collocated pixels.

Impact of systematic uncertainties in the satellite retrieval → A-priori profile shape

→ Recalculation of the TROPOMI/S5P VCDs using vertical profiles from MAX-DOAS measurements
→ Change of the a-priori profile significantly improves the agreement between TROPOMI and MAX-DOAS data sets for both NO_2 and HCHO.

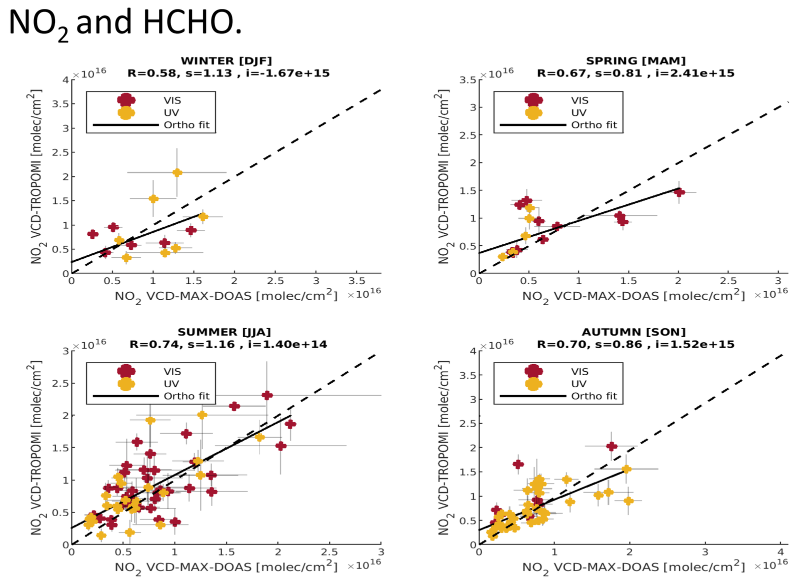

Fig. 13: Seasonal scatter plots between the tropospheric NO_2 columns derived from the dual-scan MAX-DOAS observations and the re-calculated TROPOMI tropospheric columns.

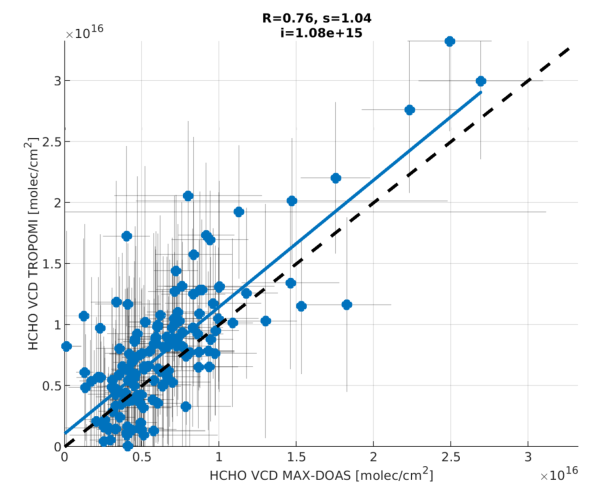

Fig. 14: Scatter plots between the tropospheric HCHO columns derived from the dual-scan MAX-DOAS observations and the re-calculated TROPOMI columns.

Take-home message

→ Two years of dual-scan MAX-DOAS NO_2 and HCHO near-surface VMRs and VCDs in Uccle, Brussels are presented here
→ The dual-scan MAX-DOAS measurements conducted in an urban area, like Brussels can:
  → better characterize the spatial variability of important pollutants, such as NO_2 and HCHO
  → improve our knowledge about the seasonality and the hotspots of NO_2 and HCHO in Brussels
  → improve validation results of satellite air quality measurements with high spatial resolution, such as TROPOMI/S5P