Cross-evaluation of tropospheric NO_2 levels over Flanders using ground-based, airborne and TROPOMI observations and the WRF-Chem model as an intercomparison platform

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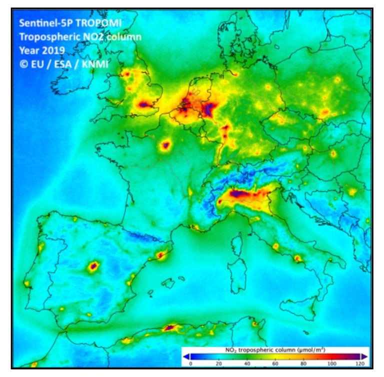
Introduction — NO_2 over Antwerp

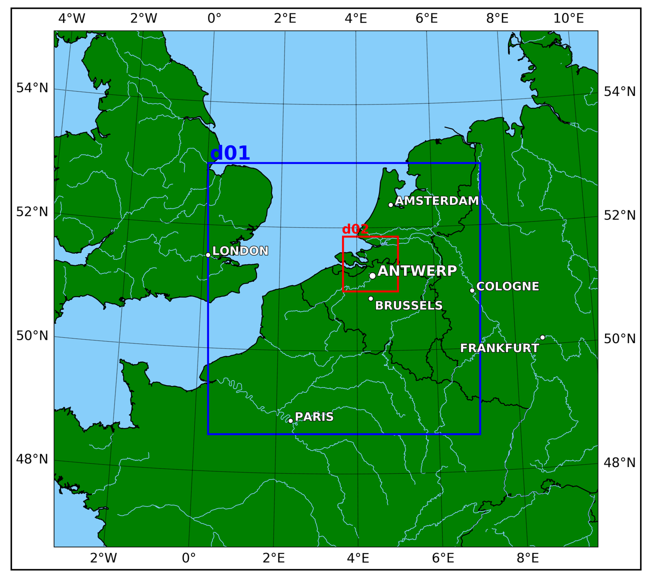
Antwerp is the most populous city in the Belgian region of Flanders, and hosts both the second largest petrochemical cluster in the world and the second biggest port in Europe. Satellite maps revealed the city of Antwerp as one of the strongest NO_2 hotspots worldwide. According to the 2017 Annual Air Report for Flanders[1], 13 out of 19 measuring sites in Antwerp showed NOx concentrations exceeding the European annual limit value.



We investigate the NO_2 distribution in Flanders, as recorded by ground-based concentration measurements, airborne data and remotely-sensed columns, with the help of the high-resolution WRF-Chem chemical-transport model as cross-comparison tool. This study also aims to build on previous TROPOMI validation campaigns over the region (**Tack** et al, 2017;2021)

Model Set Up — WRF-Chem

WRF-Chem[2,3] is the Weather Research and Forecasting (WRF) model coupled with Chemistry. The model was set up using two domains: 5km (**d01**) and 1km (**d02**) resolution.



The simulation period was set up as

SHORT: 66-hour runs from 27 June 2019 00:00 until 29 June 2019 18:00 and

LONG: 15-day runs from 15 June 2019 00:00 until 30 June 2019 00:00

The chemical mechanism used was Carbon-Bond Mechanism by Zaveri (**CBMZ**)[4]. The emissions cover the following species: CO, NOx (as NO), NH3, SO2, PM2.5, PM10, organic carbon, black carbon and disaggregated volatile organic compounds (VOCs), and the input files come from a combination of:

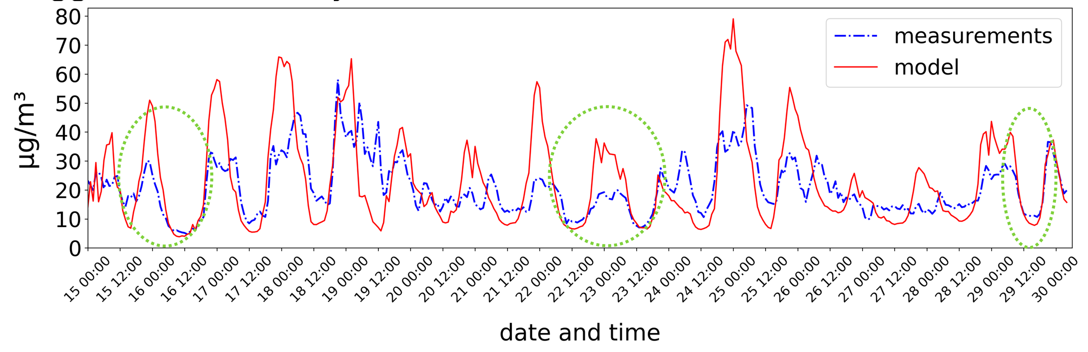
- 1km x 1km emissions from **VITO**[5] (over Flanders)
- 1km x 1km emissions from **Emissieregistratie NL**[6] (over Netherlands)
- 0.1° x 0.1° emissions from **EMEP**[7]
- 0.1° x 0.1° emissions from **EDGAR-HTAP**[8]
- 0.1° x 0.1° aircraft emissions from **CAMS**

The emissions were adjusted to account for diurnal, daily (depending on the day of the week) and monthly behaviour for the simulation period, in accordance with Crippa et al. (2020)[9].

Injection heights were incorporated over Flanders for industrial NO_2 in accordance with vertical distribution information for the emissions provided by VITO. The emissions are distributed between the surface and around 300m.

Comparison with ground-based measurements

We compare the 15-day simulation results with NO2 concentration measurements at 30 stations hosted by the Belgian Interregional Environmental Agency (IRCEL-CELINE)[10], all located around Antwerp. The modelled and observed NO2 concentrations averaged over the stations are plotted below. The measurements are tentatively corrected for interferences from PAN and HNO_3 following the suggestions made by Lamsal et al. 2008.



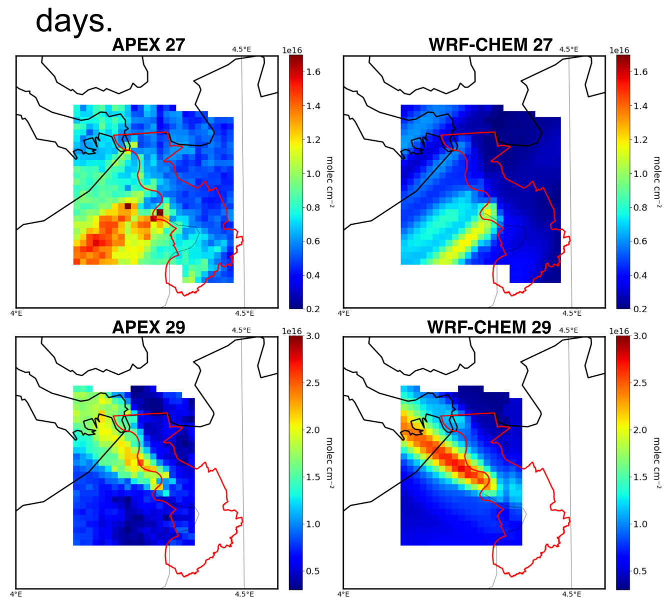
Relative bias (RB) between model and observations, calculated during daytime hours (9-17)

DAY	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29
RB	-11.3	-9	-35.3	-45.9	-57.2	-26.4	-30.3	-24.7	0.5	-44.7	-32.9	-36.2	-34.3	-27.7	-5.2

The WRF-Chem NO_2 output follows the overall diurnal shape seen by the measurements. However, there is clear overestimation of modelled NO_2 concentrations during the night, and an underestimation during the day. As seen on the Table above, the negative bias of the model is much higher during weekdays (-37% on average) than during Saturday (-13%) and Sunday (-4%). The weekends have been circled in green.

Comparison with airborne measurements

Tropospheric NO_2 vertical column densities (VCDs) are retrieved using the Airborne Prism EXperiment (**APEX**)[11,12] instrument, a pushbroom hyperspectral imager that integrates spectroscopy and 2D spatial mapping in one system. It is capable of mapping the NO_2 distribution at high spatial resolution (~75m x 120m). There were two flights over the Antwerp area in 2019, on June 27 and 29, both around midday, in order to compare with the corresponding TROPOMI overpasses. The APEX measurements have been regridded to correspond to the WRF-Chem 1x1 km² grid. These can be seen in the figures below, where each plot portrays the NO_2 tropospheric column from the corresponding source (APEX or WRF-Chem) over the two days.

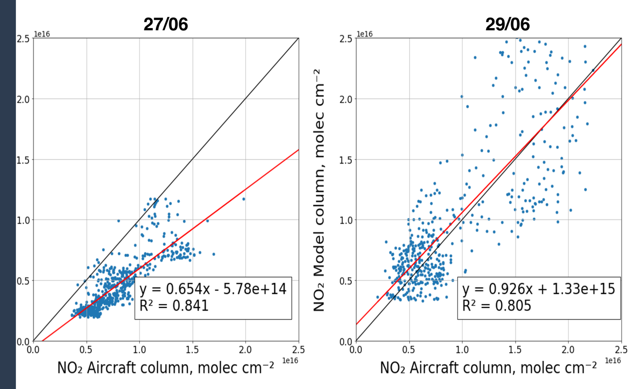


The NO_2 obtained from the model show similar features as the aircraft measurements, especially in the shape and direction of the plumes seen.

The model underestimates NO2 on the 27th of June (average model/station = 0.6) and shows better agreement on the 29th (1.01). This is in good consistency with the comparison with NO2 concentrations during daytime (see above).

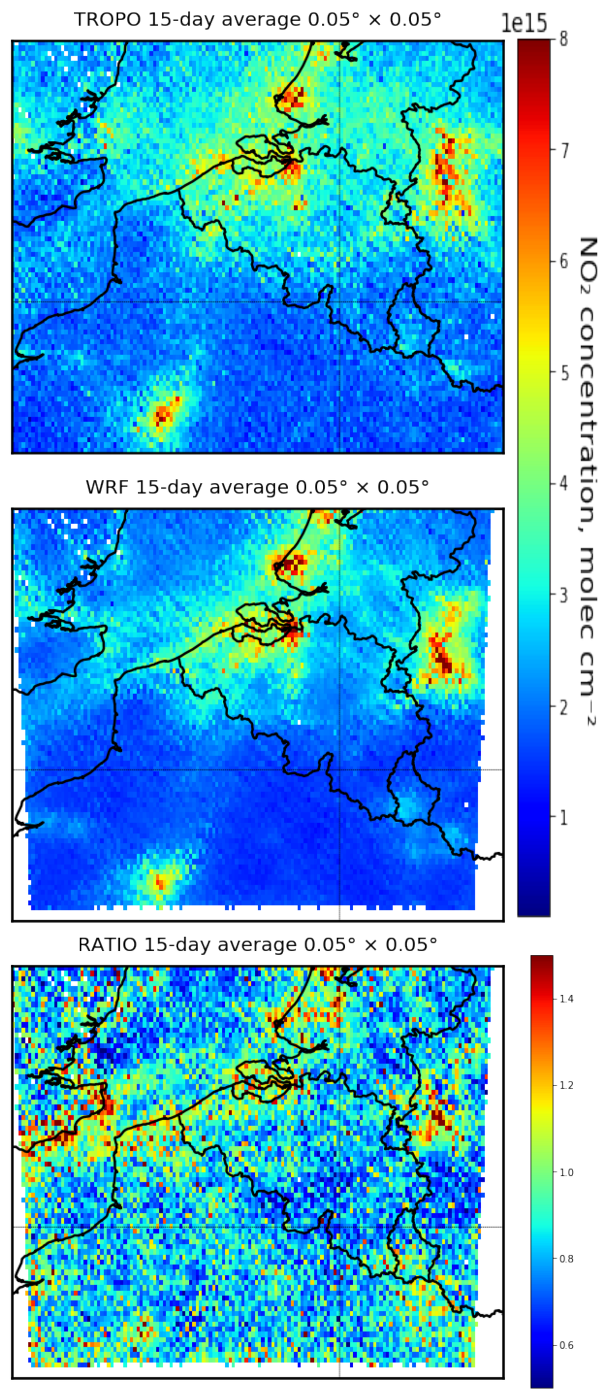
The model shows better agreement on the 29th, however there is an overestimation of NO_2 in the center of the plume where there is the highest concentration.

All of these comparisons are verified numerically by looking at the scatter plots of the model against aircraft columns, seen on the left.



Comparison with TROPOMI

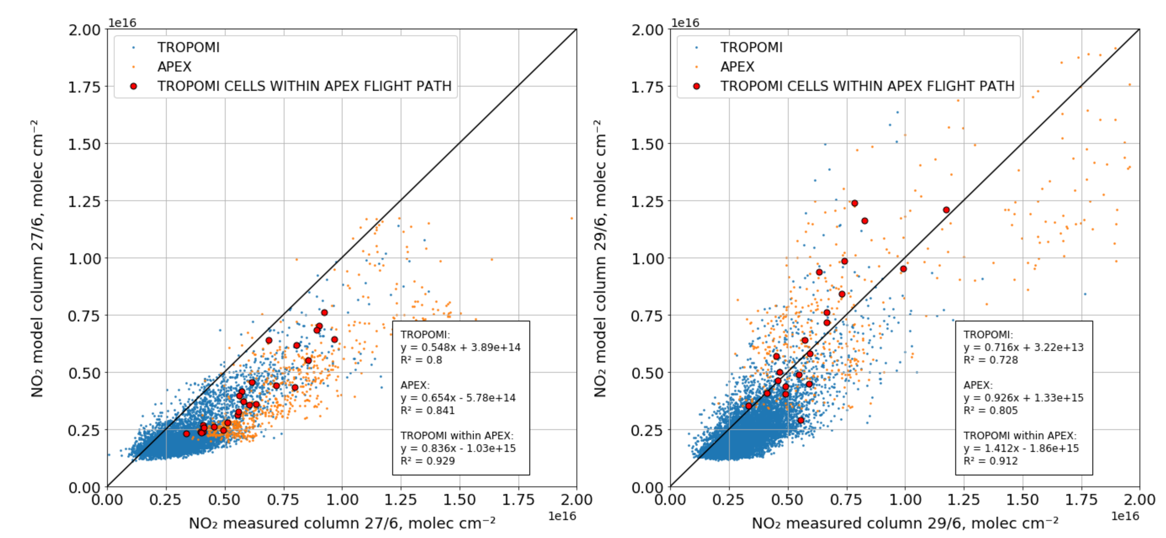
To facilitate a direct comparison between TROPOMI[13,14] and WRF-Chem, the model data was regridded into the TROPOMI resolution, using the latitude and longitude coordinates of the four corners of each TROPOMI cell. The WRF-Chem NO_2 columns were calculated using the TROPOMI averaging kernels. The appropriate quality filters were utilized, as described in the TROPOMI NO_2 ATBD.



Both measurements and model output were regridded onto a 0.05˚ x 0.05˚ grid, and averaged over the 15-day time period. The temporal average was done to remove some of the variability and noise of daily measurements.

WRF-Chem is mostly accurate at simulating the tropospheric NO_2 column over Western Europe, largely capturing the spatial patterns observed by TROPOMI. However, although the ratio of the two (model/TROPOMI) shows much noise, the model displays underestimations of TROPOMI NO_2 over less polluted areas (e.g. eastern Netherlands, Ardennes, Eiffel) and overestimations at many industrial/urban areas (Ruhr, Western Netherlands).

WRF-Chem performance was evaluated against the APEX and TROPOMI measurements on the days of the flight campaign. For TROPOMI, the entire model area and cells only within the APEX flight path were considered separately.



As discussed above, the model performs better on the 29 of June (a Saturday), and underestimates the NO2 column on the 27, suggesting too low NOx emissions on weekdays in the model. The slope of model-APEX on the 29/6 (close to 1) suggests a reasonably realistic model simulation of NO_2 distribution on that day, consistent with the comparison with station data. With respect to TROPOMI, the slope is much higher (~1.4 when selecting only pixels within the APEX flight region), suggesting that high NO_2 columns (~10^16 cm-2) are underestimated by TROPOMI.

Conclusions and Further Work

CONCLUSIONS

The results obtained in this work show that TROPOMI generally underestimates regions of high NOx emissions (seen especially over the Rühr region in Western Germany, and Amsterdam in the the Netherlands). Regions of low emissions, such as Flanders in Belgium, Eastern Netherlands and the North Sea, are overestimated by the satellite instrument.

FURTHER WORK

- Test a simple and crude "inverse-modelling" technique for improving the emissions in regions of high discrepancies between model and observed data.
- Improve the weekly cycle of emissions based on results of sensitivity tests.
- Compare the model output with the new, reprocessed TROPOMI NO_2 data when it becomes available for the simulated time period.

References and Data Access

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