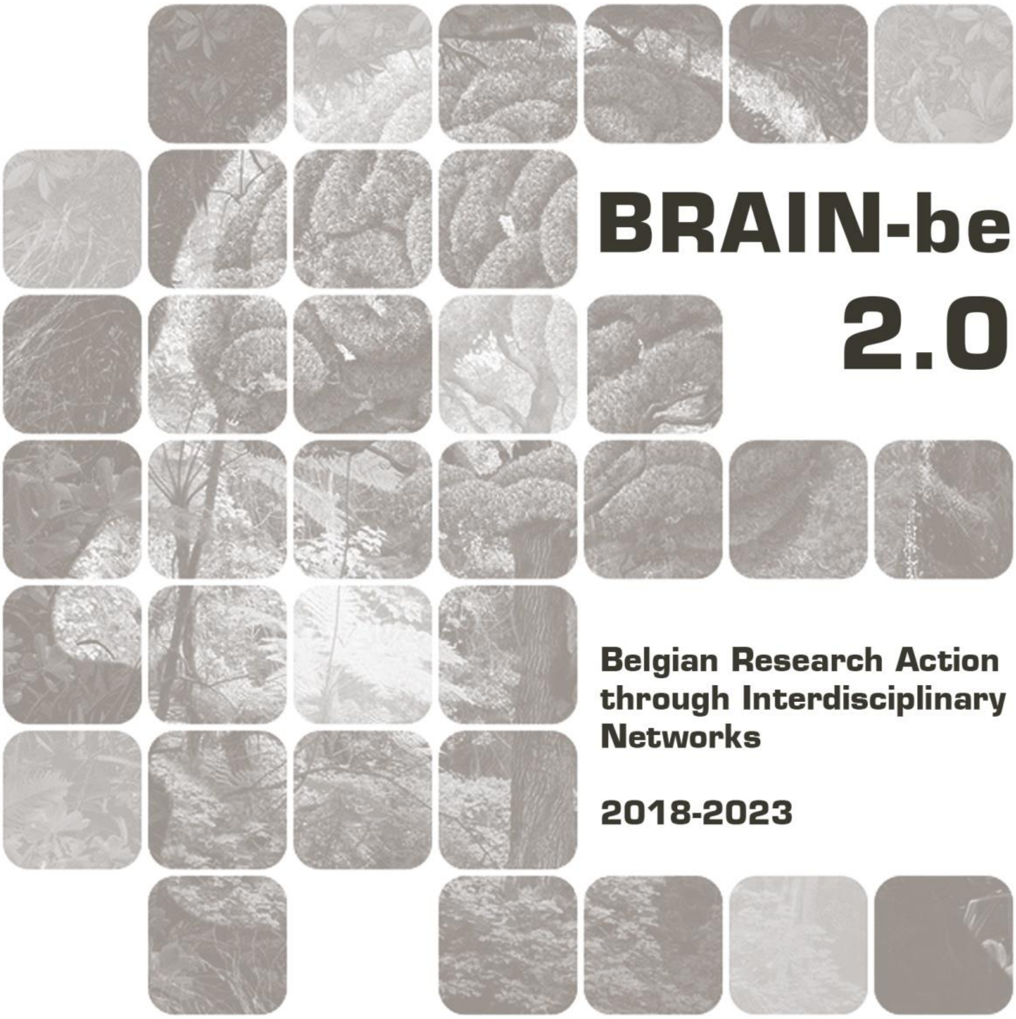

LEGO-BEL-AQ

Low-Earth and Geostationary Observations of Belgian Air Quality

TIJL VERHOELST (BIRA-IASB) – STEVEN COMPERNOLLE (BIRA-IASB) – JEAN-CHRISTOPHER LAMBERT (BIRA-IASB) – FRANS FIERENS (IRCEL-CELINE) – CHARLOTTE VANPOUCKE (IRCEL-CELINE)

Pillar 1: Challenges and knowledge of the living and non-living world

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Belgian Research Action through Interdisciplinary Networks

2018-2023

NETWORK PROJECT

LEGO-BEL-AQ

Low-Earth and Geostationary Observations of Belgian Air Quality

Contract - B2/191/P1/LEGO-BEL-AQ

FINAL REPORT

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ABSTRACT

As in most European countries, Air Quality (AQ) monitoring in Belgium has hitherto been relying mostly on in-situ measurements of near-surface concentrations, with geographical gaps between observations filled in with numerical models. However, a new global constellation of satellite sounders is being built to support detailed monitoring of AQ on the different relevant scales. As part of the Copernicus programme, the EC is contributing with Sentinel-5 Precursor TROPOMI (Low-Earth Orbit, LEO, since 2017) and the upcoming Sentinel-4 (geostationary, GEO) and Sentinel-5 (LEO) missions.

While offering near-contiguous observations of the entire domain (cloud cover permitting), observations from space also imply substantial challenges in terms of (1) spatial resolution (e.g., to resolve the Low Emission Zones), (2) relation between observed vertical column amounts and near-surface concentrations, and (3) synergistic use of multiple satellites with different observing geometries.

The aim of the LEGO-BEL-AQ project was to facilitate the use of this new-generation satellite AQ data by institutional policy makers and other stakeholders by advancing on the challenges identified above, with specific applications to the Belgian domain. Specifically, we demonstrated that policy-relevant features in the NO_2 distribution over major cities can be obtained with superresolution techniques, trading short-term temporal for spatial information. A synergistic use of satellite and in-situ data (using Regression Kriging) allows a pragmatic conversion from tropospheric columns to near-surface concentrations over the complete Belgian domain, and consequently also a confrontation to, e.g., the WHO annual exposure limit guideline of 10 microgram/m3 at the level of municipalities. Reaching the stakeholders implies a need for specific tailoring of the data and their presentation, and the use of communication channels not usually followed by the Earth Observation community.

2 SUMMARY

2.1 Context

Air Quality (AQ) in the European Low Countries is driven by complex processes covering a wide range of temporal and spatial scales, from point-like emissions of primary pollutants to intercontinental transport and the formation of secondary pollutants in a region with highly inhomogeneous land use. The responsibility for AQ regulations is distributed among different levels of public authority, from international and federal to regional and local. Informed policymaking for AQ regulation and sustainable (fossil) energy consumption requires tailored monitoring of AQ and of the impact of past and future regulations taken.

In general, governmental AQ monitoring relies on in-situ measurements of surface concentration, with geographical gaps between these (often sparse) observations filled in with interpolation techniques and numerical models incorporating meteorology and (proxies for) bottom-up emission estimates. However, a new constellation of Low-Earth Orbit (LEO) and geostationary (GEO) satellite sounders is becoming available, meant to support monitoring of AQ on the different relevant scales discussed above. Through its Copernicus programme, the European Commission is providing essential elements to this constellation, such as the Sentinel-5 Precursor (S5P, in LEO, launched in 2017 with the TROPOMI instrument on board), Sentinel-4 (GEO, to be launched in 2025), and Sentinel-5 (LEO, to be launched after 2025) suite of satellites.

While offering near-contiguous observations of the entire domain (cloud cover permitting), observations from space also imply substantial challenges that complicate the uptake by policy makers. First, in view of the size of the typical Low Emission Zones (LEZ) enforced in several (European) cities, there is a need to enhance the resolution of the satellite data. Second, there is a non-trivial relation between the column amount measured by a satellite and the near-surface concentrations actually leading to adverse health impacts. Third, the integration of observations from multiple LEO and GEO platforms into a synergistic system requires due care for their (relative) biases and for their different perception of atmospheric features.

2.2 Objectives

The overarching objective of the LEGO-BEL-AQ project was to facilitate the stakeholder uptake of satellite AQ data by advancing on the three challenges identified above, with applications specific to Belgium, its regions and cities. Essential to this was bringing together the expertise in satellite-based monitoring of tropospheric composition at BIRA-IASB with the in-situ and modelling expertise and the relation with key stakeholders and policy makers at IRCEL-CELINE. To tackle the first two challenges - enhance the spatial resolution and identify the relation between column data from satellites and near-surface concentrations - a mostly observational approach was adopted, meant to be complementary to the Chemical Transport Model method adopted in the Copernicus Atmosphere Monitoring Service (CAMS). More specifically:

The first challenge was addressed by developing oversampling/superresolution tools for satellite data, based on selective aggregation and geostatistical techniques, effectively trading temporal for horizontal resolution (WP1 of the project), and by applying these on S5P observations of tropospheric NO_2 over Belgium, with a focus on key cities such as Brussels, Antwerp, Ghent, and Liège (WP2).

Targeted outcomes were high-resolution maps and analyses of the temporal evolution (with reference to the policy in effect).

The second challenge was addressed by comparing the high-resolution S5P maps over Belgium with the in-situ measurements and the derived RIO interpolated data to assess under which conditions the high-resolution satellite maps of the NO_2 column can be considered as representative for the distribution of the NO_2 surface concentration (also in WP2);

The third challenge was addressed (WP3) by extending the OSSSMOSE observing systems metrology simulator at BIRA-IASB with dedicated GEO observation operators and applying these on the high-resolution maps produced in the 1st WP to quantify the impact of the particular viewing geometry of the geostationary sounders (located above the equator and observing our latitudes under very oblique angles) on the measurements and their consistency with the LEO platform data. Specific concerns are horizontal information smearing and obscuration effects.

To advance on the uptake by key stakeholders, a separate work package (WP4) was dedicated to user interaction: collecting requirements, ensuring availability of outcomes, and designing a roadmap for the future to take this work to an operational level.

2.3 Conclusions & recommendations

From the work performed in LEGO-BEL-AQ, the following key messages could be distilled:

a. The approach to **trade temporal for spatial resolution by aggregation and oversampling is successful** for LEO sounders such as S5P-TROPOMI, in particular when the temporal averaging window is of the order of several months.
b. Data sparseness, specifically due to cloud cover in winter, is an issue for smaller aggregation windows. **Filling data gaps due to cloud cover is advised against** as errors may be large.
c. The **increased resolution** obtained by oversampling S5P-TROPOMI tropospheric NO_2 data **brings valuable information** on the spatial distribution of NO_2 over Belgium, and on its medium to long-term temporal evolution.
d. The **spatial correlation between tropospheric NO_2 columns and corresponding near-surface concentrations as measured in-situ is (surprisingly) high** for longer temporal averages (seasonal and annual). Typical correlation coefficients range from 0.8 to 0.9.
e. This high correlation implies that a **pragmatic synergistic use of satellite and in-situ data**, using a linear regression + spatial Kriging interpolation of the residuals (a.k.a. Kriging Regression) to derive near-surface concentrations for the entire Belgian domain, is meaningful. Other avenues, such as integrating the satellite data as an additional proxy in the RIO model, also deserve exploration.
f. To study long-term temporal evolution, care must be taken to start from a **homogeneous level-2 data set**. For S5P-TROPOMI, ESA has just performed a full-mission reprocessing. These data should be the starting point for any trend study. Cal/Val results must be taken into account.
g. Still, **satellite and in-situ/model data show different long-term temporal behaviour**; Satellite data shown no/little impact from LEZ (decline in NO_2 columns in line with previous years), while in-situ and RIO show strong reductions in “urban” Belgium. Further analysis is needed.
h. Due to the fixed location of geostationary platforms, increasing spatial resolution through aggregation + oversampling does not work for these sounders (as demonstrated for GEMS). **An increase in spatial resolution will therefore require synergistic use of LEO+GEO data**.

i. **Spatial smearing biases due to large viewing and solar zenith angles** may be substantial for Sentinel-4 (and other geostationary sounders observing mid- to high latitude scenes).
j. **Interest for these applications in the Earth Observation community is strong, worldwide,** as illustrated by the collaboration initiated with the Japanese National Institute for Environmental Studies.
k. However, **awareness of the space component for AQ among environment specialists outside the EO domain is limited** and needs more exposure through channels other than those habitually used by EO scientists.
l. **Quick response times are valuable**. This became clear from our own experience in the CoViD-19 pandemic and from the discussions with FMI and potential stakeholders. To that end, an operational production of the tailored data is a prerequisite.
m. The **collaboration between BIRA-IASB and IRCEL-CELINE is already very productive** in making the bridge between EO and (in-situ) AQ monitoring (in Belgium).

Beyond the end of this project, recommended future research covers (1) further investigation into the discrepancy in temporal trends between satellite and in-situ measurements, (2) integration of the diurnal information to be obtained with Sentinel-4 when launched, (3) geographical extension to Europe and beyond, (4) inclusion of additional AQ components, such as PM2.5 estimates from aerosol optical depth data, (5) more advanced trend and exposure estimates including auxiliary data on e.g, population density, insolation, etc., (6) emission estimates, using for instance the flux divergence method, and comparison to bottom-up emission inventories, and finally, (7) further integration into the official monitoring systems.

Technical project outcomes are available through the website (http://lego-bel-aq.aeronomie.be) and/or by request to the project Principal Investigator (tijl.verhoelst@aeronomie.be).

2.4 Keywords

Air Quality trends – Nitrogen dioxide – Low Emission Zones - Remote sensing – Copernicus – ESA - Space - Satellite observations – Sentinel-5P TROPOMI - Low-Earth orbit – Sentinel-4 - Geostationary – Oversampling – Superresolution – Synergistic data use

3 INTRODUCTION

Air Quality (AQ) in the Low Countries, i.e., the coastal region of north-western Europe, consisting of Belgium, the Netherlands, and Luxembourg, is driven by complex processes covering a wide range of temporal and spatial scales, from point-like emissions of primary pollutants in a region with highly in-homogeneous land use to intercontinental transport and the formation of secondary pollutants. The responsibility for AQ regulations is distributed among different levels of public authority, from international and federal to regional and local. For instance, in recent years several major Belgian cities implemented so-called Low Emission Zones (LEZ), targeting emissions by road transport, albeit each with its own criteria and time schedule (see Table I).

Table I: List of city-level or regional Low Emission Zones (to be) enforced in Belgium as of 2024.

City or region	Start	Details
Antwerp	1 February 2017	Inner city; Stricter criteria on 1 Jan. 2020 and 1 Jan. 2025.
Brussels	1 January 2018	Almost the entire area within the large ring road R0; Stricter criteria on an almost yearly basis.
Ghent	1 January 2020	Almost the entire area within small ring road R40; Stricter criteria in 2025.
Wallonia	TBC	Unclear whether it will come into legislation.

AQ assessment in Belgium has hitherto been relying mostly on in-situ measurements of near-surface concentrations. While this measurement network is extensive, it remains relatively sparse when considering the large variability in pollutant concentrations over the Belgian domain, with many interesting regions (both urban and rural) and large heterogeneity not being sampled. Geographical gaps between observations are filled in with interpolation technique RIO (e.g., the RIO model, Janssen et al., 2008) and numerical modelling, see Figure 1 for an example map.

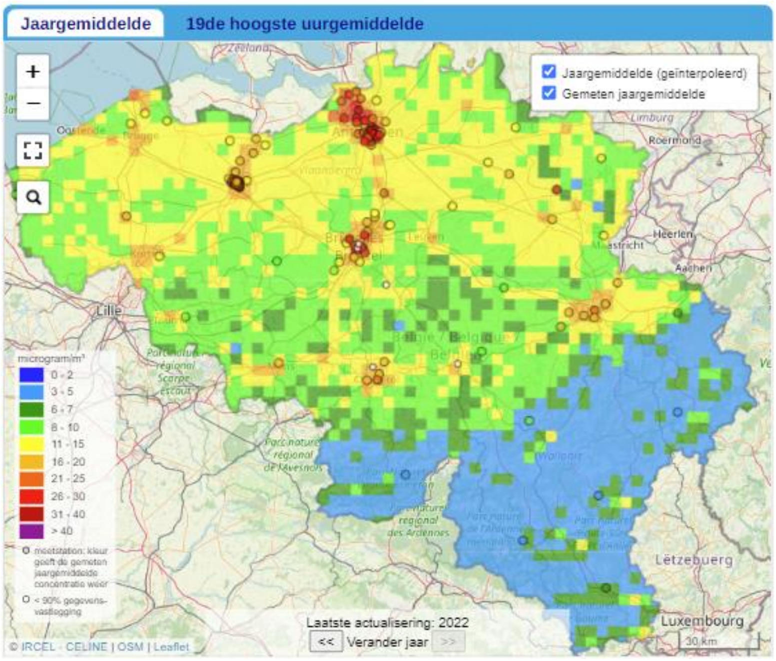

Figure 1: 2022 annual average NO_2 near-surface concentration as measured in-situ (round markers) and interpolated/modelled over the entire Belgian domain using the RIO interpolation model on a 4x4km² grid.

Pollutant concentration estimates thus depend largely on the land use proxy that supports the RIO interpolation model. While more advanced concentration estimates can be obtained by coupling with an additional model including bottom-up emission estimates and meteorological information, as implemented in the RIO-IFDM model, the underlying set of actual observations remains sparse and the emission inventories are not necessarily complete or accurate.

In this context, we can anticipate -- even for Belgium -- added value in the Earth Observation (EO) data obtained by the first of the Copernicus Sentinel series of AQ satellite sounders in Low Earth Orbit (LEO), Sentinel-5 Precursor TROPOMI, which covers the entire Belgian domain (cloud cover permitting) on a daily basis since May 2018. Moreover, Belgium will soon be observed also from a geostationary vantage point (GEO), providing hourly data (during daytime) with the Sentinel-4 UVN series and with follow-on LEO sounders (the Sentinel-5 UVNS series), all part of a global constellation of AQ-dedicated satellites that is being deployed (Figure 2).

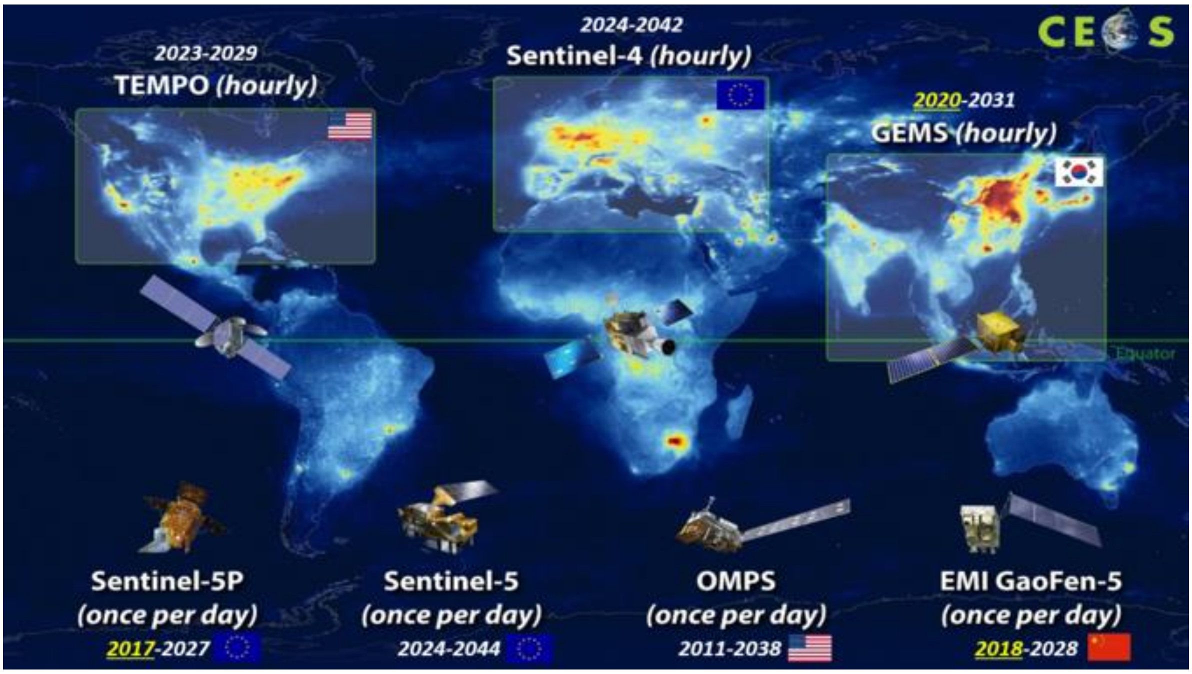

Figure 2: Collage illustrating the AQ-dedicated constellation of LEO and GEO sounders currently being put in space by various space agencies in the context of large (inter-)national programmes. Figure adapted from a collage made by the Committee on Earth Observation Satellites (CEOS).

This constellation builds on pioneering work with missions such as the GOME instrument on board of ERS-2 (1995-2003), the GOME-2 instruments on the MetOp A/B/C satellites (2007-now), and SCIAMACHY on board of ENVISAT (2002-2012). Reductions in NO2 columns over Europe but increases over China were already reported using the GOME data by Richter et al. (2005). More recently, the CoViD-19 related lockdown measures implemented all over the world provided exceptional insight in the relation between human activity and air pollution, resulting in a large body of scientific literature on satellite-derived AQ assessments (e.g., Bauwens et al., 2020; Bassani et al., 2021; Cooper et al., 2022, Fioletov et al., 2022; Zhao et al., 2022; Levelt et al., 2022). With the ever-increasing spatial resolution of the satellite sounders, also the urban environment has become a focal point (e.g., Ialongo et al., 2020; Petetier et al., 2023; Pommier, 2023).

4 STATE OF THE ART AND OBJECTIVES

Uptake of the EO AQ data introduced above by Belgian (and European) stakeholders has however remained limited. In fact, to realize the full complementary impact of this constellation of LEO and GEO satellites, i.e. to make their observations fit-for-purpose for AQ assessment applications at the different scales, several technical and scientific challenges need to be addressed. These include in particular the need to:

(1) Improve even further the horizontal resolution to map and monitor pollution on the scale of local policies (e.g. the Low Emission Zones established in several large Belgian cities);

(2) Determine the non-trivial relation between the (tropospheric) pollutant column measured by satellite instruments and the surface concentration measured by in-situ networks;

(3) Evaluate to what extent differences in measurement geometry and sensitivity between LEO and GEO observations lead to a different perception of atmospheric details (e.g., through spatial blurring and obscuration effects), hence, to an internal incoherence of the satellite constellation.

The project aimed to advance the state-of-the-art on these three topics, with applications specific to Belgium, its regions and cities, by bringing together the expertise in satellite-based monitoring of tropospheric composition at BIRA-IASB with the in-situ and modelling expertise and the relation with key stakeholders makers at IRCEL-CELINE. For challenges (1) and (2), a mostly observational approach was adopted, meant to be complementary to the CTM method adopted in the Copernicus Atmosphere Monitoring Service (CAMS).

As in much of the literature reviewed above, the LEGO-BEL-AQ project focused on tropospheric NO_2, which is both a direct pollutant with adverse pulmonary health effects and a precursor to secondary pollutants such as ozone and fine particulate matter (PM2.5), both major contributors to mortality related to air pollution. NO_2 was recently estimated to cause 1.85 million Asthma cases in children, globally and annually, either directly or by an unknown co-emitted pollutant (Anenberg et al., 2022). The main source of NO_2 is oxidation of atmospheric nitrogen during combustion processes at high-temperatures in transport, power plants, industry, and residential heating. In Belgian urban areas, on-road transport is currently expected to be the main source but this may shift to domestic heating when electrification of transport occurs more rapidly than that of heating.

5 METHODOLOGY

As an overview, the work plan of the project targeted advances:

- On the first challenge by developing oversampling tools for satellite data, based on selective aggregation (trading temporal for horizontal resolution) and geostatistical interpolation techniques (WP1 of the project), and by applying these on S5P observations of tropospheric NO_2 over Belgium, with a focus on key cities such as Brussels, Antwerp, Ghent, and Liège (WP2).

- On the second challenge by comparing the high-resolution S5P maps over Belgium with the in-situ measurements and derived gridded products (e.g., the RIO data set) to assess under which conditions the high-resolution satellite maps of the NO_2 column can be considered as representative for the distribution of the NO_2 surface concentration (also in WP2);

- On the third challenge by extending the OSSSMOSE observing systems metrology simulator at BIRA-IASB with dedicated 3D GEO observation operators and applying these on representative atmospheric model fields, digital elevation models and orbital parameters of S5P, Sentinel-4 and Sentinel-5, in order to quantify the significance of these effects on the internal coherence of the CEOS LEO+GEO AQ Constellation (WP3).

To ensure uptake by key stakeholders, a separate work package (WP4) was dedicated to user interaction: collecting requirements, ensuring availability of outcomes, and designing a roadmap for the future to take this work to an operational level.

Extensive technical details on the adopted methodologies and outcomes are provided in the next Section (6).

6 SCIENTIFIC RESULTS AND RECOMMENDATIONS

For the purpose of traceability, we describe first the data sets that were used (not produced) in the project. Subsequent subsections detail the specific methodologies and results for each of the three identified challenges.

6.1 Satellite, in-situ and model data sets

6.1.1 S5P-TROPOMI tropospheric NO_2 column data

The TROPOspheric Monitoring Instrument (TROPOMI) was launched on board of ESA's Sentinel-5 Precursor (S5P) early-afternoon (13:30 local solar overpass time at the equator) LEO satellite in October 2017. Its large wavelength coverage, from the UV to the near-IR, allows accurate and high spatial resolution (5.5x3.5km^2 since 6 August 2019) observations of a large suite of trace gases (Veefkind et al.,2012), with almost daily global coverage. The operational NO_2 product(s), publicly available since May 2018, are derived in a three-step process based on the QA4ECV community retrieval approach (Boersma et al., 2018) and on the DOMINO/TEMIS algorithm (Boersma et al., 2007,2011), as already applied to other heritage and current satellite sounders. It relies on a DOAS approach for the slant column retrieval and a Chemical Transport Model (CTM) for the stratosphere-troposphere separation. The latest processor improvements are described in vanGeffen et al. (2022) and extensive ground-based validation is reported in Ialongo et al., (2020), Judd et al., (2020) and Verhoelst et al. (2021), with the most recent validation results published in the S5P ATM-MPC VDAF Quarterly Validation Reports, available online at https://mpc-vdaf.tropomi.eu/. A key finding in these validation studies is a tendency for S5P-TROPOMI to underestimate tropospheric NO_2 columns by approximately 30%. As this bias is mostly multiplicative, it does cancel out in relative differences, e.g., from year-to-year. To ensure as accurate and homogeneous a data set as possible, we used here the full-mission reprocessed tropospheric NO_2 column data based on processor version v2.4.0 (as released on 2 March 2023 and available at https://dataspace.copernicus.eu , extended beyond the end date of the reprocessing with the offline (OFFL) v2.4.x (and above) processing. To improve on the representation of the strong spatial gradients to be expected over Belgium, a sensitivity analysis was performed using a modified product, for which the original a priori vertical profile (from the TM5-MP global analysis at 1x1degree^2) was replaced with profile information from the regional (Europe only) CAMS ensemble (0.1x0.1degree^2), as described in Douros et al. (2022). Quality filtering was performed using the provided *qa_value* information, which is a single-number summary of a large set of quality indicators. The threshold was put at 0.75, as recommended in the Product Readme File (PRF).

6.1.2 Near-surface in-situ concentration measurements

In Belgium, monitoring of air quality lies within the responsibility of the regions. NO_2 concentrations in ambient air are measured with the reference method NBN EN 14211 (2012) : "Ambient air - Standard method for the measurement of the concentration of nitrogen dioxide and nitrogen monoxide by chemiluminescence" in the three regions. In 2022 the Belgian network included 97 stations measuring NO_2 of which 56 are located in Flanders, 16 in Brussels and 25 in Wallonia. The stations cover the whole territory and all stations types are represented (urban, rural, traffic and industrial oriented), with some more density in urban and industrial areas. Measurements of this telemetric network are published hour-by-hour on the websites of the Belgian regional and interregional environment agencies. Within the cities of Ghent, Antwerp, Brussels and Liège, the automated network is complemented with a network of passive tubes, sampling bi-weekly, mainly to assess the concentrations in street canyons, but these data were not used in the current project.

6.1.3 RIO model data

Although the Belgian measurement network is quite extensive, obtaining coverage of the full Belgian territory requires estimates in between measurement sites. Near-surface air quality concentrations at locations where no in-situ measurements are available are filled in via the interpolation technique RIO (Janssen et al., 2008). The core of the RIO model is the Ordinary Kriging method, but to fulfil the criterium of equal spatial representation of the samples, a detrending and retrending step is needed. These steps before and after the Kriging interpolation, account for the local characteristics of the sampled locations and interpolation locations. The local characteristics are defined by a parameter derived from the Corine Land Cover classes (CLC2006), the so-called 'spatial proxy' and show a clear relation with the long-term series of air quality concentrations. By using the information on land cover, the gaps between the fixed measurements can be estimated on an hourly basis and covering the whole area with a resolution of 4 km. The RIO model is operational at the Belgian Interregional Environment Agency (IRCEL) to evaluate the air quality in Belgium in near-real time (hourly) and for assessment purposes (historical). RIO results are equally used as background within the high resolution modelling chain of RIO-IFDM and ATMO-Street which use detailed information on meteorology and emissions, inherently subject to uncertainties (detailed documentation available at: https://www.irceline.be/en/documentation/models/rio-ifdm).

6.1.4 Administrative boundaries

To be able to aggregate the NO_2 data over the areas corresponding to the different levels of public authority, administrative boundaries for Belgian municipalities, provinces, and regions were obtained from the Belgian Federal Public Service (FPS) Finance - General Administration of Patrimonial Documentation (GAPD), through the Geoportal of the Belgian federal institutions (http://geo.be). These boundaries are provided in either EPSG:3812 (Belgian Lambert 2008) or EPSG:4258 (European Terrestrial Reference System, 1989) projection as shape files and converted to WGS84 (World Geodetic System 1984) coordinates for the present work.

6.2 WP1: Spatio-temporal aggregation and oversampling

A key aim of this project was to produce satellite-based maps of NO_2 at a spatio-temporal resolution corresponding to the scale of AQ policies that are being implemented, while keeping in mind the strengths and weaknesses of the (UV-Vis) remote sensing. In practice, this implies the need for spatial oversampling to resolve the urban and industrial NO_x features. Achieving true superresolution requires some temporal aggregation, which is also needed to deal with persistent cloud cover in winter, and to limit the impact of other short-term meteorological conditions. To address the needs of local stakeholders, we optionally aggregate the high-resolution data to administrative entities, such as the Belgian regions and the 581 Belgian municipalities. These processing steps are described below.

6.2.1 Spatial oversampling/superresolution

The aim here is to trade the temporal resolution of individual daily measurements for increased spatial resolution, which is an acceptable trade-off in view of the spatial and temporal scales of the AQ policies that are being implemented, e.g., the LEZ zones. Many approaches have been explored in the EO community to produce L3 (i.e., regularly gridded) data on a grid finer than the native satellite pixel size, with varying degrees of complexity (and hence CPU needs) and resolution gain (see e.g., De Foy et al., 2009, Clarisse et al., 2019, Cersosimo et al., 2020, and references therein). As a tractable starting point, we have chosen here to use a straightforward area-weighted pixel averaging on a regular lat-lon grid, as implemented in the HARP toolbox (https://atmospherictoolbox.org/harp/ and http://stcorp.github.io/harp/doc/html/operations.html). This routine calculates the area overlap between each intersecting L2 pixel and target L3 grid cell, in 2-D geometry (an acceptable approximation given the small pixels and grid cells targeted here, both having typical sizes of a few km). This area overlap is used as a weight in the averaging for a given grid cell. Mathematically:

$$VCD_{L3,j} = \frac{\sum_i w_i VCD_{L2,i}}{\sum_i w_i}, \text{ with } \mathrm{w_i} = \frac{\mathrm{Area_{L2,i \cap L3,j}}}{\mathrm{Area_{L3,j}}}$$

where *i* runs over all L2 pixels intersecting the *j*-th L3 grid cell within the temporal averaging window. The sum of the weights, $\sum w_i$, is available in the output files in order to have some quantitative information on the density of the data behind the L3 product. Temporal averaging windows must be chosen large enough to ensure $\sum w_i >> 1$ for each grid cell, because only then will we see a reduction in correlations between neighbouring grid cells due to contributions from the same L2 pixels. Indeed, as the orbit repeat cycle of S5P is 227 orbits within 16 days, L2 pixels will have slightly different locations for each of 15 consecutive days and this is essential to achieve true superresolution and not just an oversampled image.

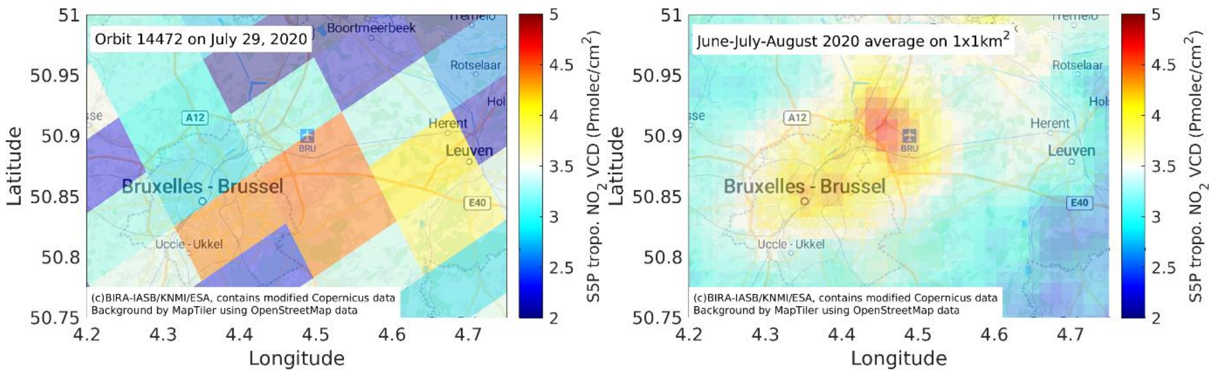

Figure 3: Comparison between S5P-TROPOMI tropospheric NO_2 columns measured in a single orbit passing over Brussels, with pixel sizes of at best 5.5x3.5 km^2 (left-hand panel), and the gridded and oversampled 3-month average for June-August 2020 at a 1x1 km^2 grid resolution (right-hand panel). The area covered and the color scale are identical in both panels. Note that activity levels at this time were still impacted by CoViD-19 related restrictions.

Figure 6 demonstrates the gain in spatial resolution achieved when gridding 3 months of (summer time) data on a grid corresponding to 1x1 km^2 at the map center (0.009x0.0143 degree^2), allowing the differentiation between Brussels city center, the logistics area near the national Airport in Zaventem in the North, and the university town of Leuven to the East, which was in the advected plume of Brussels on July 29, 2020, but doesn't seem to be a significant source itself.

While the true resolution of the oversampled image is probably not as high as the 1x1 km^2 sampling chosen here, the observed structures suggest a resolution well above that of a single S5P-TROPOMI overpass. Further evidence that the combination {3 months, 1km} is a good compromise can be obtained from a semi-variogram (Matheron, 1963), as illustrated in Figure 4. The nugget being close to zero indicates that there are neither large unresolved structures nor significant random noise at the

1km spatial scale, both of which would lead to non-zero variance at the smallest lags (Dominy et al., 2003; Francois-Bongarcon et al., 2004).

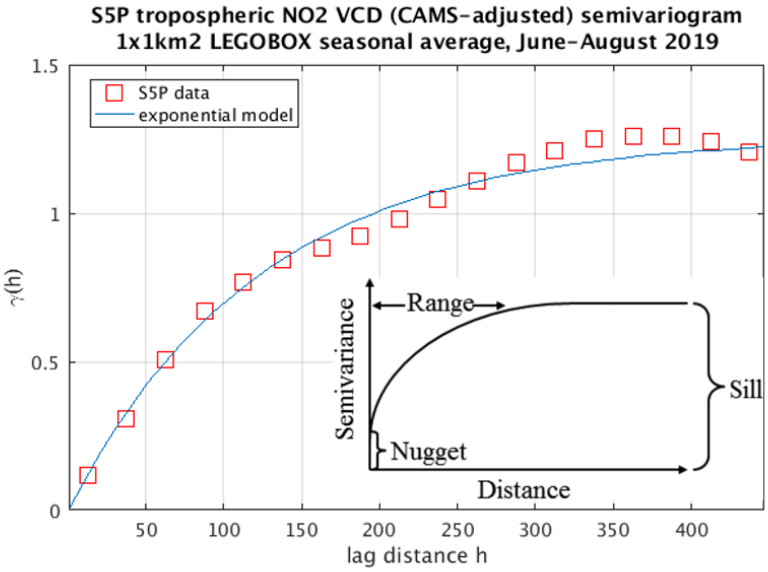

Figure 4: Semi-variogram of the oversampled S5P data for summer 2019 on all of Belgium (and part of the neighbouring countries), and the corresponding fitted exponential model. Explanatory inset courtesy of Samiran Banerjee (North Dakota State University).

6.2.2 Meteorological constraints

While daily maps, based on only one or a couple of overpasses, can reveal very specific events and conditions, they will in most cases suffer large gaps due to clouds or other retrieval-complicating conditions (such scenes being filtered out through the *qa_value* filter). Moreover, at these temporal scales, the observed NO_2 field is heavily affected by the meteorological conditions and as such not necessarily representative for the typical pollution levels. In view of the objectives of the current work, interesting temporal windows are rather in the months-to-season range. While gap-free NO_2 maps of Belgium can be obtained in summertime from as little as 1 week of data, heavy cloud cover in winter translates into minimum temporal averaging windows of at least a month (Figure 5).

A couple of days with strong winds from a single prevailing direction (often from the South-West in Belgium) may impact strongly a weekly or monthly NO_2 map. Depending on the targeted use of the map, it may be advisable to filter out such days with strong winds. However, we found that for averages over multiple months, the variability in wind direction ensures that emission sources still pop up, as these “stack up” consistently, while the advected plumes are diluted over the various directions. Consequently, the data shown here were not filtered on wind speed.

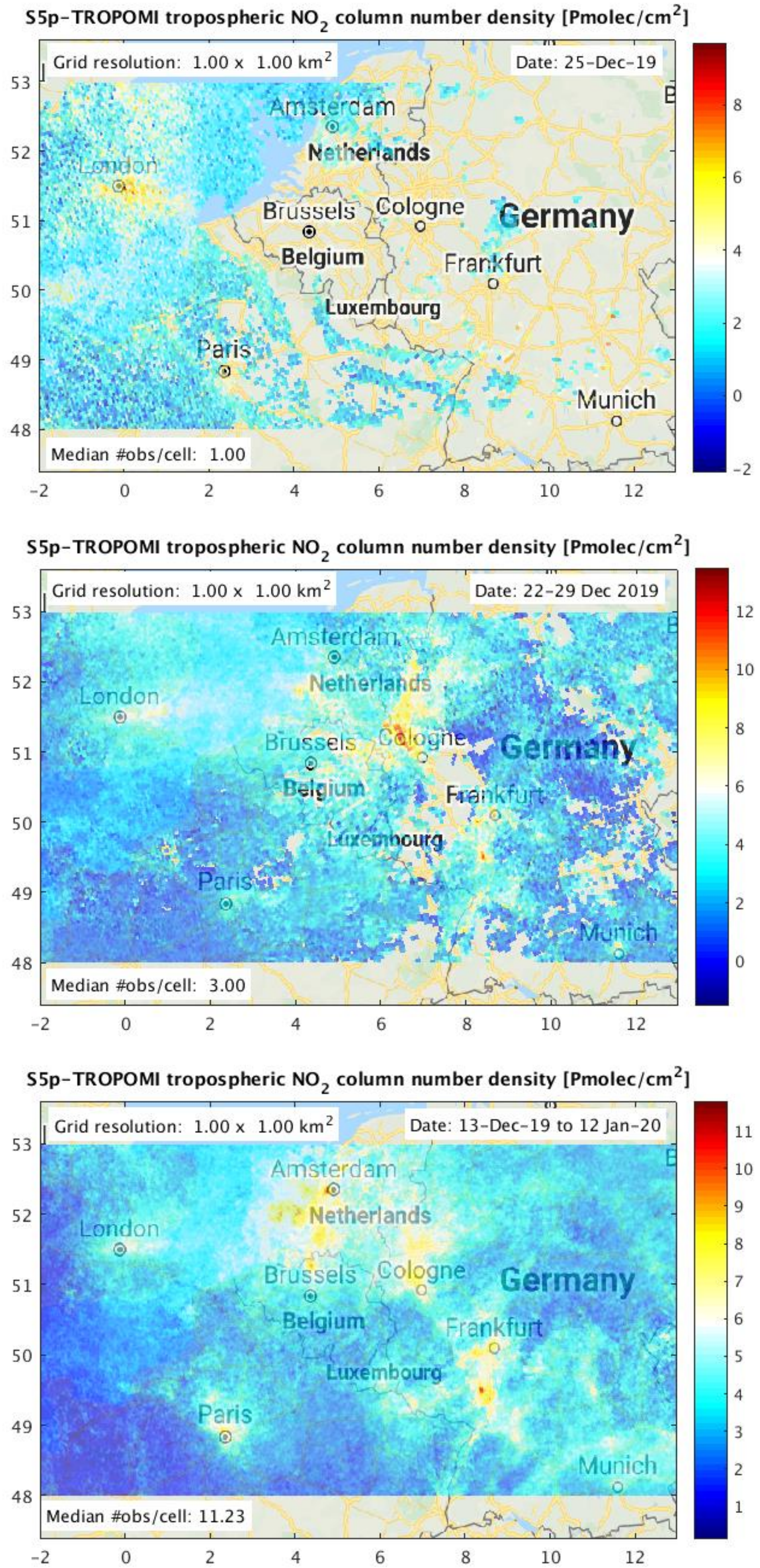

Figure 5: Illustration of typical spatial data gaps due to cloud cover for either a single day of winter-time measurements (top panel), a week of data (middle panel), and a month of data (bottom panel).

6.2.3 Diurnal variability

While the nominal overpass time of S5P is 1:30pm (local solar time for the sub-satellite point at the equator), mid- and high-latitude regions may benefit from multiple overpasses per day. For Belgium, the main overpass occurs around 12:30pm UTC, and additional overpasses occasionally occur one orbit earlier (11am UTC) and one orbit later (2pm UTC). As the concentrations of NO_x depend strongly on the insolation (and on variations in emissions throughout the day), mixing overpasses of different local solar times may lead to artificial structures (e.g., striping). This effect is most pronounced when including low-sun observations obtained close to twilight. For this reason, we remove observations with Solar Zenith Angle (SZA) >75 degrees. Particularly in winter this implies some data loss.

6.3 WP2(a): Application on S5P TROPOMI NO2 data over Belgium

6.3.1 Main Belgian NO_2 hot spots

Figure 6 and Figure 7 contain, respectively, a multi-year (2019-2023) summer map of the entire Belgian domain and a set of city-level maps for four major urban areas. A temporal average over May, June, and July was chosen as the best compromise between normal (non-vacation) activity levels and short photochemical NO_2 lifetime, to keep blurring due to advection as small as possible.

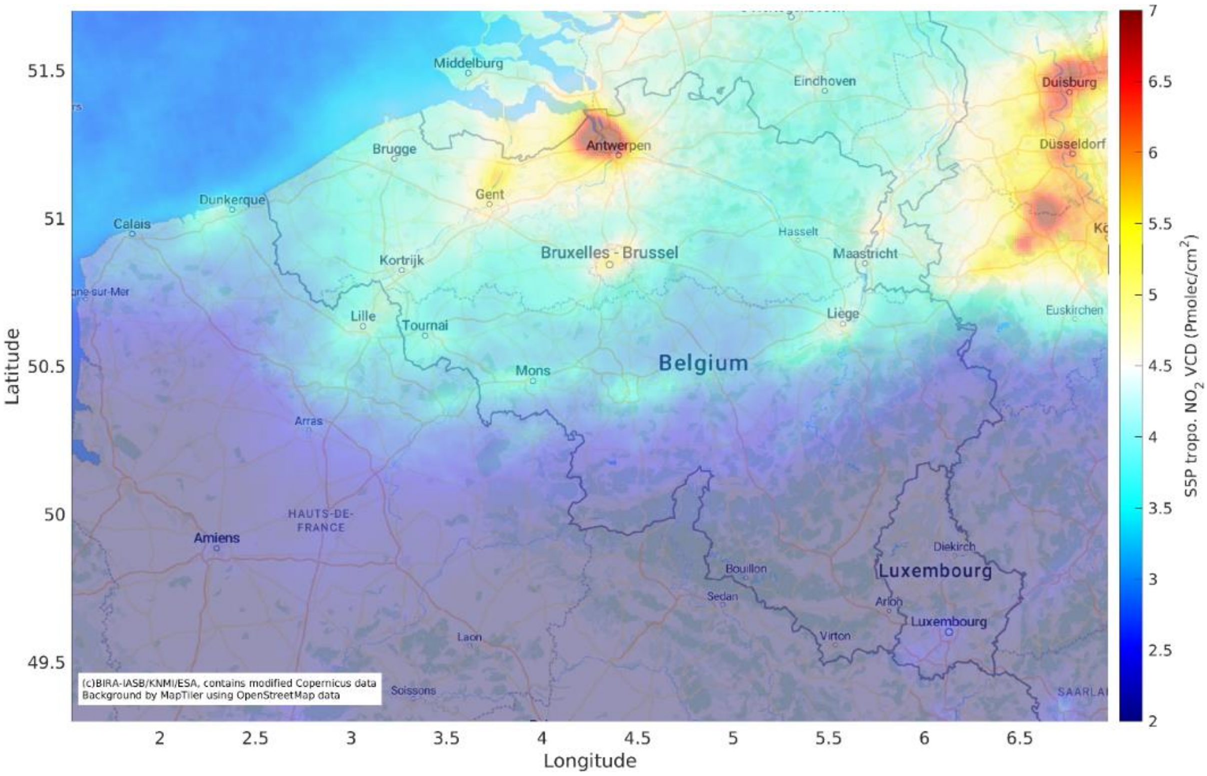

Figure 6: Illustration of the result of spatial oversampling and temporal aggregation on S5P-TROPOMI tropospheric NO_2 columns, for all of Belgium, using 5 years of May-June-July data (2019-2023).

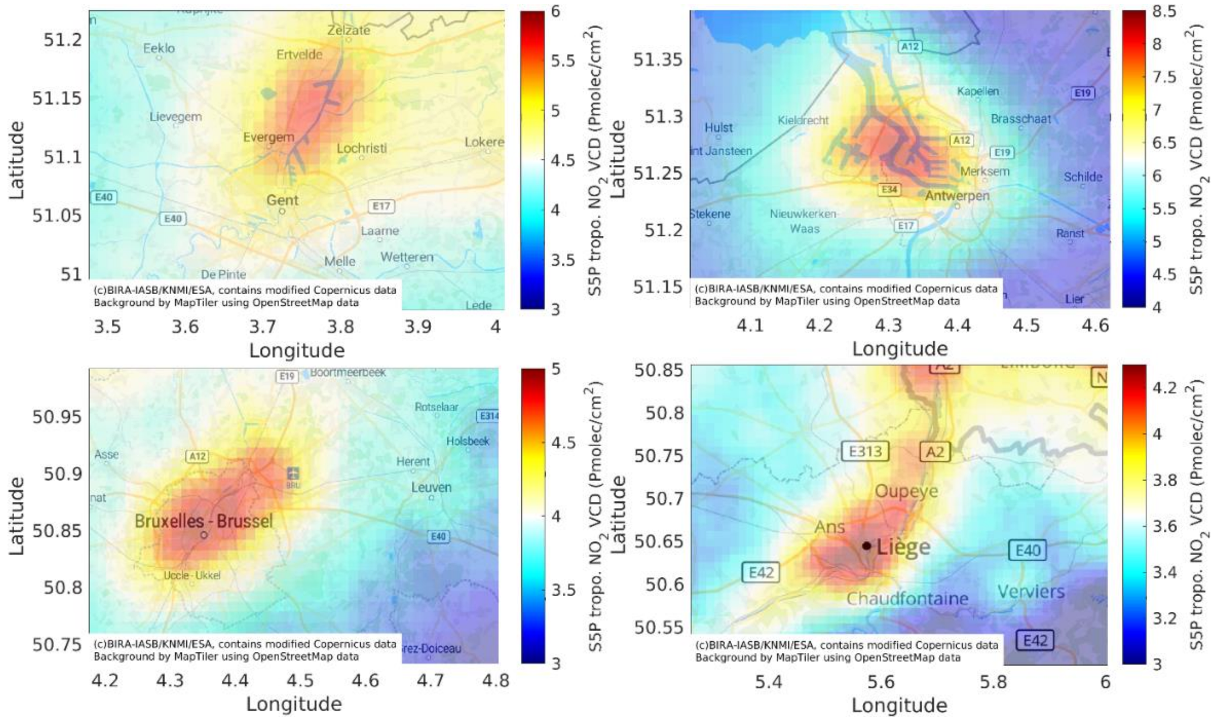

Figure 7: Similar to Figure 6 but focussing on four interesting urban/industrial areas with dedicated colour scales (note that absolute levels are very different for the four urban areas).

The overall picture for Belgium is one of a strong North-South gradient, with peak pollution levels observed over Antwerp, followed by Ghent, Brussels, Liège and a few urbanized corridors along either highways or waterways. During these summer months, the harbours of Antwerp and Ghent display much more elevated NO_2 levels than what is observed over the city center. Note that there is nevertheless a significant difference in absolute NO_2 levels between both cities, summer-time peak values over the Antwerp harbour being some 40% above those over the Ghent harbour. Note also that these long-term averages reveal the persistent structures only. In case of unfavorable wind conditions, the pollution plumes do cover the city centres (though perhaps to some extent at an altitude that safeguards the local population). Over Brussels, we see both the city center and the industrial/logistics hub near the national airport in the North-East (see also Figure 3). Liège is another NO_2 hot spot with elevated levels both over the city center and in the industrial zone of Herstal (North-East of the city center), and also over the nearby rural town of Visé (West of the "A2" highway marker in the image). A potential contributor there could be a cement factory.

6.3.2 At the level of municipalities

Municipalities are typically the small-scale stakeholders targeted by this work. As such, one of the products developed in the project are S5P-TROPOMI average tropospheric NO_2 columns for each Belgian municipality, as a monthly, seasonal, and yearly scale. Example maps of these aggregated data are visualized in Figure 8. These example maps contain spring-time averages (March through May) at overpass time for 2019 and 2020. The clear reduction of NO_2 levels in 2020 is a consequence of both the reduced activity levels (CoViD-19 lockdown measures) and a particularly sunny spring.

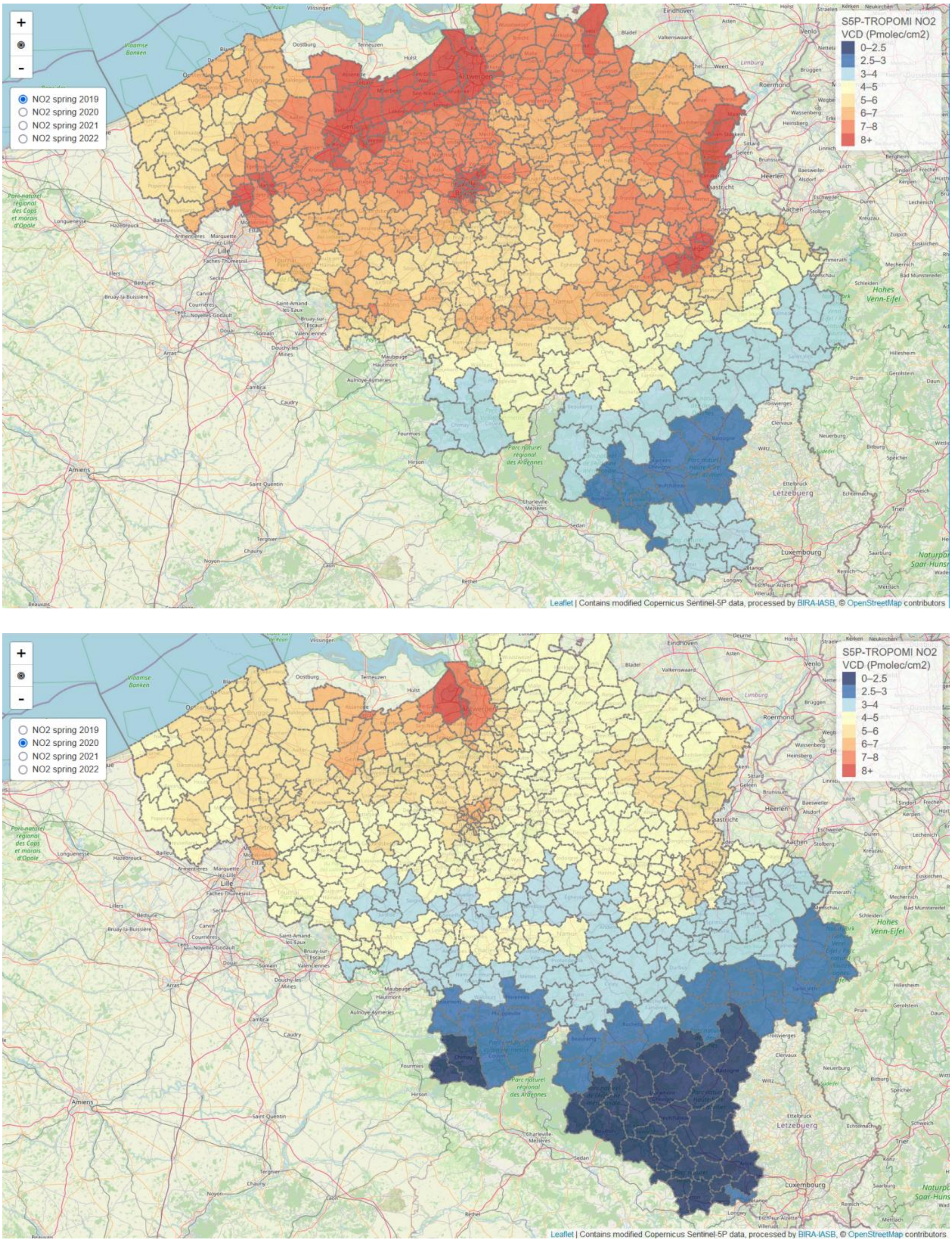

Figure 8: Spring-time (March-April-May) noon mean tropospheric NO_2 vertical column density (VCD) from S5P-TROPOMI observations aggregated and oversampled using the tools described in Section 6.2 and averaged within municipality boundaries. Top panel corresponds to spring 2019, the bottom panel to spring 2020. The significant reduction in NO_2 levels was due to both the reduced activity levels (CoViD-19 lockdown measures) and particularly sunny weather.

One of the driving questions for the developments reported here concerns the impact of (local) policies meant to improve air quality, such as LEZs, and whether this impact can be observed with space-based instrumentation that allows observations of the entire Belgian domain, as a complement to the accurate but relative sparse in-situ instruments. While the LEZ in Antwerp and Brussels were initiated just before the launch of S5P-TROPOMI, initial criteria were relatively modest, e.g., Brussels banning only diesel cars not satisfying the Euro-1 norm and putting no constraints on petrol cars. Criteria have been gradually tightened since, and one could thus expect a trend in traffic-related pollution levels over the past 5 years. Figure 9 visualises the evolution of the monthly mean tropospheric NO_2 vertical column densities (VCDs) observed by S5P-TROPOMI over three municipalities:

- Brussels, i.e. the commune in the very heart of the Brussels region (and thus of the Brussels LEZ),
- Leuven, a strongly growing provincial university town (without a LEZ, but still adopting various green policies),
- Sankt Vith, a typical rural, background commune, close to the border with Germany and Luxemburg.

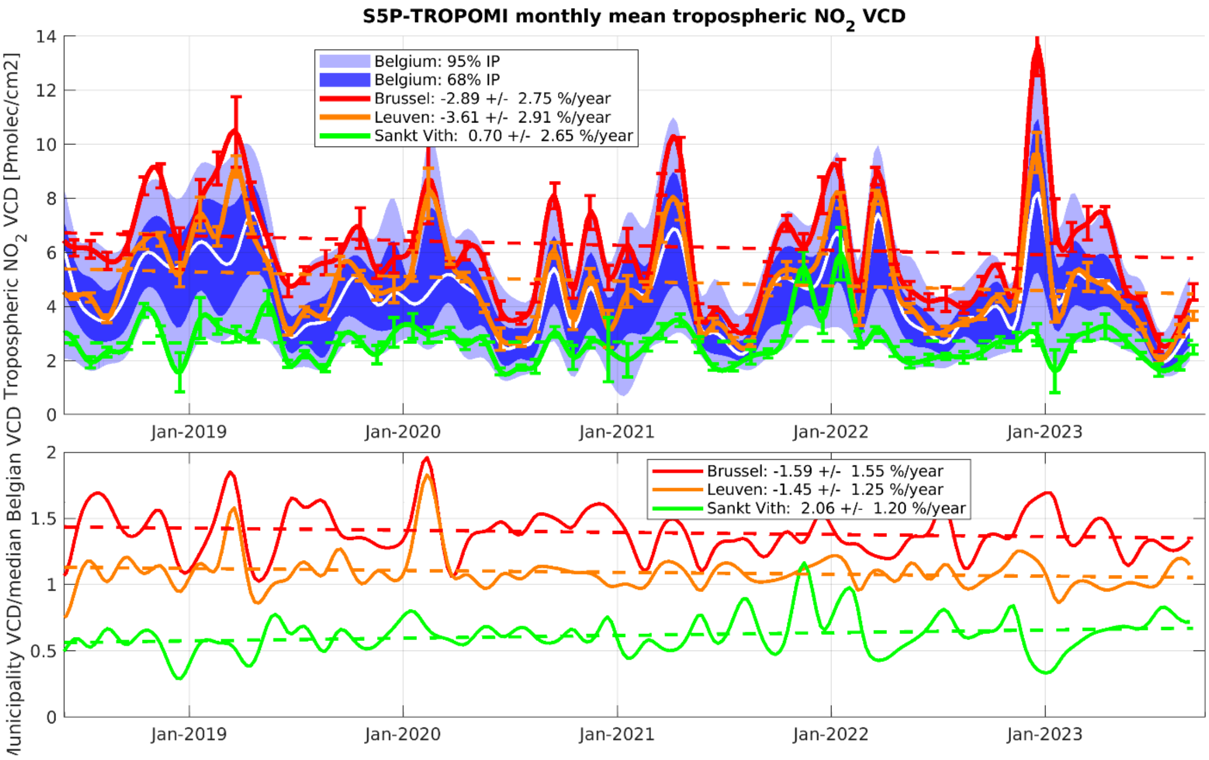

Figure 9: Upper panel: Time evolution of the monthly mean tropospheric NO_2 VCD for the ensemble of 581 Belgian municipalities (blue) and for three communes representative for different urban/background conditions in Belgium. Error bars represent the variation in observed VCD within each month. Lower panel: Time evolution of the VCDs over these three communes relative to the median of the ensemble.

Because of the strong inter- and intra-annual variability of NO_2, due to meteorological variations mostly, accurate trend determination over a period of only 5 years is not trivial. In an attempt to remedy this, the bottom panel of Figure 9 shows the evolution of the VCD over these communes normalized by the Belgian median VCD. As is common nomenclature in atmospheric sciences, we refer to these normalized VCDs as "*anomalies*". Temporal variations that hold for the entire Belgian domain, such as the annual cycle and to some extent the impact of the CoViD-19 related measures in 2020 and 2021, cancel out in this normalization. The derived trends in VCD and in VCD anomalies are based on a robust linear regression on exactly 5 years of data (June 2018 - June 2023) to further minimize the impact of the (seasonal) natural variability. The uncertainties on the trend are based on the variance of the residuals w.r.t. the linear relation, which is the most conservative uncertainty estimate as both an uncertainty propagation and a bootstrapping approach yield smaller uncertainties.

Overall, we find a (borderline significant) decrease of the NO_2 columns of -2.4 ± 2.5 %/year over Belgium as a whole over the past 5 years. Both Brussels and Leuven show a marginally stronger decrease of -2.9 ± 2.8 %/year and -3.6 ± 2.9 %/year respectively, while Sankt Vith experiences no decrease (we find an insignificant positive trend of 0.7 ± 2.7 %/year). After normalization with the Belgian median VCD, Brussels and Leuven both show a relative decrease of about -1.5 %/year, while Sankt Vith shows a relative increase of about 2 %/year. The absolute decrease rates around -3 %/year that we find for most Belgian cities are fully in line with previous estimates of -2 to -4 %/year for Western urban regions using AURA-OMI data from 2005 to 2019 (Goldberg et al., 2021b). Whether this implies that the LEZ have not (yet) had a substantial impact on the NO_2 VCDs over their area, or that their impact is more widespread, can not directly be answered here.

6.4 WP2(b): Comparison to near-surface data (measured and RIO model)

A major challenge in the use of satellite data for AQ monitoring is the relation between the observed tropospheric vertical column density (VCD), i.e. the integrated concentration from the surface up to the tropopause at an altitude of 8-10 km and the near-surface concentrations (NSC) that actually determine the health impacts. In particular the amount of vertical mixing in the planetary boundary layer (PBL), wind-driven advection (at various altitudes), and the presence of NOx sources in the free troposphere (e.g., lightning) can complicate this relation. A large body of literature explores potential methods to relate the VCD to the NSC, most often including meteorological information on PBL height and winds (e.g., Chan et al., 2021). On the other hand, surprisingly strong correlations have been found between VCD's and NSCs for long-term average NO_2 levels (e.g., Cersosimo et al., 2020). Below, we investigate the correlation between our high-resolution but temporally averaged satellite VCD maps and the NSCs measured/estimated in (1) the point-like in-situ measurements and (2) the 4-km resolution RIO model described in 6.1.

To make the data sets comparable, only near-surface data near the S5P overpass time were used, and for the comparison to RIO, the satellite data maps were first averaged to the 4-km resolution and projected on the Belgian Lambert 1972 grid of the RIO model. A detailed quantitative assessment is provided in Figure 10. The figure is based on 2021 annual average data but is representative for all years hitherto covered by S5P.

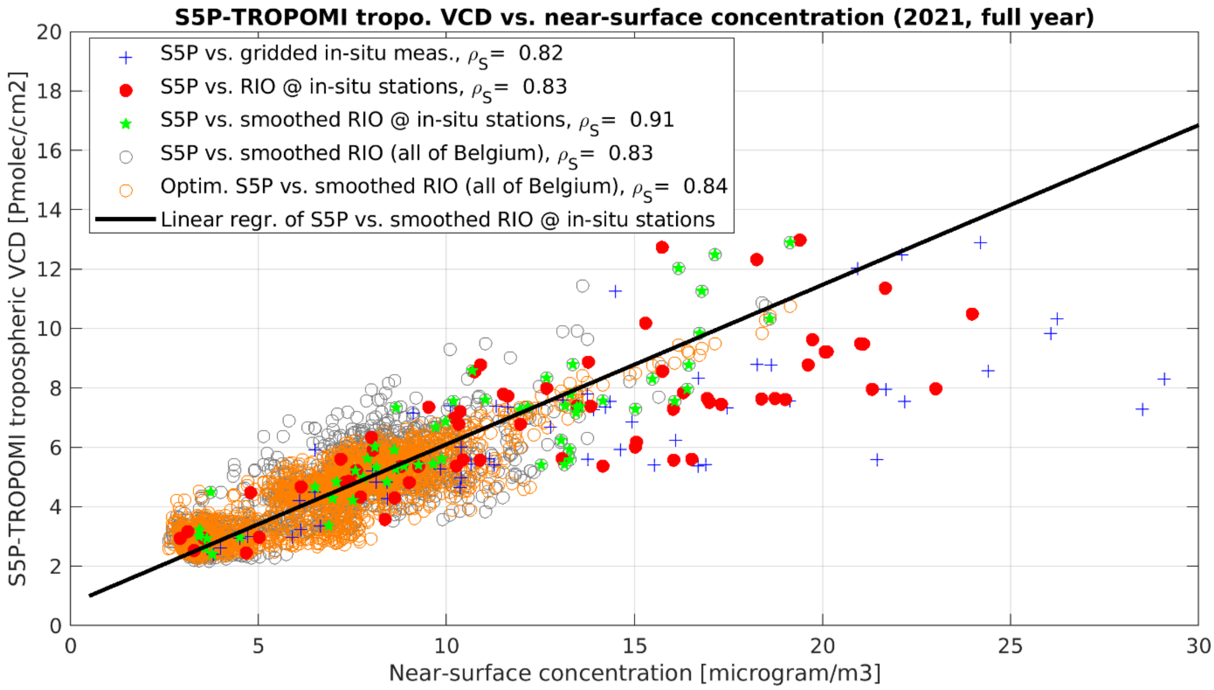

Figure 10: Comparison between the 2021 annual average S5P-TROPOMI tropospheric VCDs and the corresponding near-surface concentrations from either the in-situ measurements or the (smoothed) RIO model, for the entire Belgian territory at a 4-,km spatial resolution and taking only data at the S5P overpass time (near noon). See text for a detailed description of the different markers.

Several types of comparisons are performed in this analysis:

- Blue crosses: S5P-TROPOMI at 4-km resolution versus the in-situ measurements gridded at the (RIO) 4-km resolution grid. In particular in several of the major cities, where multiple in-situ instruments are located within a 4-km area, this gridding addresses some of the representativeness issues of e.g., instruments positioned close to major roads or intersections. The Spearman rank correlation coefficient (ρ_S = 0.82) indicates a good correlation, but a non-linear behaviour at high NSC is clear.
- Red disks: S5P-TROPOMI at 4-km resolution versus the RIO model grid cells covering in-situ instruments. These are the locations where the RIO model is (1) directly constrained by actual measurements and (2) helping to address representativeness issues through its underlying use of the CORINE land-cover data set. This is confirmed by the tighter relation at high NSC when compared to the direct comparison (the blue crosses), albeit with an only marginally better ρ_S = 0.83.
- Green asterisks: Similar to the red disks discussed above, but using a slightly spatially smoothed version of the RIO model, by using a 2D Gaussian filter with σ = 4 km. Indeed, as can be observed in Figure 1, the RIO model at 4-km resolution displays large pixel-to-pixel differences not observed in the S5P data. This may be due to some level of undersampling in the RIO output w.r.t. the true resolution of some underlying model data, and/or an effectively worse spatial resolution in the S5P-TROPOMI maps (either due to the instrumental limitations or due to atmospheric smoothing inherent to a long-term average tropospheric VCD). This slight spatial smoothing of the RIO model data improves the agreement considerably, resulting in a ρ_S = 0.91 and a clear, tight, linear relation.

- Grey circles: similar to the green asterisks discussed above: S5P-TROPOMI at 4-km resolution versus the RIO model grid cells, but now for the entire Belgian domain, i.e., not solely for those grid cells where in-situ measurements are located. For the majority of the grid cells, RIO relies on an interpolation method (combining land-cover data and Ordinary Kriging). As the S5P-TROPOMI data do contain multiple measurements per grid cell (of the order of 100), the weaker correlation (ρ_S = 0.83 versus the ρ_S = 0.91 obtained above) suggests that the satellite observes features that are not well captured by the RIO model interpolation scheme.
- Orange circles: similar to the grey circles discussed above: S5P-TROPOMI at 4-km resolution versus all RIO model grid cells, but now using an optimised S5P-TROPOMI map, where the optimisation consists of an adjustment per grid cell which is based on a Kriging interpolation of the residuals in a linear regression (black solid line, a.k.a. Kriging Regression) between S5P-TROPOMI and smoothed RIO at the in-situ instrument locations (i.e., the green asterisks). This adjustment represents the local nature of the relation between VCD and NSC not captured by a simple linear regression. While this optimisation only marginally improves the overall rank correlation, it does clearly reduce the scatter, in particular at higher NSC. Presumably this is due to most of the high NSCs occurring close to measuring stations, where the adjustment factors are well constrained.

In general terms, as in the Italian study by Cersosimo et al. (2020), we find large correlations between S5P-TROPOMI and in-situ measurements directly, and even more so between S5P-TROPOMI and (a slightly smoothed) RIO at the locations of in-situ instruments, i.e. where RIO was directly constrained by measurements and not relying on (much) interpolation. While Figure 10 is based on annual averages, similar analysis on seasonal data (not shown) reveals similar agreement. On monthly scales, especially in winter when cloud cover complicates the satellite measurements, the scatter in the comparisons increases substantially.

6.5 WP2(c): A pragmatic conversion from VCD to NSC

In view of the strong correlations between long-term average VCDs and NSCs described above, a pragmatic conversion from the former to the latter using the in-situ measurements and a linear model can be expected to yield valuable results, as for instance already demonstrated for health-impact assessments in the United States by Goldberg etal. (2021, and reference therein). Below, this conversion approach is adopted, albeit with two improvements:

i. Instead of using the in-situ measurements directly, we use the smoothed RIO model at the locations of the in-situ instruments, as this addresses to a large extent the representativeness and horizontal resolution differences between satellite and in-situ. In essence, it is the high-spatial resolution CORINE land-cover data set supporting RIO that facilitates this resolution matching.
ii. We supplement the linear regression with the local adjustment factors described earlier, i.e., a Kriging interpolation of the residuals w.r.t. the linear regression, which is aimed to capture some of the local variations in the VCD-NSC relation. This combination is known as Regression Kriging (Hengl, 2007, and references therein).

This synergistic use of both the satellite and in-situ data exploits the complementarity in both types of data, and can be expected to be virtually insensitive to the reported low bias in S5P-TROPOMI tropospheric VCDs.

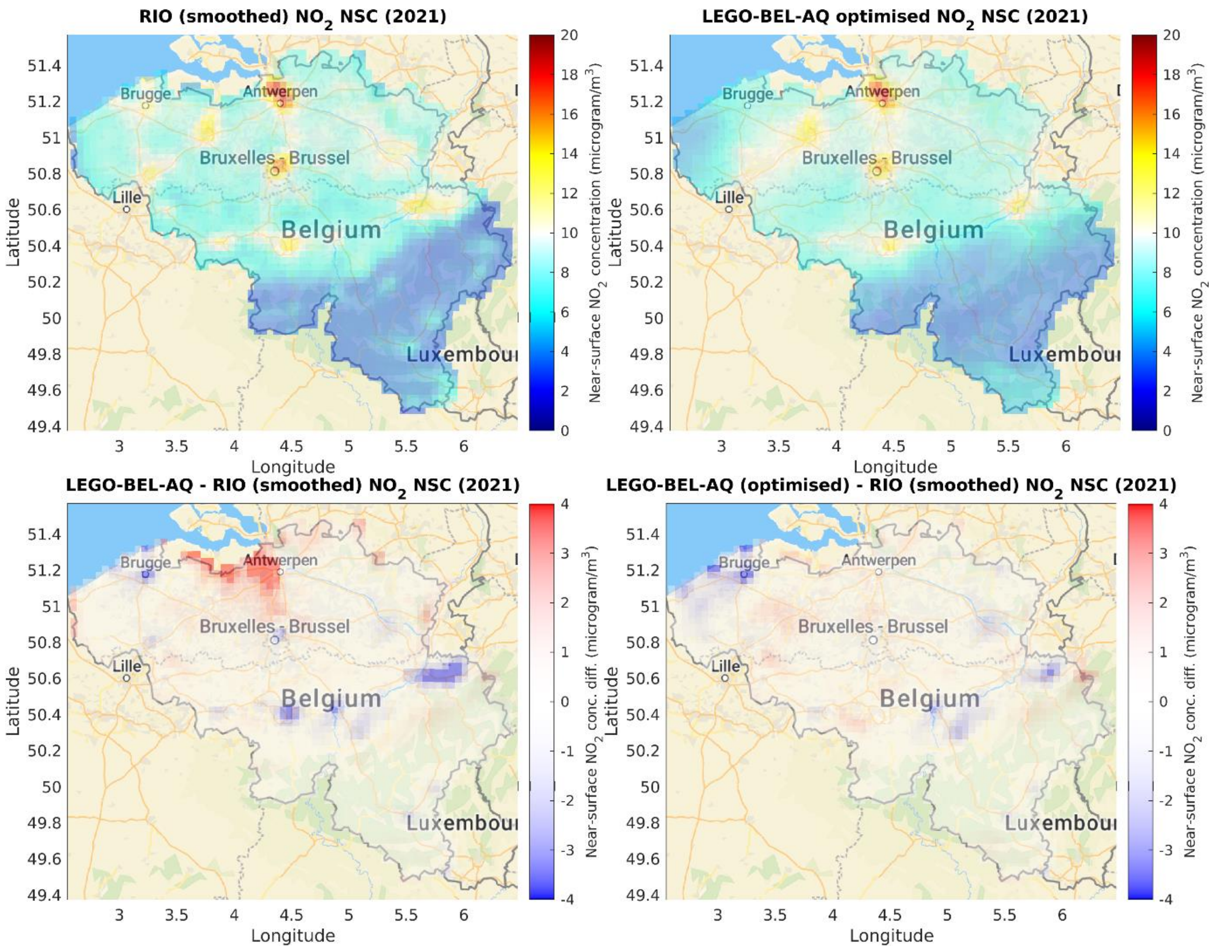

Figure 11: Comparison between the smoothed (σ = 4km) RIO near-surface concentrations and those derived from a synergistic use of S5P-TROPOMI tropospheric VCDs and RIO grid cells covering in-situ measuring instruments. The top row contains the NSC maps: smoothed RIO in the left-hand panel and optimised synergistic product in the right-hand panel. The color scale is centered (white) on the WHO annual exposure guideline (10 microgram/m^3). The bottom row contains the absolute differences for both the pure linear regression (left-hand panel), which preserves the spatial structure of the satellite VCDs, and for the optimised data set (right-hand panel).

Figure 11 visualises the results of this conversion for 2021. The agreement, in particular between RIO and the optimised S5P map (top row), is clear but should not be surprising for the main hot spots as these are well-sampled by in-situ instruments that are used here to tie the satellite data set to the ground "truth". Differences (visualized in the bottom panels), however, do exist. It is meaningful to make a distinction between (1) the raw, linear conversion from VCD to NSC, and (2) the optimised approach (with Kriging interpolation of the residuals). The former preserves the spatial structures seen in the VCD as a single linear conversion is used for the entire domain. The latter removes the spatial structure of the residuals in the linear regression. The bottom left panel of this figure shows that a pure linear conversion leads to strong positive differences over Antwerp (especially to the West), and negative differences over the major city centres, and over smaller cities such as Brugge, Hasselt, Charleroi, Namur, and to the East of Liège. When including the optimisation scheme (bottom right in Figure 11), differences at Antwerp, Charleroi and Brussels disappear, as expected because these cities are well sampled with in-situ instrumentation (yielding residuals in the linear regression that are used in the optimisation), but that does not necessarily imply biases in the satellite data or shortcomings in the conversion (e.g., due to pollution being present only above chimney height), but may also be related to representativeness issues or biases in the in-situ instrumentation. Also, some differences over smaller cities and more rural areas remain after optimisation, in particular for Brugge and Eupen. These locations may benefit from the installation of an in-situ instrument.

6.5.1 Uncertainty quantification

Various approaches are possible to determine an uncertainty on these satellite+in-situ+model based near-surface concentrations. Different contributors to the total uncertainty are - in presumed descending order of importance - (1) the measurement uncertainty on the satellite VCDs, (2) the natural variability in the relation between VCD and NSC, (3) the measurement uncertainty on the in-situ measurements, and (4) the uncertainty on the RIO model as translator between the point-like in-situ measurements and the 4x4km^2 resolution NSC. A propagation of the satellite measurement uncertainty though the process of temporal aggregation, spatial oversampling, and conversion is unlikely to give reliable results as the differentiation between random and systematic components (and the spatio-temporal correlation lengths involved) is not detailed enough (neither ex-ante or from validation). A more pragmatic and conservative estimate can be obtained from the scatter between satellite-derived, in-situ, and RIO NSCs, as shown in Figure 10. Table II lists mean difference (bias) and standard deviation of several meaningful comparisons. Robust similar estimators (median and half of the 68% interpercentile) yield roughly the same numbers. Both relative (in %) and absolute numbers (in microgram/m^3) are provided, but perusal of the scatter graphs suggests the relative numbers to be most meaningful as the deviations clearly depend on the NSC level. The improvement in scatter after optimisation is only marginal due to the limited number of pixels receiving a substantial adjustment, e.g., only 100 out of almost 4000 pixels are changed by more than 2 microgram/m^3.

Table II: Statistical properties of the differences between satellite-derived, in-situ, and RIO NSCs, presented here as indicators of the uncertainty on the NSCs.

Comparison	Mean difference	Standard deviation
LEGO vs. smoothed RIO @ stations	1.6% (0.0 µg/m^3)	19% (2.0 µg/m^3)
LEGO vs. smoothed RIO (full domain)	7.2% (0.3 µg/m^3)	21% (1.4 µg/m^3)
LEGO optim. vs. smoothed RIO (full domain)	4.0% (0.1 µg/m^3)	20% (1.2 µg/m^3)

As these bias (mean difference) and dispersion numbers also include the uncertainty on the in-situ measurements and the differences due to differing resolution and representativeness, the actual uncertainty on the satellite-derived NSCs will be lower. Attributing half of the dispersion to the satellite measurements and the shortcomings inherent to a linear conversion, we estimate the random and systematic uncertainty on the annual average LEGO VCDs to be about 14% and 5% respectively.

6.5.2 Temporal agreement

As one of the main drivers of this work concerns the temporal evolution of the pollutant concentrations, the temporal agreement between the satellite and near-surface monitoring data needs to be verified. In Figure 12, the time series of monthly means over the entire Belgian domain are compared.

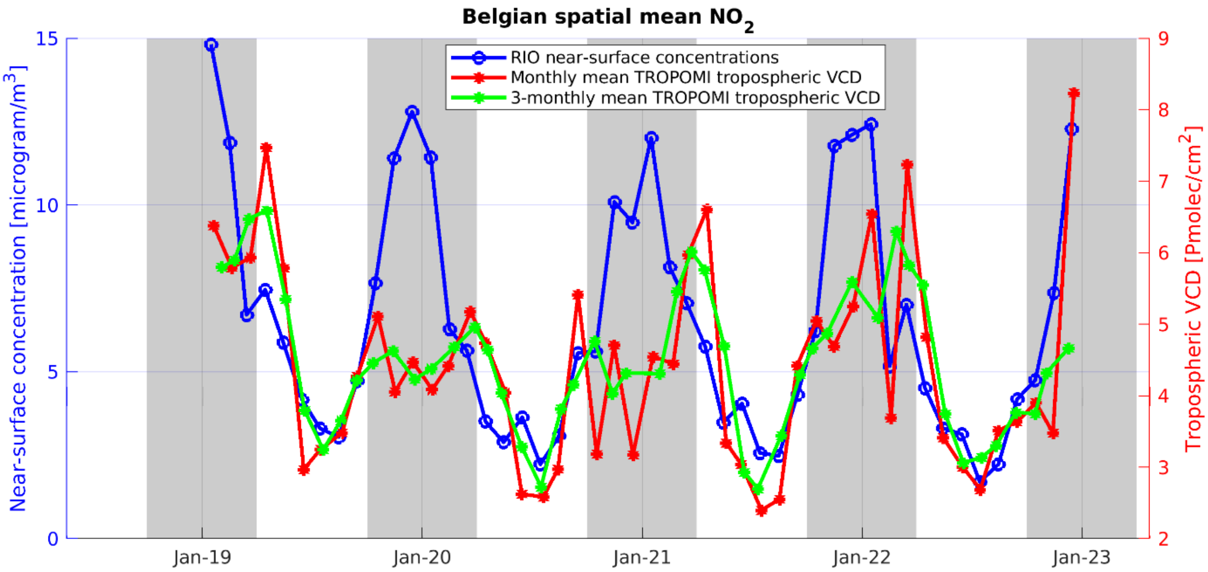

Figure 12: Comparison between the time series of monthly means: tropospheric VCDs from S5P-TROPOMI (right-hand scale) versus near-surface concentrations from the RIO model (left-hand scale) over the entire Belgian domain. The agreement is poor in the period from October to March (indicated in grey).

While both data sets are reasonably consistent over the summer periods (April to September), large differences occur in winter. The overall temporal correlation is of the order of $r \approx 0.6$, while for April to September it is $r \approx 0.8$. A regression analysis (not shown) confirms the poor agreement when NSCs averaged over Belgium exceed 7-8 µg/m^3, i.e., in winter. Large uncertainties on the satellite monthly mean VCDs are to be expected in winter due to cloud cover, but the systematic and strong underestimation of the seasonal amplitude is striking. It remains to be investigated whether this can be fully explained by sampling issues in the satellite data. In view of these results, trend analysis on the satellite VCDs should probably be limited to spring and summer-time data.

The VCD-to-NSC conversion method using the near-surface data to calibrate the VCD-NSC relation on an annual scale can be expected to absorb a large part of these biases, but the non-random year-by-year evolution of the regression coefficients presented in Table III indicates also different long-term temporal behaviour.

Table III: Robust regression coefficients between annual mean RIO near-surface concentrations and S5P-TROPOMI tropospheric VCDs, using only grid cells covering in-situ measuring instruments (i.e., corresponding to the regression line in Figure 10).

	2019	2020	2021	2022
Slope	0.36	0.43	0.46	0.51
Intercept	1.77	1.02	0.80	0.96

This different long-term behaviour is illustrated in Figure 13, which visualizes the ratio (in either NSC or VCD) of 1:30pm summertime NO_2 for RIO and S5P. Only April through September are used here to avoid the large sampling biases related to cloud cover (see Figure 12) in winter. While reductions up to 50% are seen in the RIO data over the center of Belgium, much more modest (typically 10-20%) and dispersed reductions are observed in the S5P data.

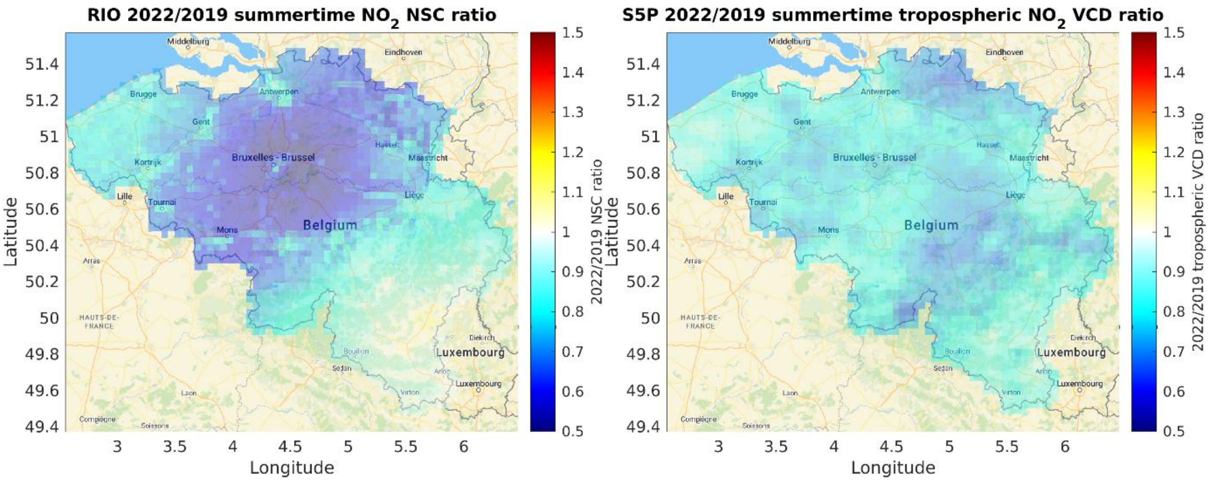

Figure 13: Comparison between the RIO (left-hand panel, NSCs) and S5P-TROPOMI (right-hand panel, tropospheric VCDs) 1:30pm summertime (April to September) ratio between 2022 and 2019 NO_2 levels.

The cause of this discrepancy is at the moment not understood. Some considerations to be made are:

- From validation with ground-based remote sensing, the well-known clear-sky bias in the satellite data appears to be multiplicative, which implies it should cancel out in a 2022/2019 ratio (if it is stable over time).
- If there is some additive bias in the satellite data, or if the satellite VCDs contain an additional NO_2 component well above the surface (e.g., in the free troposphere), this would appear in the intercept of the regression between VCD and NSC. The intercept is indeed not zero (see Table III), but subtracting the intercept for 2022 and 2019 before creating the ratio map only exacerbates the discrepancy.
- While sampling biases due to cloud cover are unlikely to be large during summer months, the winter-time comparisons to the in-situ NSCs do suggest that the amplitude of cloud-related biases may be large, and it is now well known that cloud cover over Western Europe is showing a clear negative trend (less and less clouds) due to climate change. This needs to be explored further.
- Chinese research revealed a similar discrepancy between satellite-derived and in-situ NO_2 trends over several Chinese megacities. In that work, an aerosol optical depth (an important influence quantity in the satellite NO_2 retrieval) decreasing over time was found to artificially reduce the satellite derived NO_2 trends (Yuhang Zhang, Peking University, private communication). This needs to be explored for the Belgian case as well.
- The reductions seen in the RIO map are significantly larger than the average trend observed in the past decade(s). The trend between land-cover type and NO_2 anomaly that supports the RIO model may be outdated (it is based on 2014-2017 data). This needs to be explored further.

6.5.3 The synergistic data at municipality level

The synergistic data set, i.e., the satellite VCD data converted to near-surface concentrations by Kriging regression against the RIO data (where they cover in-situ measuring sites), ties the spatial information in the spatially contiguous satellite data to the near-surface concentration “truth” (as in unaffected by (1) temporal sampling issues and (2) assumptions on the vertical distribution). As the regression is done on a per annum basis, annual maps (at overpass time) can be created at the municipality level for each of the years hitherto covered by the S5P-TROPOMI mission, and these will follow the long-term trend as represented by the RIO model. The result for the 1st and last complete year are shown in Figure 14.

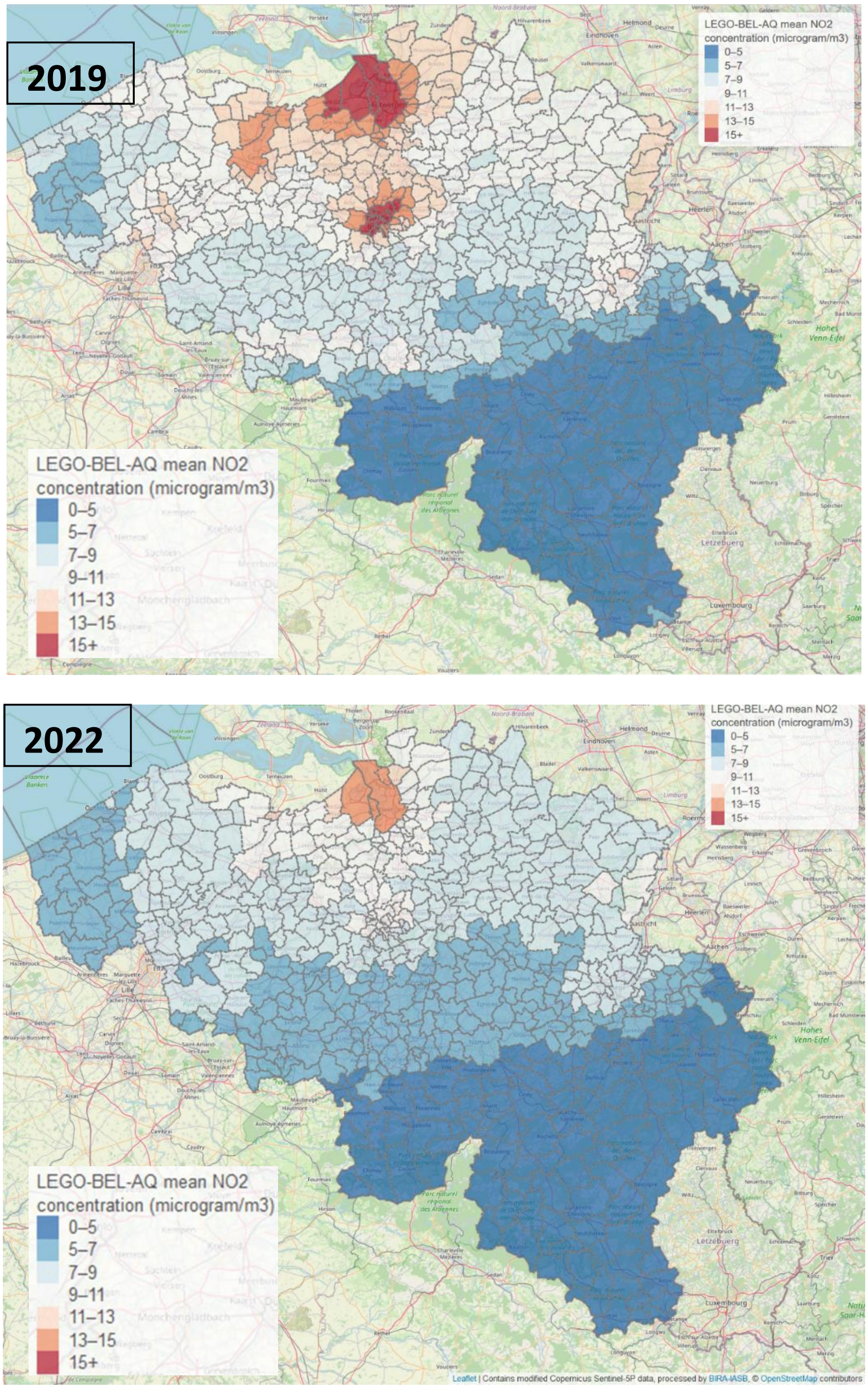

Figure 14: Maps of annual mean early afternoon NO_2 concentrations, averaged per municipality, for 2019 (top panel) and 2022 (bottom panel), as calculated with a synergistic use of both S5P TROPOMI NO2 tropospheric vertical column densities and RIO near-surface concentration data. The color scale is chosen to reflect compliance with the WHO annual exposure limit guideline (10 µg/m³), although that refers to the full daily exposure, not just the early afternoon situation presented here.

An additional step could be to use the diurnal information in the in-situ measurements to convert these early afternoon exposure maps into full 24-hour yearly exposure maps, but this was outside the scope of the current project and will be left as a future development.

6.6 WP3: Preparing for the geostationary sounders: an oblique viewing geometry

As introduced in Section 4, the constellation of satellite sounders dedicated to AQ monitoring that is being put in place consists of both low-earth-orbit (LEO) and geostationary (GEO) sounders. For Europe, the geostationary mission is Sentinel-4 on the Meteosat Third Generation-Sounder (MTG-S) platform. The first of two such instruments (S4-A) is to be launched in the course of 2025. The unique capability of a geostationary platform w.r.t. LEO is the measurement of the diurnal cycle: Sentinel-4 will map (a large part of) Europe almost every hour (sunlight permitting). This is of particular interest when measuring species with short (photochemical) lifetimes, such as NO2, showing a complicated diurnal behaviour driven by time-varying emissions (e.g., morning and evening traffic rush hours) and meteo-dependent losses/sinks (advection and photolysis). On the other hand, as a geostationary platform is only possible above the equator, Sentinel-4 will observe our latitudes under a highly oblique viewing zenith angle (VZA, see also Figure 15).

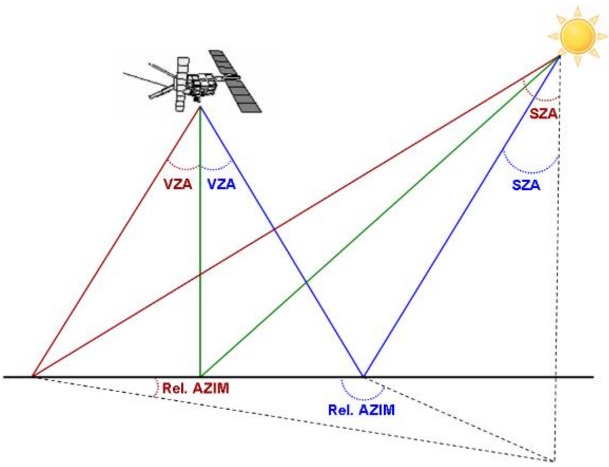

Figure 15: Schematic of the light-path travelled from the sun to the detector onboard a satellite, and the angles that characterize this light path: the Solar Zenith Angle (SZA), the Viewing Zenith Angle (VZA) and the relative azimuth angle. Reproduced from Vandenbussche et al., GEOmon Technical Notes, 2011.

6.6.1 Identified complications of the oblique viewing angle

This specific viewing geometry will potentially complicate the satellite retrievals through:

- Complex obscuration effects, by strong orographic features and by clouds. In particular observations in the shadow of clouds that are not directly in the line of sight (but in the path of the solar radiation) may be biased.
- Complex surface reflectance effects. Baseline reflectivity data sets do not differentiate for viewing angle, though the use of Bi-Directional Reflectance Functions (BRDF) is now becoming commonplace, e.g., the GE_LER approach for S5P-TROPOMI (Loyola et al., 2020).
- Horizontal smearing/smoothing effects. The satellite is sensitivity along the lines-of-sight depicted in Figure 15. Consequently, it captures information not just above the nominal ground pixel but from a complex atmospheric transect covering many km around the ground

pixel. If the distribution of the target species is very inhomogeneous (as is usually the case in polluted environments), this leads to errors not accounted for by the retrieval uncertainty budget (which typically assumes a horizontally homogeneous atmosphere).

The obscuration effects due to clouds have recently been looked into by Kylling et al. (2022), who found significant biases near cloud edges for both LEO and GEO sounders. From this follows a recommendation to not only remove cloud-contaminated pixels (usual practice), but also adjacent pixels. A follow-up paper, led by BIRA-IASB, looked into mitigation strategies at the retrieval stage (Yu, H., et al., “Impact of 3D cloud structures on the atmospheric trace gas products from UV–Vis sounders – Part 2: Impact on NO2 retrieval and mitigation strategies”, AMT, 2022).

The horizontal smoothing effect for a geostationary sounders had hitherto not yet been studied. Extensive such assessments have been performed for both LEO and ground-based remote sensing (see e.g., Lambert et al., 2012 and Verhoelst et al., 2015) and we built on those developments to do a first assessment for the upcoming GEO Sentinel-4.

6.6.2 Tailoring the OSSSMOSE observing system simulator and it’s inputs

The assessment of horizontal smoothing errors was performed with the Observing System of Systems Simulator for Multi-mission Synergies Exploration (OSSSMOSE) at BIRA-IASB. It was developed over the course of the past decade to study smoothing and sampling issues in LEO and ground-based remote sensing, and their intercomparison, with a particular focus on understanding the uncertainty budget of data comparisons as performed in the context of satellite data validation. The system is described in detail in both Verhoelst et al. (2015) and Lambert et al. (2018), with the former paper including an extensive application on total ozone column intercomparisons. The general lay-out is portrayed in Figure 16.

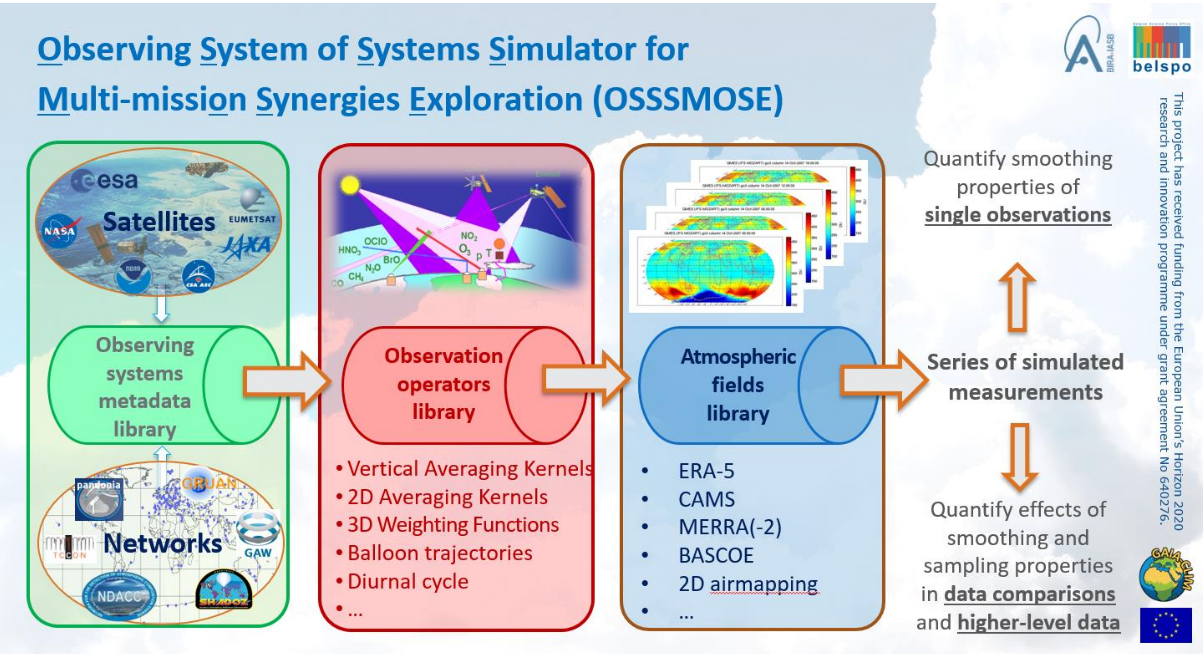

Figure 16: Schematic overview of the OSSSMOSE observing system simulator at BIRA-IASB, used here to simulate spatial smoothing effects in geostationary NO_2 observations over Belgium as planned with the upcoming Sentinel-4 missions.

The system contains/requires three components/inputs:

a. Parameters characterizing the observing system, such as orbital parameters for a satellite sounder.
b. Observation operators, which are a parametrization/functional description of the actual 2D/3D spatial distribution of the measurement sensitivity. These can be based either on pure geometrical arguments, or RT modelling, or Averaging Kernels if available.
c. A representation of the actual, inhomogeneous atmospheric field that will be observed. This can be either a model, reanalysis, or observational data set (satellite, aircraft,…) with as most important requirement that it's spatio-temporal resolution is high enough to resolve the field at the size of the observation operators.

For (a), we assumed Sentinel-4 to be located above (0°N,0°E) at an altitude of 36.000km above the Earth's surface. The viewing zenith angle is therefore approximately 58° at Belgian latitudes.

As observation operator (b) we chose to perform a horizontal projection along the line-of-sight of an estimated "effective" measurement height. This approach is common to that used to derive the actual measurement location of several ground-based remote sensing systems (e.g., for direct-sun measurements with Pandora instruments or FTIR spectrometers).

The effective height is defined as a weighted mean height, where the weights are determined by multiplying, for each altitude level, the satellite sensitivity (as estimated in a vertical Averaging Kernel, AK) with the actual NO_2 concentration.

The AK was taken from S5P-TROPOMI retrievals as we didn't have access yet to simulated Sentinel-4 AKs.

The vertical profile of the NO2 concentration was taken from the IMAGESv2 Chemical Transport Model (CTM) run at BIRA-IASB (Müller & Brasseur, 1995; Stavrakou et al., 2013). It uses ERA-interim meteorological fields and extensive emission inventories to calculate the evolution of 117 chemical compounds at high temporal and vertical resolution over the entire globe. The output for a grid cell covering the centre of Belgium (Uccle, near Brussels) is visualized in Figure 17 . This case represents the most complex situation, with significant anthropogenic NOx emissions during daytime. While a mid-latitude rural (unpolluted) site would be characterized by a sharp decrease and increase of NO_2 at sunrise and sunset respectively this photo-chemical loss during daytime is here completely filled in by the emissions. Only well into the free troposphere (around 10 km altitude) does the behavior resemble that of an unpolluted regime. In fact, quite striking and requiring further investigation is the apparent increase in NO_2 concentrations also above the PBL. This is at odds with the expectation that mostly vertical mixing within the PBL brings the emissions to higher altitudes.

The effective altitude for this case study came out to be about 5-6 km above the surface. Projected along the line-of-sight to Sentinel-4, this would imply a horizontal displacement of about 8 km to the South.

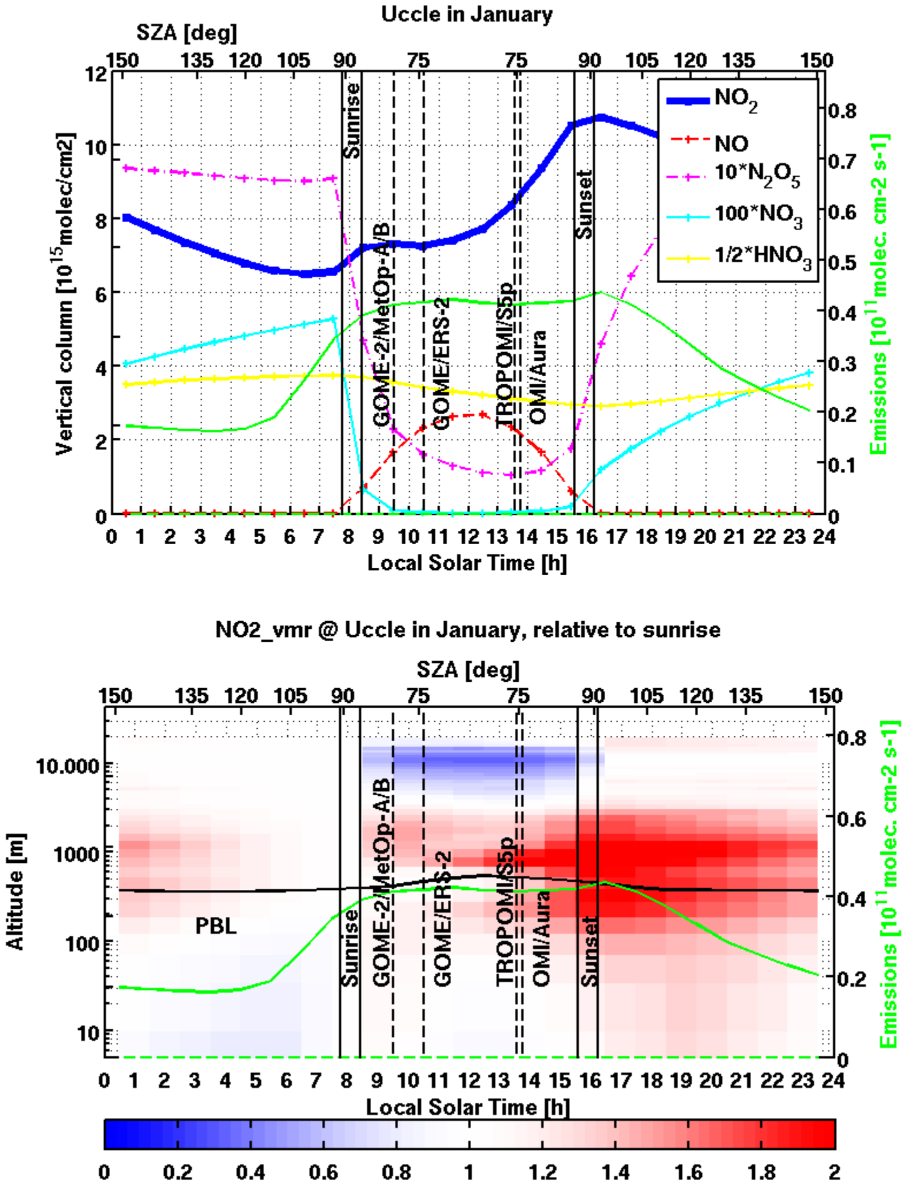

Figure 17: Upper panel: Diurnal cycle of the tropospheric column of NO_2 and related species at Uccle, near Brussels. Sunrise and sunset conditions are marked,and so are the overpass times of several key air quality satellite sounders. The bright green curve (right-hand axis) represents the combined NOx emissions, both anthropogenic and biogenic. Lower panel: Diurnal cycle of the concentration (volume mixing ratio, VMR) of NO2 as a function of altitude and normalized to the value at sunrise. The upper bound of the planetary boundary layer (PBL) is indicated (reproduced from Verhoelst et al., ESA Living Planet Proceedings, 2016).

As atmospheric field representing the "truth" at high horizontal resolution (c), we used a 1x1km^2 multi-annual mean S5P-TROPOMI NO_2 map produced in WP1 and WP2 (Sections 6.2 and 6.3 of this report). The difference can then be computed between this "truth" and the same field shifted 8 km to the South. While this is a simplification of the real situation in which the line-of-sight traverses the inhomogeneous atmosphere at an oblique angle, it is meant to serve as a qualitative indication of the potential effect and its amplitude. The result is visualized in Figure 18, in which the top panel represents a simulation with this 8 km displacement, while the bottom panel represents a more optimistic situation of only 2 km displacement.

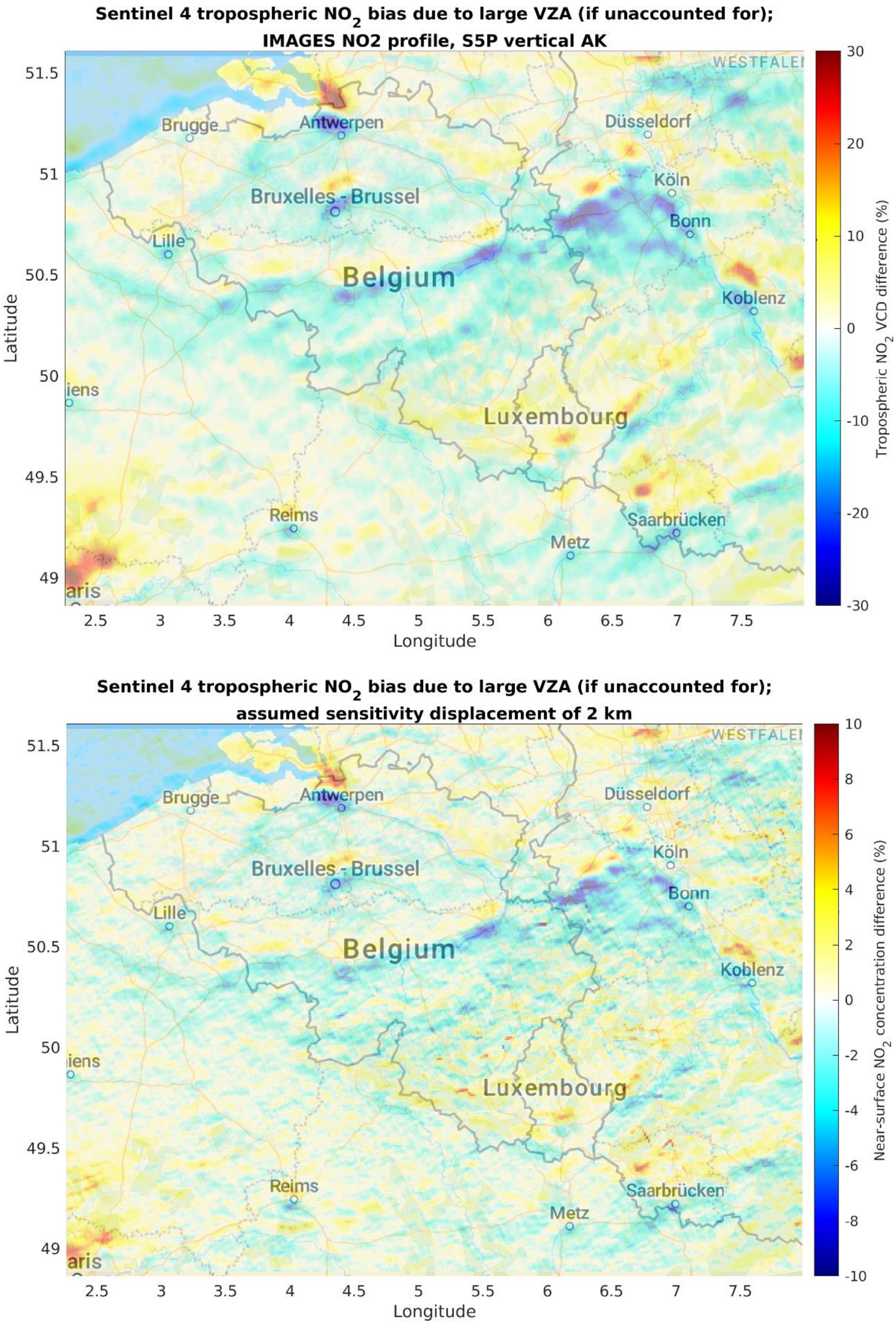

Figure 18: Simulated bias in Sentinel-4 observations of tropospheric NO_2 VCD over Belgium due to the highly oblique viewing angles of a geostationary platform (located above the equator. Top panel: using the 8 km displacement of sensitivity to the South derived from the assumed vertical NO_2 and satellite sensitivity profiles. Bottom panel: similar simulation but using a more optimistic 2 km displacement. Note the different color scales.

The main feature is a systematic underestimation, by up to 30% for 8 km displacement and 10% for 2 km displacement, at the Southern edge of regions with elevated NO_2 levels, such as the major cities, and an overestimation of similar magnitude at the Northern side. This amplitude is not negligible w.r.t. the typical measurement uncertainty, and it is a systematic effect as this viewing geometry is fixed in time.

In addition to this effect due to the oblique viewing angle, a similar effect exists for the line-of-sight between the sun and the back-scattering surface. As Sentinel-4 will observe during daylight hours, low-sun conditions will be sampled in the morning and evening, leading to displacements in sensitivity to the East and West respectively. Because of this varying azimuth angle (throughout the day and throughout the year), the resulting bias will impact the observed diurnal cycle. Such a simulation was outside the scope of this project, but it needs to be explored further to assess the accuracy of the diurnal cycle in Belgian NO_2 concentrations to be derived from the upcoming Sentinel-4 observations.

A final point to be made under this topic is the (absence of a) potential for spatial oversampling (as developed in WP1 and WP2 for LEO sounders) on the GEO data. As the geostationary platforms do not generate on-ground pixels with varying geo-location from orbit-to-orbit, an essential element to create superresolution, aggregation and oversampling of these GEO data does not provide any resolution benefit. This property was independently remarked and communicated to the community by Andreas Richter (IUP Bremen) at the TROPOMI 5-year anniversary event in Taormina (October 2022). His corresponding slide is copied below (Figure 19).

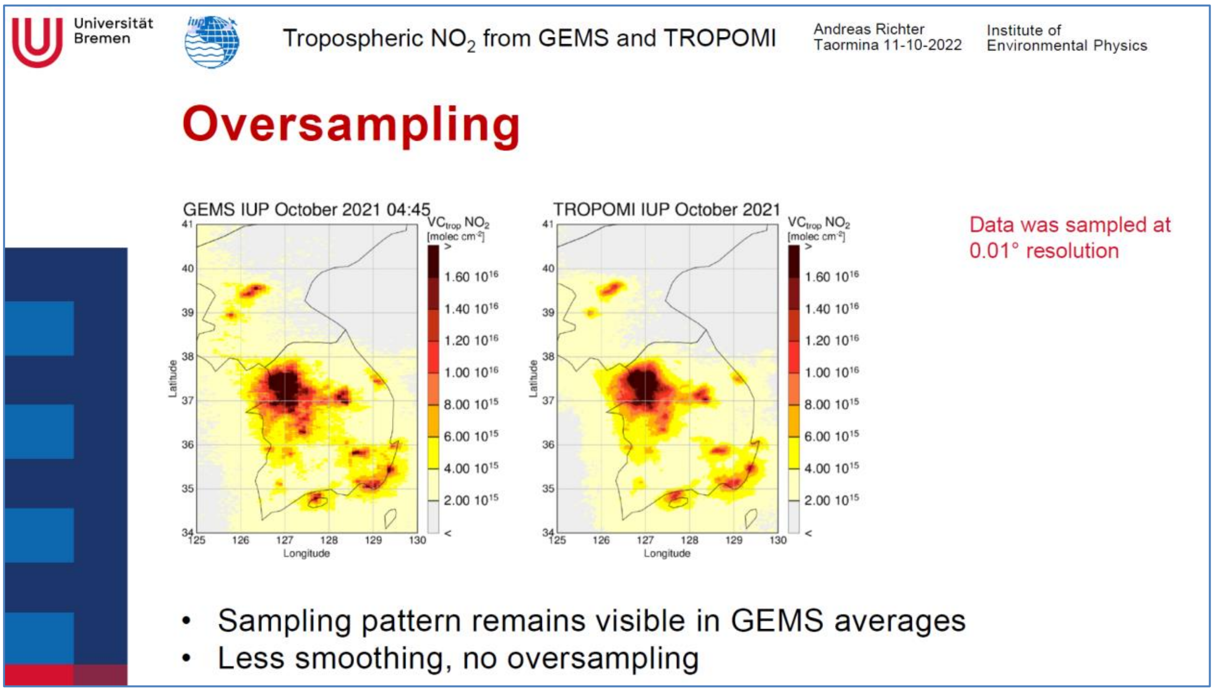

Figure 19: Analysis of the first GEMS geostationary AQ data confirms that the stable on-ground sampling pattern is in fact counterproductive to the oversampling potential. Slide courtesy of Andreas Richter (IUP Bremen), presented at the S5P-TROPOMI 5-year anniversary conference in Taormina, Italy, October 2022.

6.7 WP4: User consultation, outreach, and roadmap

To facilitate the user uptake of the AQ EO data, a separate work package was dedicated to user consultation and outreach. Thanks to the expertise of IRCEL in monitoring air quality and its relations with policy makers and key stakeholders, we confirmed or identified the reasons and main challenges for uptake of the air quality results provided by the EO community. Besides the technical challenges inherent to EO and (partially) targeted in WP1 through WP3, such as resolution, temporal sampling, and vertical sensitivity, other hurdles are (1) legislation on the official AQ reporting (e. g., at European level), which stipulates that the reporting needs to be based on in-situ instrumentation (except in the cleanest, most remote regions), and (2) general awareness of - and access to - the EO data.

Similar lessons were learned and communicated to us in a dedicated meeting by the Finnish Meteorological Institute, who have an excellent track record in EO data uptake (see Section 7.3).

While legislation was outside the scope of the project, improving awareness and data access were key objectives. Consequently, in the project, significant effort was put into dissemination and valorisation initiatives. These are elaborated in more detail in the following Section (7). In short, the following (types) of activities were carried out:

- Presentation of the project and its outcomes to targeted AQ stakeholders:
    - IRCEL coordination committee (including representatives of the regional administrations)
    - The ISSeP Groupe de Travail en Observation de la Terre (GTEO)
    - The European Commission Directorate-General Defence Industry and Space (EC DG DEFIS)
    - The project advisory board, including representatives from the EO community and the EC DG Environment, Clean Air Unit.
    - The general public (including His Majesty the King) at BIRA-IASB open doors
    - The European Air Quality Expert Group meeting (still to come, April 2024)
- Presentation of the project and its outcomes to the EO community at 9 international conferences and symposia (7 oral presentations, 2 posters)
- Access to project outcomes through a dedicated website (http://lego-bel-aq.aeronomie.be)
- A publication in a journal aimed at Belgian and Dutch AQ professionals (Tijdschrift Lucht)

Overall, from the stakeholder perspective, and in part thanks to the work performed in the LEGO-BEL-AQ project, it is now recognized that it is important to explore the possible complementarity of the EO data to the currently used monitoring and modelling systems. Indeed, the aim of user uptake should not be to go in competition with or to replace current methods, but rather to arrive at a synergistic use that ultimately completes and improves our view on Air Quality and its dependence on targeted policy.

As an illustration of the success of our outreach endeavours, we entered a collaboration with the Japanese National Institute for Environmental Studies (NIES) who are interested in the application of our system to the Japanese territory (see Section 7.5 for details).

Future activities identified as important to pursue, are:

a. further investigation into the discrepancy in trends between satellite and in-situ data,
b. integration of the diurnal information to be obtained with Sentinel-4 when launched,
c. geographical extension to Europe and beyond,
d. inclusion of additional AQ components, such as PM2.5 estimates from satellite aerosol optical depth data,
e. emission estimates, using for instance the flux divergence method, and comparison to bottom-up emission inventories,
f. further integration into the official monitoring systems.

Finally, the productive collaboration consolidated in this project between the EO community as represented by BIRA-IASB and the AQ monitoring stakeholders represented by IRCEL-CELINE, should continue to facilitate user uptake of EO data and an improved AQ monitoring system for Belgium (and beyond).

7 DISSEMINATION AND VALORISATION

As described in Section 6.7, user consultation and outreach were an important component of the project. Below, we detail the various avenues we elaborated. An article covering the material from WP1 and WP2, i.e., the work on high-resolution S5P-TROPOMI NO_2 data over Belgium, will be submitted to a peer-reviewed journal soon. At the time of writing of this report, the manuscript is receiving final polishing.

7.1 Website

A project website (http://lego-bel-aq.aeronomie.be) was developed and hosted at BIRA-IASB (Figure 20).

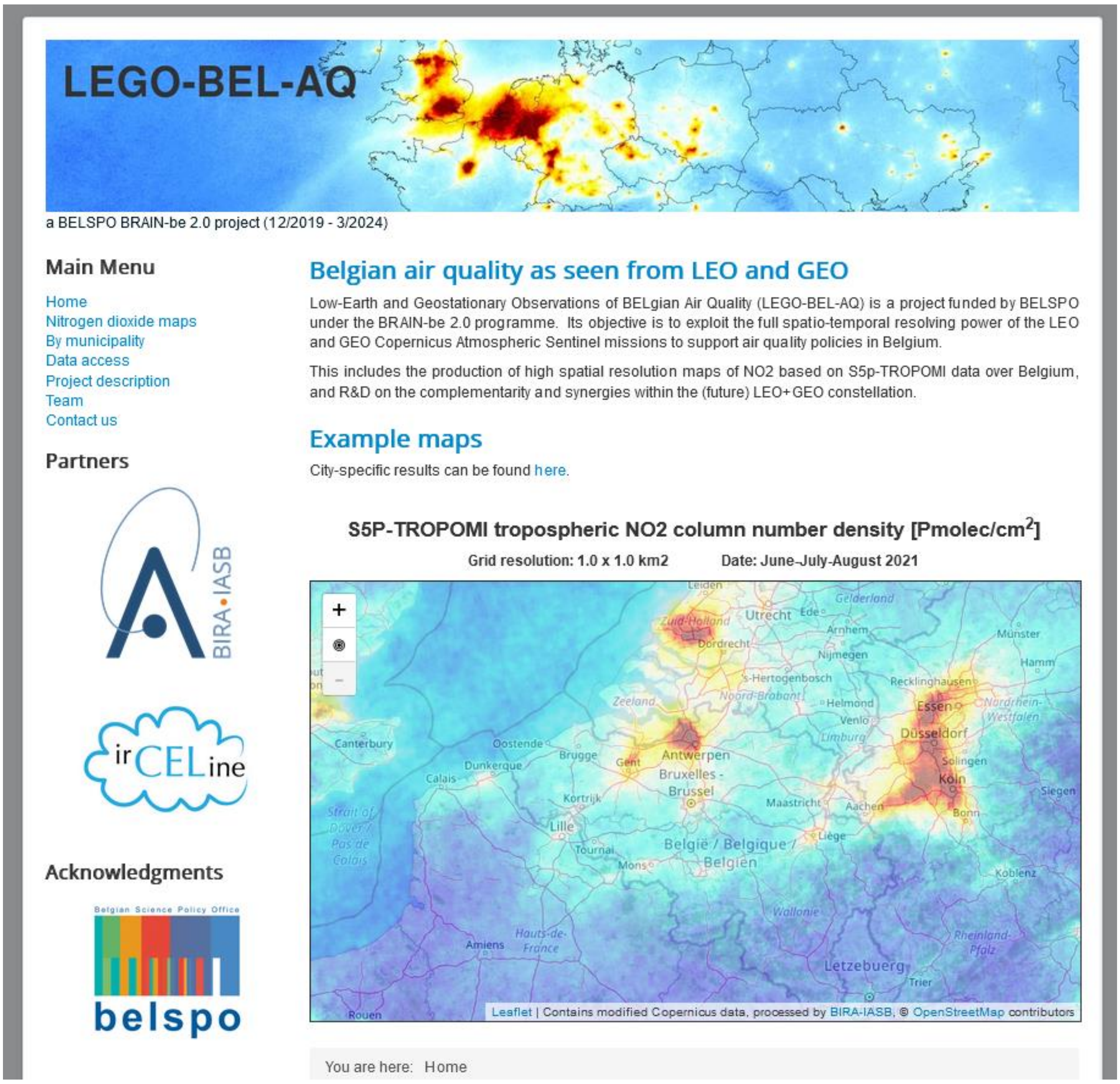

Figure 20: Landing page of the LEGO-BEL-AQ project website at http://lego-bel-aq.aeronomie.be

It contains a description of the project and the outcomes, both as static material and as interactive maps (which required substantial technical work, but a detailed description of that is not relevant for this report). It will be maintained also after project end. Data files are provided through an ftp server, with access details provided on the website.

7.2 Outreach opportunities

A significant set of outreach and stakeholder interaction opportunities was seized. Air quality stakeholders not directly involved in the space component were reached at the following events:

- Webinar of the Institut Scientifique de Service Public (*ISSeP*) Groupe de Travail en Observation de la Terre (GTEO) ; online project presentation ; 25 and 27 November 2020.
- Visit of a representative of the European Commission Directorate General Defence and Space Industry, Maria Berdahl, at BIRA-IASB, oral project presentation, 21 November 2022.
- Coordination commission IRCEL (representatives of ISSeP, AWAC, VMM), oral project presentation, 28 February 2024.
- BIRA-IASB Open Doors (general public), interactive session, September 24-25, 2022.
- Visit at BIRA-IASB by His Majesty the King (oral project presentation), 30 January 2024.

The project was also presented to the international EO community on several occasions:

- 2021 EUMETSAT Meteorological Satellite Conference, online oral presentation
- 2021 ESA ATMOS virtual conference (poster + short oral presentation)
- 2022 ESA Living Planet Symposium, Bonn, Germany, oral presentation
- 2022 EUMETSAT Meteorological Satellite Conference, Brussels, Belgium, oral presentation
- 2022 ESA S5P 5-year anniversary workshop, Sicily, poster
- 2023 EGU General Assembly, Vienna, Austria, oral presentation
- 2023 EUMETSAT Meteorological Satellite Conference, Malmö, Sweden, oral presentation
- 2023 AGU Fall Meeting, San Francisco, USA, oral presentation
- 2024 ACTRIS Science Conference, Rennes, France, oral presentation (planned)

Overall, interest in the LEGO-BEL-AQ work has been – and continues to be - very strong.

7.3 Lessons learned from the Finnish experience

From presentations at several EO-related conferences, we learned that the Finnish Meteorological Institute has been highly successful in facilitating uptake of satellite AQ data in both their national reporting and by private entities in their effort to reduce their emissions of pollutants (in particular in the CleanTech industry). To facilitate detailed discussions on their experiences and lessons learned, we organized an online meeting with FMI, BIRA-IASB, and IRCEL-CELINE. The meeting consisted of a presentation by Iolanda Ialongo and Henrik Virta on their strategy, achievements, and lessons learned with regard to user uptake, followed by a Q&A-type discussion. The cover and conclusions slides of the FMI presentation are copied in Figure 21.

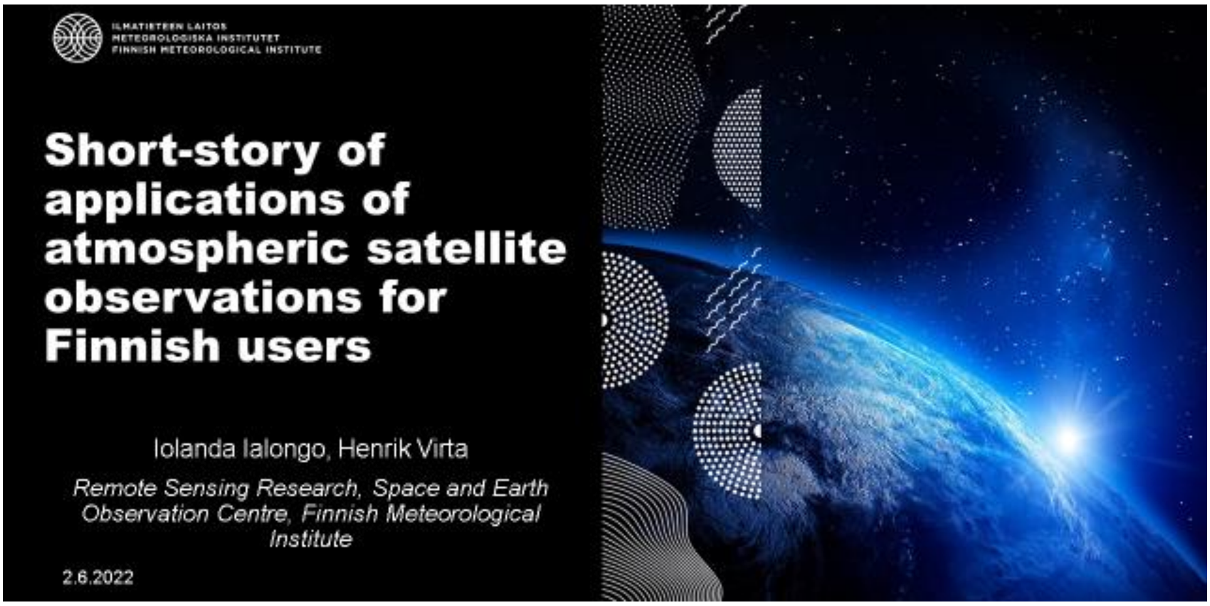

Figure 21: Cover and conclusion slide from a presentation by FMI colleagues in a dedicated knowledge-transfer meeting involving FMI, BIRA-IASB, and IRCEL-CELINE in February 2022.

7.4 Publication in “Tijdschrift Lucht”

A crucial observation is that many potential users, especially at the more local levels, are unaware of the developments in the space component and what they might bring to them. To act on this, we wrote a short overview article on satellite-based AQ monitoring aimed at (local) environment experts in the Netherlands and Belgium in the dedicated jounal “Tijdschrift Lucht”, published by the “network van milieuprofessionals” (https://www.vvm.info/home). The table of contents of issue 2, 2022, which includes a small cartoon created specifically for our contribution by the journal’s illustrator, is reproduced in Figure 22.

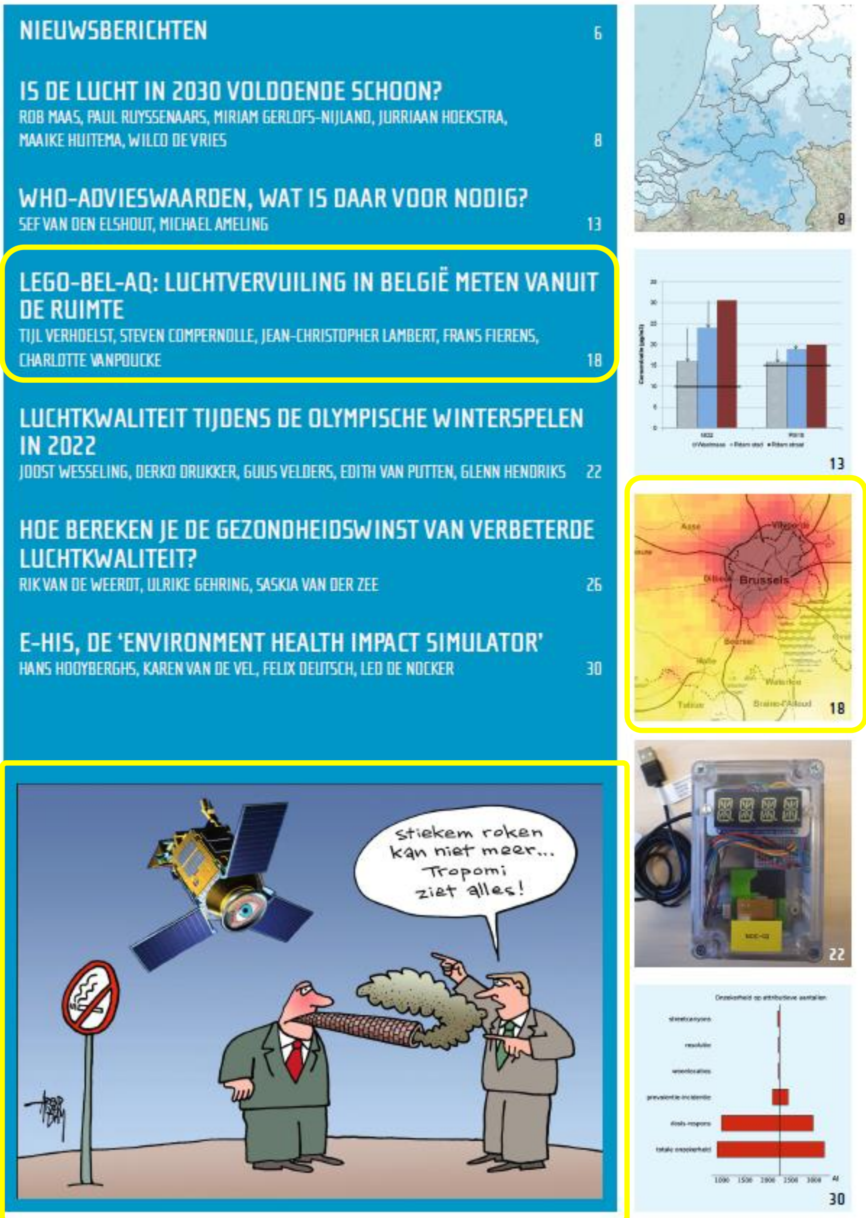

Figure 22: Table of contents of issue 2, 2022, of the Dutch journal "Tijdschrift Lucht", for which we wrote an overview/introductory article on (Belgian) AQ monitoring from Space.

7.5 Application to Japan

An encouraging outcome of our outreach activities was a request for collaboration by the head of the Atmosphere group at the Japanese National Institute for Environmental Studies (NIES), Hiroshi Tanimoto. He saw potential in an application of the LEGO-BEL-AQ toolset on Japan and its many highly populated and heavily polluted (mega-)cities. This work was initiated in early 2023 and first outcomes (e.g., Figure 23) are very promising. Several NIES representatives visited BIRA-IASB in February 2024 and attended the LEGO-BEL-AQ final meeting at BELSPO. This work will be continued, including the comparison to – and synergistic use with - the Japanese in-situ measurements. This 2nd case study is an illustration of the potential for geographical extension of the LEGO-BEL-AQ work.

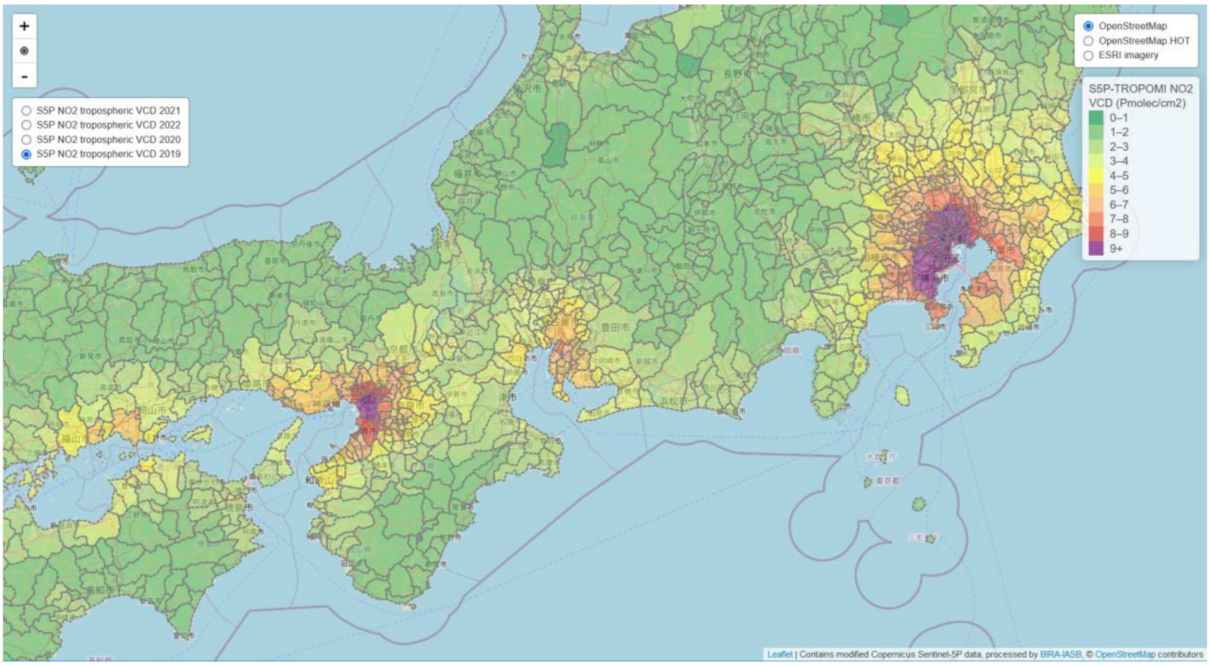

Figure 23: Application of the LEGO-BEL-AQ tools on S5P-TROPOMI data over Japan, including aggregation over the Japanese districts. The major urban agglomeration on the right is Tokyo, the smaller one on the left Osaka.

8 PUBLICATIONS

8.1 Peer-reviewed papers

A paper to be submitted for peer review and dedicated to the project outcomes from WP1 and WP2 is in a final phase of polishing:

- **Verhoelst, T.,** Compernolle, S., Lambert, J.-C., Fierens, F., Vanpoucke, C.: *Policy-relevant Air Quality information from satellite remote sensing: A case study on Belgium*, in prep., 2024

Besides this dedicated article, the team has contributed to several peer-reviewed papers of interest to the project:

- Petetin, H., Guevara, M., **Compernolle, S.,** Bowdalo, D., Bretonnière, P.-A., Enciso, S., Jorba, O., Lopez, F., Soret, A., and Pérez García-Pando, C.: *Potential of TROPOMI for understanding spatio-temporal variations in surface NO2 and their dependencies upon land use over the Iberian Peninsula,* EGUsphere [preprint], https://doi.org/10.5194/egusphere-2022-1056, 2022.
- Souri, A. H., Johnson, M. S., Wolfe, G. M., Crawford, J. H., Fried, A., Wisthaler, A., Brune, W. H., Blake, D. R., Weinheimer, A. J., **Verhoelst, T**., **Compernolle, S.,** Pinardi, G., Vigouroux, C., Langerock, B., et al..: *Characterization of Errors in Satellite-based HCHO / NO2 Tropospheric Column Ratios with Respect to Chemistry, Column to PBL Translation, Spatial Representation, and Retrieval Uncertainties*, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-410, 2022.
- Douros, J., Eskes, H., van Geffen, J., Boersma, K. F., **Compernolle, S**., Pinardi, G., Blechschmidt, A.-M., Peuch, V.-H., Colette, A., and Veefkind, P*.: Comparing Sentinel-5P TROPOMI NO2 column observations with the CAMS-regional air quality ensemble,* EGUsphere [preprint], https://doi.org/10.5194/egusphere-2022-365, 2022.
- van Geffen, J., Eskes, H., **Compernolle, S**., Pinardi, G., **Verhoelst, T., Lambert, J.-C**., Sneep, M., ter Linden, M., Ludewig, A., Boersma, K. F., and Veefkind, J. P.: Sentinel-5P *TROPOMI NO2 retrieval: impact of version v2.2 improvements and comparisons with OMI and ground-based data*, Atmos. Meas. Tech., 15, 2037–2060, https://doi.org/10.5194/amt-15-2037-2022, 2022.
- Levelt, P. F., Stein Zweers, D. C., Aben, I., Bauwens, M., Borsdorff, T., De Smedt, I., Eskes, H. J., Lerot, C., Loyola, D. G., Romahn, F., Stavrakou, T., Theys, N., Van Roozendael, M., Veefkind, J. P., and **Verhoelst, T**.: *Air quality impacts of COVID-19 lockdown measures detected from space using high spatial resolution observations of multiple trace gases from Sentinel-5P/TROPOMI*, Atmos. Chem. Phys., 22, 10319–10351, https://doi.org/10.5194/acp-22-10319-2022, 2022.
- **Verhoelst, T**., **Compernolle, S**., Pinardi, G., **Lambert, J.-C**., Eskes, H. J., Eichmann, K.-U., Fjæraa, A. M., Granville, J., Niemeijer, S., Cede, A., Tiefengraber, M., Hendrick, F., Pazmiño, A., Bais, A., Bazureau, A., Boersma, K. F., Bognar, K., Dehn, A., Donner, S., Elokhov, A., Gebetsberger, M., Goutail, F., Grutter de la Mora, M., Gruzdev, A., Gratsea, M., Hansen, G. H., Irie, H., Jepsen, N., Kanaya, Y., Karagkiozidis, D., Kivi, R., Kreher, K., Levelt, P. F., Liu, C., Müller, M., Navarro Comas, M., Piters, A. J. M., Pommereau, J.-P., Portafaix, T., Prados-Roman, C., Puentedura, O., Querel, R., Remmers, J., Richter, A., Rimmer, J., Rivera Cárdenas, C., Saavedra de Miguel, L., Sinyakov, V. P., Stremme, W., Strong, K., Van Roozendael, M., Veefkind, J. P., Wagner, T., Wittrock, F., Yela González, M., and Zehner, C., *Ground-based validation of the Copernicus Sentinel-5P TROPOMI NO2 measurements with the NDACC ZSL-*

DOAS, MAX-DOAS and Pandonia global networks, Atmos. Meas. Tech., 14, 481–510, https://doi.org/10.5194/amt-14-481-2021, 2021.

8.2 Other papers

- "*LEGO-BEL-AQ : luchtvervuiling in België meten vanuit de ruimte*", **T. Verhoelst, S. Compernolle, J.-C. Lambert, F. Fierens, C. Vanpoucke**, sollicited contribution, Tijdschrift Lucht, Issue 2, 2022.

8.3 Presentations at conferences and symposia

- **Verhoelst, T**., Compernolle, S., Lambert, J.-C., Fierens, F., Vanpoucke, C., *Synergistic Monitoring of Air Quality Policy from Space and In Situ: A Case Study on Belgium*, ACTRIS Science Conference 2024, 13-16 May 2024. (oral)

- **Verhoelst, T**., Compernolle, S., Lambert, J.-C., Fierens, F., Vanpoucke, C., *From LEO and GEO Satellite Remote Sensing to Policy-Relevant Air Quality Information: A Case Study on Belgium*, AGU23, 11-15 December 2023. (oral)

- **Verhoelst, T**., Compernolle, S., Lambert, J.-C., Fierens, F., Vanpoucke, C., *LEGO-BEL-AQ: a prototype satellite-based service for policy-relevant Air Quality information in Belgium*, EUMETSAT Meteorological Satellite Conference, Malmö, Sweden, 11-15 September 2023. (oral)

- **Verhoelst, T**., Compernolle, S., Lambert, J.-C., Fierens, F., Vanpoucke, C., *Monitoring Belgian air quality with LEO and GEO atmospheric composition data*, EGU General Assembly 2023, 23-28 April 2023. (oral)

- **Verhoelst, T**., Compernolle, S., Lambert, J.-C., Fierens, F., Vanpoucke, C.: *Exploiting the full spatio-temporal resolution of S5P NO2 data for Belgian air quality monitoring*, ESA S5P 5-years anniversary meeting, Taormina, Italy, 10-14 October 2022. (poster)

- **Verhoelst, T.**, Compernolle, S., Lambert, J.-C., Fierens, F., Vanpoucke, C.: *Tailoring LEO and GEO atmospheric composition EO data for Belgian air quality monitoring*, EUMETSAT Meteorological Satellite Conference, Brussels, 19-23 September 2022. (oral)

- **Verhoelst, T**., S. Compernolle, Lambert, J.-C., Fierens, F., Vanpoucke, C.: *Monitoring Belgian Air Quality from space through the synergistic use of the Sentinel constellation*, ESA Living Planet Symposium (LPS 2022), Bonn, Germany, 23-27 May 2022. (oral)

- **Verhoelst, T.,** Compernolle, S., Lambert, J.-C., Fierens, F., Vanpoucke, C.: *Tailoring Atmospheric Data from the Sentinel-4/5(p) Constellation in Support of Belgian Air Quality Policies*, ESA ATMOS conference, 22-26 November 2021, online event. (poster)
- **Verhoelst, T**., Compernolle, S., Lambert, J.-C., Fierens, F., Vanpoucke, C.: *Tailoring atmospheric data from the Sentinel-4/5(p) constellation in support of Belgian Air Quality policies*, EUMETSAT Meteorological Satellite Conference 2021, Virtual meeting, 20-24 September 2021. (oral)

8.4 Presentations at workshops and meetings

- **Verhoelst, T. & Compernolle, S.:** BIRA-IASB seminar: *Sentinel-5P NO2: from global validation to local application***,** 3 June 2022.
- **Verhoelst, T.** , Compernolle S., Lambert, J.-C., Fierens, F., Vanpoucke, C.: *LEGO-BEL-AQ: Low-Earth and Geostationary Observations of BELgian Air Quality*, ISSeP GTEO Air-Atmosphère, 25,27 November 2020, online

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- Alessandro Fassó (UniBergamo, Italy)
- Laura Judd (NASA Langley Research Center, USA)
- Vicente Franco and Christian Büns (European Commission Directorate-General Environment, Clean Air Unit, Brussels, Belgium)

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10 REFERENCES

Anenberg, S.C.; Mohegh, A.; Goldberg, D.L.; Kerr, G.H.; Brauer, M.; Burkart, K.; Hystad, P.; Larkin, A.;Wozniak, S.; Lamsal, L. Long-term trends in urban NO2 concentrations and associated paediatric asthma incidence: estimates from global datasets. The Lancet Planetary Health 2022, 6, e49–e58. https://doi.org/https://doi.org/10.1016/S2542-5196(21)00255-2.

Bassani, C.; Vichi, F.; Esposito, G.; Montagnoli, M.; Giusto, M.; Ianniello, A. Nitrogen dioxide reductions from satellite and surface observations during COVID-19 mitigation in Rome (Italy). Environmental Science and Pollution Research 2021 https://doi.org/10.1007/s11356-020-12141-9.

Bauwens, M., Compernolle, S., Stavrakou, T., Müller, J.-F., vanGent, J., Eskes, H., et al. (2020). Impact of coronavirus outbreak on NO_2 pollution assessed using TROPOMI and OMI observations. *Geophysical Research Letters*, 47, e2020GL087978. https://doi.org/10.1029/2020GL087978

Boersma, K.F.; Eskes, H.J.; Richter, A.; De Smedt, I.; Lorente, A.; Beirle, S.; van Geffen, J.H.G.M.; Zara, M.; Peters, E.; Van Roozendael, M.; et al. Improving algorithms and uncertainty estimates for satellite NO2 retrievals: results from the quality assurance for the essential climate variables (QA4ECV) project. Atmospheric Measurement Techniques 2018, 11, 6651–6678. https://doi.org/10.5194/amt-11-6651-2018.

Boersma, K.F.; Eskes, H.J.; Veefkind, J.P.; Brinksma, E.J.; van der A, R.J.; Sneep, M.; van den Oord, G.H.J.; Levelt, P.F.; Stammes, P.; Gleason, J.F.; et al. Near-real time retrieval of tropospheric NO2 from OMI. Atmos. Chem. Phys. 2007, 7, 2103–2118. https://doi.org/10.5194/acp-7-2103-2007.

Boersma, K.F.; Eskes, H.J.; Dirksen, R.J.; van der A, R.J.; Veefkind, J.P.; Stammes, P.; Huijnen, V.; Kleipool, Q.L.; Sneep, M.; Claas, J.; et al. An improved tropospheric NO2 column retrieval algorithm for the Ozone Monitoring Instrument. Atmos. Meas. Tech. 2011, 4, 1905–1928. https://doi.org/10.5194/amt-4-1905-2011.

Cersosimo, A.; Serio, C.; Masiello, G. TROPOMI NO2 Tropospheric Column Data: Regridding to 1 km Grid-Resolution and Assessment of their Consistency with In Situ Surface Observations. Remote Sensing 2020, 12. https://doi.org/10.3390/rs12142212.

Chan, K.L.; Khorsandi, E.; Liu, S.; Baier, F.; Valks, P. Estimation of Surface NO2 Concentrations over Germany from TROPOMI Satellite Observations Using a Machine Learning Method. Remote Sensing 2021, 13. https://doi.org/10.3390/rs13050969.

Clarisse, L.; Van Damme, M.; Clerbaux, C.; Coheur, P.F. Tracking down global NH3 point sources with wind-adjusted superresolution. Atmospheric Measurement Techniques 2019, 12, 5457–5473. https://doi.org/10.5194/amt-12-5457-2019.

Cooper, M.J.; Martin, R.V.; Hammer, M.S.; Levelt, P.F.; Veefkind, P.; Lamsal, L.N.; Krotkov, N.A.; Brook, J.R.; McLinden, C.A. Global fine-scale changes in ambient NO2 during COVID-19 lockdowns. Nature 2022, 601, 380–387.

Dominy, S.C.; Platten, I.M.; Raine, M.D. Grade and geological continuity in high-nugget effect gold–quartz reefs: implications for resource estimation and reporting. Applied Earth Science 2003, 112, 239–259, https://doi.org/10.1179/037174503225003116.

Douros, J.; Eskes, H.; van Geffen, J.; Boersma, K.F.; Compernolle, S.; Pinardi, G.; Blechschmidt, A.M.; Peuch, V.H.; Colette, A.; Veefkind, P. Comparing Sentinel-5P TROPOMI NO2 column observations with the CAMS-regional air quality ensemble. EGUsphere 2022, 2022, 1–40. https://doi.org/10.5194/egusphere-2022-365.

François-Bongarçon, D. Theory of sampling and geostatistics: an intimate link. Chemometrics and Intelligent Laboratory Systems 2004, 74, 143–148. Special Issue: 50 years of Pierre Gy's Theory of Sampling Proceedings: FirstWorld Conference on Sampling and Blending (WCSB1) Tutorials on sampling: Theory and Practice, https://doi.org/https://doi.org/10.1016/j.chemolab.2004.03.015

Fioletov, V.; McLinden, C.A.; Griffin, D.; Krotkov, N.; Liu, F.; Eskes, H. Quantifying urban, industrial, and background changes in NO2 during the COVID-19 lockdown period based on TROPOMI satellite observations. Atmos. Chem. Phys. 2022, 22, 4201–4236. https://doi.org/10.5194/acp-22-4201-2022.

Goldberg, D.L.; Anenberg, S.C.; Kerr, G.H.; Mohegh, A.; Lu, Z.; Streets, D.G. TROPOMI NO2 in the United States: A Detailed Look at the Annual Averages, Weekly Cycles, Effects of Temperature, and Correlation With Surface NO2 Concentrations. Earth's Future 2021, 9, e2020EF001665, https://doi.org/https://doi.org/10.1029/2020EF001665.

Goldberg, D.L.; Anenberg, S.C.; Lu, Z.; Streets, D.G.; Lamsal, L.N.; McDuffie, E.E.; Smith, S.J. Urban NOx emissions around the world declined faster than anticipated between 2005 and 2019. Environmental Research Letters 2021, 16, 115004. https://doi.org/10.1088/1748-9326/ac2c34.

Hengl, T.; Heuvelink, G.B.; Rossiter, D.G. About regression-kriging: From equations to case studies. Computers & Geosciences 2007, 33, 1301–1315. Spatial Analysis, https://doi.org/https://doi.org/10.1016/j.cageo.2007.05.001.

Ialongo, I.; Virta, H.; Eskes, H.; Hovila, J.; Douros, J. Comparison of TROPOMI/Sentinel-5 Precursor NO2 observations with ground-based measurements in Helsinki. Atmospheric Measurement Techniques 2020, 13, 205–218. https://doi.org/10.5194/amt-13 512-205-2020.

Janssen, S.; Dumont, G.; Fierens, F.; Mensink, C. Spatial interpolation of air pollution measurements using CORINE land cover data. Atmospheric Environment 2008, 42, 4884–4903. https://doi.org/https://doi.org/10.1016/j.atmosenv.2008.02.043.

Judd, L.M.; Al-Saadi, J.A.; Szykman, J.J.; Valin, L.C.; Janz, S.J.; Kowalewski, M.G.; Eskes, H.J.; Veefkind, J.P.; Cede, A.; Mueller, M.; et al. Evaluating Sentinel-5P TROPOMI tropospheric NO2 column densities with airborne and Pandora spectrometers near New York City and Long Island Sound. Atmospheric Measurement Techniques 2020, 13, 6113–6140. https://doi.org/10.5194/amt-13-6113-2020.

Levelt, P. F., Stein Zweers, D. C., Aben, I., Bauwens, M., Borsdorff, T., De Smedt, I., Eskes, H. J., Lerot, C., Loyola, D. G., Romahn, F., Stavrakou, T., Theys, N., Van Roozendael, M., Veefkind, J. P., and Verhoelst, T.: Air quality impacts of COVID-19 lockdown measures detected from space using high spatial resolution observations of multiple trace gases from Sentinel-5P/TROPOMI, Atmos. Chem. Phys., 22, 10319–10351, https://doi.org/10.5194/acp-22-10319-2022, 2022.

Liu, F.; Tao, Z.; Beirle, S.; Joiner, J.; Yoshida, Y.; Smith, S.J.; Knowland, K.E.; Wagner, T. A new method for inferring city emissions and lifetimes of nitrogen oxides from high-resolution nitrogen dioxide observations: a model study. Atmos. Chem. Phys. 2022, 22, 1333–1349. https://doi.org/10.5194/acp-22-1333-2022.

Matheron, G. Principles of geostatistics. Economic Geology 1963, 58, 1246–1266, [https://pubs.geoscienceworld.org/segweb/economic5g7e7ology/58/8/1246/3481854/1246.pdf]. https://doi.org/10.2113/gsecongeo.58.8.1246.

Petetin, H.; Guevara, M.; Compernolle, S.; Bowdalo, D.; Bretonnière, P.A.; Enciso, S.; Jorba, O.; Lopez, F.; Soret, A.; Pérez Garciia-Pando, C. Potential of TROPOMI for understanding spatio-temporal variations in surface NO2 and their dependencies upon land use over the Iberian Peninsula. EGUsphere 2022, 2022, 1–43. https://doi.org/10.5194/egusphere-2022-1056

Pommier, M. Estimations of NOx emissions, NO2 lifetime and their temporal variation over three British urbanised regions in 2019 using TROPOMI NO2 observations. Environ. Sci.: Atmos. 2023, https://doi.org/10.1039/D2EA00086E.

Pseftogkas, A.; Koukouli, M.E.; Segers, A.; Manders, A.; Geffen, J.v.; Balis, D.; Meleti, C.; Stavrakou, T.; Eskes, H. Comparison of S5P/TROPOMI Inferred NO2 Surface Concentrations with In Situ Measurements over Central Europe. Remote Sensing 2022, 14. https://doi.org/10.3390/rs14194886.

van Geffen, J.; Eskes, H.; Compernolle, S.; Pinardi, G.; Verhoelst, T.; Lambert, J.C.; Sneep, M.; ter Linden, M.; Ludewig, A.; Boersma, K.F.; et al. Sentinel-5P TROPOMI NO2 retrieval: impact of version v2.2 improvements and comparisons with OMI and ground-based data. Atmospheric Measurement Techniques 2022, 15, 2037–2060. https://doi.org/10.5194/amt-15-2037-2022.

Veefkind, J.; Aben, I.; McMullan, K.; Förster, H.; de Vries, J.; Otter, G.; Claas, J.; Eskes, H.; de Haan, J.; Kleipool, Q.; et al. TROPOMI on the ESA Sentinel-5 Precursor: A GMES mission for global observations of the atmospheric composition for climate, air quality and ozone layer applications. Remote Sensing of Environment 2012, 120, 70–83. The Sentinel Missions - New Opportunities for Science, https://doi.org/https://doi.org/10.1016/j.rse.2011.09.027.

Verhoelst, T.; Compernolle, S.; Pinardi, G.; Lambert, J.C.; Eskes, H.J.; Eichmann, K.U.; Fjæraa, A.M.; Granville, J.; Niemeijer, S.; Cede, A.; et al. Ground-based validation of the Copernicus Sentinel-5P TROPOMI NO2 measurements with the NDACC ZSL-DOAS, MAX-DOAS and Pandonia global networks. Atmospheric Measurement Techniques 2021, 14, 481–510. https://doi.org/10.5194/amt-14-481-2021.

Zhao, C.; Zhang, C.; Lin, J.; Wang, S.; Liu, H.; Wu, H.; Liu, C. Variations of Urban NO2 Pollution during the COVID-19 Outbreak and Post-Epidemic Era in China: A Synthesis of Remote Sensing and In Situ Measurements. Remote Sensing 2022, 14. https://doi.org/10.3390/rs14020419.