

# Using satellite data to help quantify Scottish greenhouse gas emissions

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## 1 Executive summary

### 1.1 Aims

The Paris Agreement aims to keep global mean temperatures to within 2° C Celsius of pre-industrial levels, with an aspirational aim of remaining within 1.5° C. To achieve this, global carbon emissions (principally CO<sub>2</sub>) have to at least halve every decade over the next century. In line with the agreement, Scotland has committed to achieving net-zero emissions by 2045. We therefore require accurate and frequently updated knowledge of human-driven emissions. Robust monitoring is essential if we are to verify progress.

At present, greenhouse gas (GHG) emissions for Scotland are published annually, approximately 18 months after the period to which they relate. The current approach combines annual production and usage statistics with estimates of how much carbon is emitted per unit measure of production and usage. An alternative approach is to look to the atmosphere. This study examines how satellite observations of the atmosphere could be used to build on existing modelling efforts and report GHG emissions well in advance of the present estimates.

In this report, we describe the software we have developed to download and interpret publicly available satellite observations of tropospheric NO<sub>2</sub>, as a proxy for fossil fuel emissions of CO<sub>2</sub> (ffCO<sub>2</sub>). The observations cover three spatial areas: onshore Scotland; the Scottish zone of the UK continental shelf; and the subset of the Scottish zone corresponding to the location of oil and gas platforms.

We used data from two satellite instruments: the Ozone Monitoring Instrument (OMI) and the TROPOspheric Monitoring Instrument (TROPOMI). Developed by the Netherlands and Finland, OMI is a spectrometer aboard NASA's Aura spacecraft that measures reflected solar radiation at visible to ultraviolet wavelengths. TROPOMI, also built by the Netherlands and launched in 2017, is the most advanced multispectral imaging spectrometer to date, extending OMI's capabilities. It sits aboard the European Space Agency's Sentinel-5 Precursor, part of its Copernicus Programme to monitor air pollution.

In the longer-term, the data collected can be used to improve emission estimates of ffCO<sub>2</sub> over Scotland in the context of the 2009 Scottish Climate Act and its 2019 amendment which commits to achieving net-zero emissions by 2045.

## 1.2 Summary of main observations

- We used variations in tropospheric NO<sub>2</sub> observed by the Ozone Monitoring Instrument (OMI) and TROPOspheric Monitoring Instrument (TROPOMI) satellite instruments, mostly reflecting changes in surface emissions, as a proxy for the combustion source of CO<sub>2</sub>.
- We found sufficient coverage of OMI and TROPOMI NO<sub>2</sub> column data to support robust estimates at monthly and annual intervals for the three target areas. OMI provides a consistent dataset from 2004 onwards, with TROPOMI providing higher spatially resolved data from 2018 onwards.
- We found the retrieval uncertainty of the tropospheric NO<sub>2</sub> data effectively described individual uncertainties associated with changes in cloud cover, surface albedo and the angle of the sun at this latitude, and consequently used it to calculate weighted mean statistics of the data.
- TROPOMI weighted means over the three Scotland zones are 6-38% larger than OMI values in 2018, but 10-70% lower in subsequent years. We attribute the positive bias of TROPOMI to its ability to sample more emission hotspots by virtue of its better horizontal spatial resolution, and we have linked the negative bias to a progressively larger number of measurements that have been flagged as poor quality by the TROPOMI team for which we do not currently understand.
- Taking advantage of OMI's 17-year record, our calculations found significant downward trends in NO<sub>2</sub> of approximately -6%/yr for the three Scottish zones considered.
- Much more could be achieved using TROPOMI. For example, its data could be used to study emissions changes from Scottish counties and from large cities. If this were to be pursued, it might be worth investing in ground-based remote sensing instruments that measure tropospheric NO<sub>2</sub> in a similar way to the satellites, but from the ground upwards.

## 2 Contents

1	Executive summary .....	1
1.1	Aims .....	1
1.2	Summary of main observations .....	2
2	Contents .....	3
	Glossary .....	4
3	Introduction .....	5
2.1	Outline of study .....	6
2.2	Satellite observations of NO <sub>2</sub> .....	6
2.3	Data analysis approach .....	8
2.1.1	Activity 1 .....	8
2.1.2	Activity 2 .....	9
2.1.3	Activity 3 .....	10
2.4	Results .....	10
2.4.1	Activity 1 .....	10
2.4.2	Activity 2 .....	11
2.4.3	Activity 3 .....	17
4	Conclusion .....	18
5	References .....	21
6	Annexes .....	22
	Annex 1: Diagnostics in OMI and TROPOMI data files .....	22
	Annex 2: Technical details of data analysis approach .....	29
	Activity 1 .....	29
	Activity 2 .....	29
	Annex 3: Technical details of results .....	30
	Activity 1 .....	30
	Annex 4: Importance of using quality assurance flags to filter data .....	31

## Glossary

**CO<sub>2</sub>**: Carbon dioxide

**ffCO<sub>2</sub>**: contribution of atmospheric carbon dioxide from fossil fuel

**geojson**: open standard format designed for representing simple geographical features

**GOME-2**: Global Ozone Monitoring Experiment-2 satellite instrument

**LEO**: Low-Earth orbit, which is typically lower than 1000 km above Earth's surface.

**Level 2 data product**: derived geophysical product described at the same resolution as the unprocessed instrument data.

**NO<sub>2</sub>**: nitrogen dioxide.

**OMI**: Ozone Monitoring Instrument satellite instrument

**Scottish Zone**: describes the various limits of the continental shelf with one of the limits being the Scottish Adjacent Waters Boundary.

**Sun synchronous orbit**: a low-Earth orbit in which the satellite passes over any given point of the planet's surface at the same local mean solar time

**TROPOMI**: TROPospheric Monitoring Instrument satellite instrument.

### 3 Introduction

The Paris Agreement aims to keep global mean temperatures to within 2° C Celsius of pre-industrial levels, with an aspirational aim of remaining within 1.5° C. To achieve this, carbon emissions (principally CO<sub>2</sub>) have to at least halve every decade over the next century (Rockström, et al. 2017). Parties to the Paris Agreement will track progress using five-year global stocktakes from 2023 onwards. We therefore require accurate and frequently updated knowledge of human-driven emissions. The Scottish Government has made a statutory commitment to meet a net-zero target by 2045 (Climate Change (Emissions Reduction Target) Scotland Act 2019). Robust monitoring of emissions is essential if we are to verify progress.

The current approach of compiling national CO<sub>2</sub> emissions combines annual production and usage statistics with estimates of how much carbon is emitted per unit measure of production and usage. Uncertainties in these values and the diversity of approaches used by different countries hamper the international comparison of estimates (Andres et al, 2012). Under the Paris Agreement, parties are required only to report national emission totals.

An alternative approach is to look to the atmosphere. Global atmospheric growth rates of CO<sub>2</sub> accurately reflect the (im)balance between surface emissions and uptake. However, atmospheric CO<sub>2</sub> measurements cannot tell us where the sampled air has been or what it has experienced. CO<sub>2</sub> has a natural biogeochemical cycle that modulates the atmospheric increase from anthropogenic emissions and consequently their impacts on Earth's climate. It is important that we can separate the influence of fossil fuel emissions from natural fluxes on atmospheric CO<sub>2</sub>.

One method of inferring fossil fuel CO<sub>2</sub> (ffCO<sub>2</sub>) is to use a trace gas that is co-emitted during the combustion process as a proxy for ffCO<sub>2</sub>. Many such gases are available from air quality networks and retrieved from current space-borne and ground-based sensors, but the most prominent candidate is nitrogen dioxide (NO<sub>2</sub>) [Konovalov et al, 2016; Goldberg et al, 2019; Reuter et al, 2019]. It has a short lifetime in the lower troposphere (<1 day), allowing for plumes to be easily identified. Space-borne sensors are typically launched in Low-Earth Orbits that pass over a particular region at the same local time every day. The instruments that are used to infer NO<sub>2</sub> columns collect solar-backscattered radiances at UV wavelengths where there are prominent NO<sub>2</sub> absorption features. These wavelengths are affected by cloud so tropospheric NO<sub>2</sub> columns (molec/cm<sup>2</sup>) are only available during cloud-free scenes, which are identified using co-retrieved cloud parameters.

In this report, we describe the software we have developed to download and interpret satellite observations of tropospheric NO<sub>2</sub> over Scotland. Using NO<sub>2</sub> as a proxy for ffCO<sub>2</sub> is consistent with the Scottish Government's strategy to meet its GHG reductions by targeting emissions from transport and industry. Having the ability to ingest satellite observations of NO<sub>2</sub> in the current GHG emission modelling framework provides constraints to improve CO<sub>2</sub> estimates, particularly during unanticipated perturbations to the Scottish economy, e.g., Covid-19.

Satellite observations of tropospheric NO<sub>2</sub> are available from a number of space-borne sensors but only the latest sensors have sufficient spatial resolution to map column enhancements of interest to the Scottish Government. We have focused on tropospheric

NO<sub>2</sub> column data collected by the Ozone Monitoring Instrument (OMI, 2004-present) and the TROPOspheric Monitoring Instrument (TROPOMI, 2017-present).

## 2.1 Outline of study

The aim of this study is to develop a capability to use satellite column observations of NO<sub>2</sub> as a proxy for ffCO<sub>2</sub> to help estimate GHG emissions for Scotland as part of wider ongoing Scottish Government activity.

The main objectives of the project were to:

- 1) Develop Python computer code to automate extraction of satellite observations of NO<sub>2</sub> from online repositories and to calculate daily, weekly, monthly, and annual mean values for a) Scotland (onshore); b) the Scottish zone of the UK continental shelf; and c) subset of the Scottish zone corresponding to the location of oil and gas extract platforms.
- 2) Develop a robust methodology, written in the Python computer language, to select or reweight observations based on their quality.
- 3) Deliver a consistent time series of historical data to which recent data (i.e. newer sensors) can be appended.

Python is an interpreted, high-level and open-source programming language. It is freely available and widely used, and runs on multiple platforms. We used the Github online code repository to maintain version control of the software we developed. The repository is currently private, meaning users must be invited to view and download the code. Code documentation is available on the Github project page and the code is commented throughout.

In Section 3 we provide a brief description of the satellite data we use and the methods we have employed to process these data to generate a consistent time series of satellite observations of NO<sub>2</sub>. In Section 4, we report our key findings, including a narrative on the results we have reported, with the strengths and limitations of our work. In Section 5 we conclude by discussing the gaps, risks, and opportunities associated with this work.

## 2.2 Satellite observations of NO<sub>2</sub>

We use data from two satellite instruments that share a common heritage: OMI and TROPOMI. Developed by the Netherlands and Finland, OMI is a spectrometer aboard NASA's Aura spacecraft that measures reflected solar radiation at visible to ultraviolet wavelengths. TROPOMI, also built by the Netherlands and launched in 2017, is the most advanced multispectral imaging spectrometer to date, extending OMI's capabilities. It sits aboard the European Space Agency's Sentinel-5 Precursor, part of its Copernicus Programme to monitor air pollution.

Table 1 summarises the main relevant details associated with the instruments. We use the NASA OMI NO<sub>2</sub> data product that shares many retrieval details of the TROPOMI retrieval (ATBD, 2019). In particular, these retrievals share the common spectral fitting window 405-465 nm and take into account the same interfering gases in the same window. NO<sub>2</sub> data from other instruments, e.g. Global Ozone Monitoring Experiment (GOME-2), have either much larger spatial footprints (80x40 km<sup>2</sup>) or very different overpass times (0930) that cannot be easily combined with data collected later in the day because of the diurnal variation in nitrogen oxide emissions from various sectors.

Table 1: Overview of OMI and TROPOMI satellite sensors.

Instrument	Launch date	Orbit, equatorial overpass time	Spatial resolution of ground pixel	NO <sub>2</sub> data product reference
OMI	15th July 2004	Sun-synchronous, 1330	24x13 km <sup>2</sup>	OMI DUG, 2012
TROPOMI	13th October 2017	Sun-synchronous, 1330	7 x 3.5 km <sup>2</sup> 5.5 x 3.5 km <sup>2</sup> from 6/8/19	PUM, 2019

Retrieval of tropospheric NO<sub>2</sub> generally involves a three-step procedure: 1) direct fitting of slant column densities to observed calibrated level 1 observed spectra; 2) separation of the slant column densities into their stratospheric and tropospheric components; and 3) conversion of these into vertical column densities.

The resulting **level 2 data product**, described on the satellite orbit tracks, is what we use for this project. The level 2 data products allow us to access measurements on the finest possible spatial and temporal scale, thereby maximising the number of cloud-free scenes. They are delivered with the scene-dependent retrieval diagnostics, data quality and assurance flags, and other parameters that will help to de-weight poor retrievals (PUM, 2019). They are also subject to ongoing calibration/validation activities (e.g. Ialongo et al, 2020) that ensure we use the best available data.

Previous versions of the OMI data product we use have been evaluated using a range of ground-based remote sensing column data, aircraft profile data, and surface in situ data that are available typically on a campaign basis (e.g., Boersma et al, 2008; Brinksma et al, 2008; Bucsela et al, 2008; Celarier et al, 2008; Lamsal et al, 2014). Over the US, OMI tropospheric NO<sub>2</sub> columns were typically within 20% of aircraft profile data but differences could be as large as 50% over coastal regions (e.g., Boersma et al, 2008; Bucsela et al, 2008; Celarier et al, 2008; Lamsal et al, 2014). Over the US and mainland Europe, OMI tropospheric NO<sub>2</sub> columns typically have a negative bias of 10-30% compared with surface remote sensing column data (Celarier et al, 2008; Lamsal et al, 2014) with the largest negative biases found over urban centres (e.g., Brinksma et al, 2008). Using a global model of atmospheric chemistry and transport as an intermediary, researchers have found that OMI tropospheric NO<sub>2</sub> columns were consistent with surface NO<sub>2</sub> data but with a seasonal bias that was largest during winter months over the US. A multi-year comparison of OMI tropospheric NO<sub>2</sub> columns with ground-based data collected over Japan show a consistent seasonal variation with an annual mean negative bias of the order of 10-15% [Lamsal et al, 2014]. Similarly, the TROPOMI tropospheric NO<sub>2</sub> product has been evaluated using a range of ground-based remote sensing data (e.g., Griffin et al, 2019; Chan et al, 2020; Ialongo et al, 2020; Zhao et al, 2020), with column generally agreeing within 10%.

We download OMI NO<sub>2</sub> data from NASA Goddard Earth Sciences Data and Information Services Center (GES DISC), as part of their Earthdata system ([https://acdisc.gesdisc.eosdis.nasa.gov/data/Aura\\_OMI\\_Level2G/OMNO2G.003/](https://acdisc.gesdisc.eosdis.nasa.gov/data/Aura_OMI_Level2G/OMNO2G.003/)).

Access to these data requires a (free) NASA Earthdata account and authorisation to download the NASA GES DISC data archive. The online repository contains all daily files stored in yearly directories. As part of this study we have downloaded the OMI NO<sub>2</sub> record relevant to the three Scottish zones to September 2020.

We download TROPOMI files via an Application Programming Interface (API) that is provided through the `sentinelat` python module (<https://sentinelat.readthedocs.io/en/stable/api.html>). This API connects to the Copernicus SciHub (<https://scihub.copernicus.eu/dhus/#/home>) and downloads files from this server.

Annex 1 summarises the retrieval diagnostics that accompany the OMI and TROPOMI level 2 data products. We discuss below how we use this auxiliary information to determine weights for data selection. Level 2 orbital files for OMI represent ~400 MB/day and for TROPOMI represent ~4.5 GB/day.

## 2.3 Data analysis approach

Figure 1 illustrates the approach we have taken to deliver our objectives. We have separated this approach into three distinct activities.

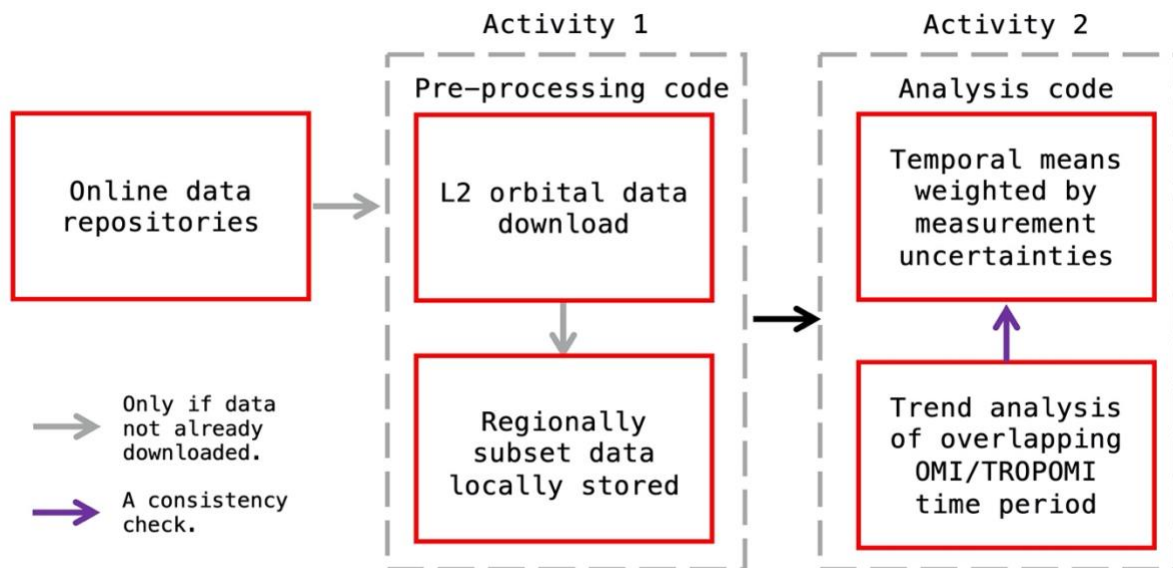


Figure 1: Overview of the processing software developed in this project.

### 2.1.1 Activity 1

Level 2 data for OMI is stored as Hierarchical Data Format Release 5 (HDF or He5), and TROPOMI data is stored online as Network Common Data Form (NetCDF) files. Both file formats can be accessed using the same methods, which we provide. The HDF and NetCDF formats have the advantage they are self-describing, including all the information that describes each data field, and is machine-independent and can be read easily by RStudio.

We developed Python code to download level 2 tropospheric NO<sub>2</sub> data, as described above, and code to subset these global data for Scotland (onshore) and the Scottish zone of the UK continental shelf (GERS, 2019), which includes offshore oil and gas platforms. We will retain data from the borders and over the North Sea, allowing us to identify plumes that originate from outside of Scotland (Figure 2).

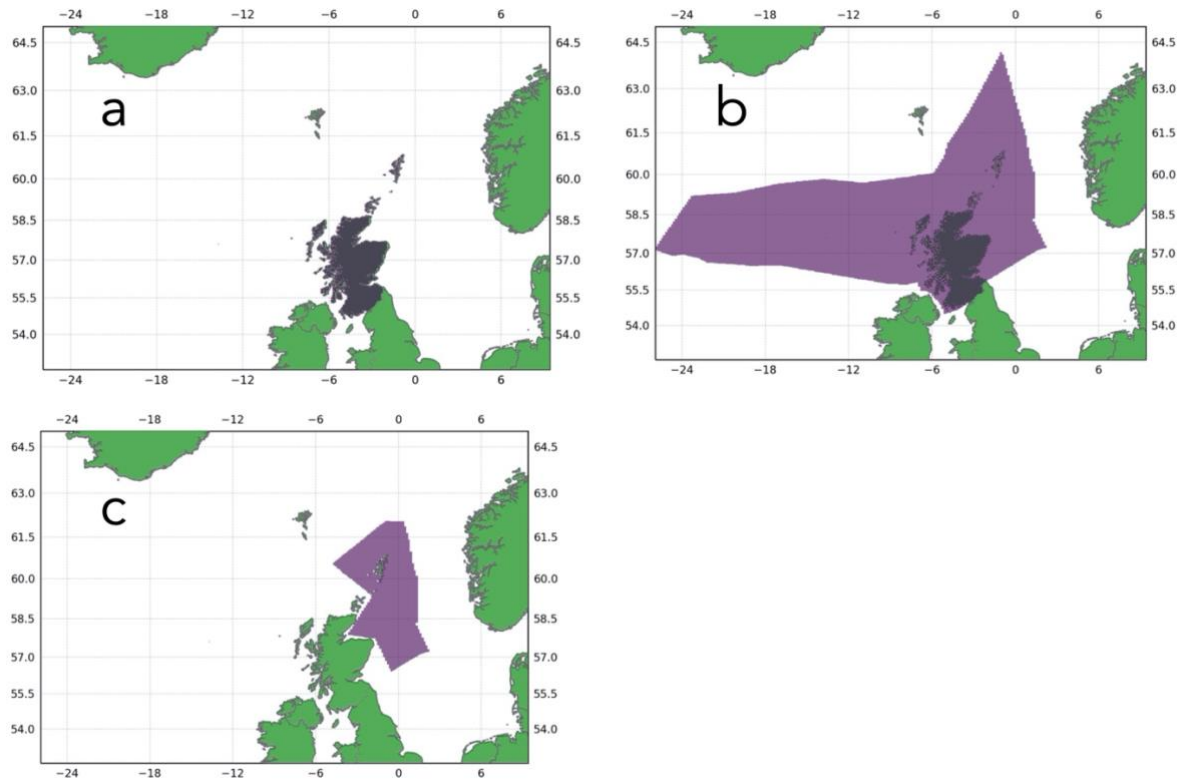


Figure 2: Data masks used to a) Scotland (onshore); b) the Scottish zone of the UK continental shelf; and c) subset of the Scottish zone corresponding to the location of oil and gas extraction platforms.

Further details about the mechanism used to download the data and the OMI and TROPOMI data products are found in Annex 2.

### 2.1.2 Activity 2

The OMI and TROPOMI Level 2 data products include a range of auxiliary variables and variables that describe the Bayesian retrieval of tropospheric NO<sub>2</sub> columns (Annex 2), which we retain in the sampled NetCDF files. Common filtering criteria include fractional cloud cover and solar zenith angle. Based on experience, these (and other) factors are typically described well by the data retrieval uncertainty but we examined them in Section 5. Data quality and assurance flags are metrics informed by some combination of retrieval diagnostics, e.g. chi-squared fit values and corresponding data retrieval uncertainty.

We have developed Python code to generate weighted daily, weekly, monthly, seasonal and annual means taking into account reported uncertainties for individual measurements (Annex 2), although as we discuss below the mean values that correspond to daily and weekly values are most sensitive to changes in data availability from cloud cover. We also report the uncertainty on the weighted mean values (Annex 2).

### 2.1.3 Activity 3

In the third and final activity, we critically assess tropospheric NO<sub>2</sub> trends across the central belt of Scotland for both OMI and TROPOMI as an approach to determine the consistency of the independent data, although some of the supportive narrative is provided in Activity 2. Based on previous comparisons of total NO<sub>2</sub> columns from OMI and TROPOMI, OMI data have a statistical uncertainty twice that of TROPOMI (van Geffen et al, 2020) but this will be reflected in the retrieval uncertainties.

## 2.4 Results

In this section we describe the outcome of the three activities defined in the previous section. The Python code we have developed to download, subsample, and analyse (Activity 3) is available in a password-protected GitHub repository, which we updated regularly throughout the project to minimise risk associated with Covid-19 and to ensure strict version control of software.

An overview of the code flow developed as part of this project and details of the password-protected code repository is found in Annex 3.

### 2.4.1 Activity 1

Figure 3 describes how code logic used to download and process level 2 tropospheric NO<sub>2</sub> data from OMI and TROPOMI repositories.

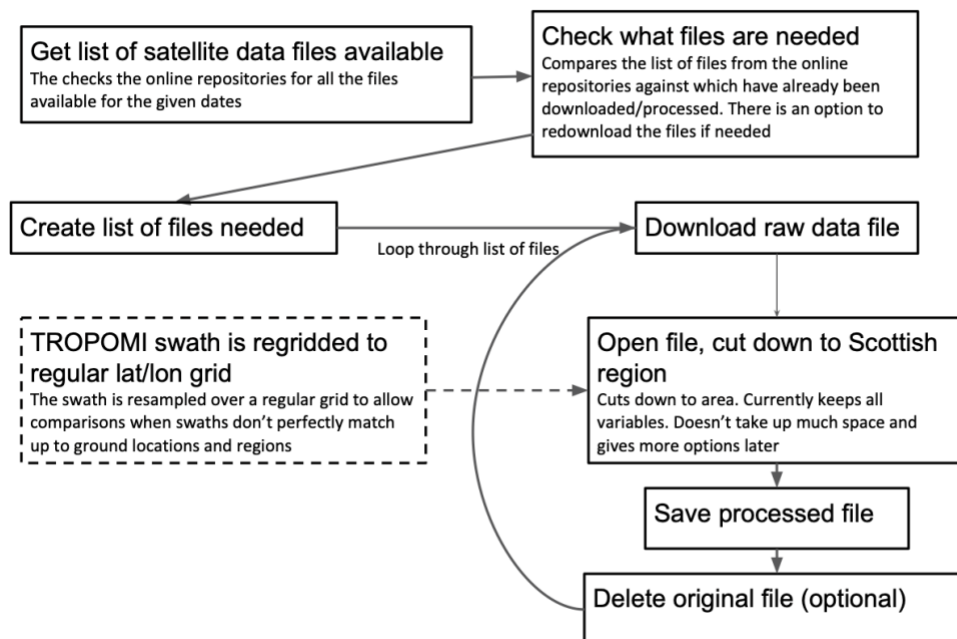


Figure 3: Overview of the code flow developed as part of this project.

All downloaded satellite data is stored in the folder 'Downloaded\_Data' in the software suite. This has further sub directories named 'OMI' and 'TROPOMI'. In each of these there are two folders named 'Raw' and 'Processed'. Once a file has been downloaded from the respective online repositories it is stored in the 'Raw' folder under the correct satellite. After processing, the file containing only the Scottish zone is stored in the 'Processed' directory and the original file in the 'Raw' directory will be deleted unless

otherwise specified. The software will detect if this file structure already exists in the specified directory and will create the folders if needed. Further details of directory and file structures are reported in Annex 3.

Table 2 provides a summary of the data volumes associated with the data downloaded and processed for OMI and TROPOMI.

Table 2 Summary of data volumes associated with OMI and TROPOMI tropospheric data. The asterisk refers to data files corresponding to the files that have been cut to the three Scottish zones.

	OMI	TROPOMI
Date Start	2004-10-01	2018-04-30
Date End	2020-09-13	2020-09-13
Months of Data	191	29
Number of Files	5793	2486
Volume Per File	1.9 MB	136.4 MB
Total Volume*	11.39 GB	353.91 GB

#### 2.4.2 Activity 2

Both OMI and TROPOMI have a data quality flag that highlights individual data points that are unphysical or should not be used. In terms of the weighted mean statistics (Annex 2) we could set these data to have very large uncertainties (small weights) but we have decided to discard them before proceeding with the statistical analysis because they are unphysical. For OMI, the literature suggest we should only consider data that have a value of 0 for the VcdQualityFlags (Annex 1). The TROPOMI use a different quality flag approach with qa\_value that has a value that ranges from 0 to 1. The scientific literature suggest we only use data with a qa\_value of 0.75 and above, but we find that adopting this value over Scotland, where there is a high likelihood of clouds and low solar zenith angles during winter, result in a large fraction of available data being discarded. Based on a trade-off analysis of qa\_value and data volume, we retain data with qa\_value values larger and equal to 0.5. Adopting these filters removes 7% of the OMI data over Scotland and 70% of the TROPOMI data over Scotland. Without these filtering steps, we have identified a number of days in which there are spurious features that cannot be readily explained (e.g. Annex 4), even after consulting with the instrument retrieval teams, and result in incorrect mean statistics.

We use the retrieval uncertainty as our individual data weights for the weighted mean statistics. To ensure we understand this approach works, we plotted in Figure 4 the scene-

dependent retrieval uncertainty for OMI and TROPOMI against cloud fraction, solar zenith angle, and surface albedo.

Cloudy scenes will influence the light path through the atmosphere and impact the quality of the retrievals. Figure 4 shows clearly that the uncertainty of OMI NO<sub>2</sub> retrievals rapidly increases with cloud fraction, as expected. The equivalent TROPOMI plot (Figure 4b) shows the opposite relationship. We find this is due to the application of the qa\_value that takes into account cloud effects on the retrievals. Figure 4b now reflects that cloud-free columns are associated with larger NO<sub>2</sub> columns and associated retrieval uncertainties. Our use of the retrieval uncertainty as weights for the weighted mean statistics for TROPOMI will now reflect mainly error due to solar zenith angle and surface albedo as described below.

Large solar zenith angles, i.e. winter months when the Sun is low in the sky, are associated with extended light paths through the atmosphere that are more difficult to describe and will be associated with large horizontal dimensions. Figure 4 shows that as solar zenith angle decreases, as we approach summer when the Sun is higher up in the sky and closer to its zenith, the NO<sub>2</sub> retrieval uncertainty decreases, as expected.

The higher the albedo the larger fraction of radiation is returned to the satellite via the atmosphere, thereby increasing the signal to noise of the observation. Figure 4 confirms that the NO<sub>2</sub> retrieval uncertainty generally decreases as surface albedo increases. These results provide confidence in our use of the NO<sub>2</sub> retrieval uncertainty as weights for the weight mean statistics.

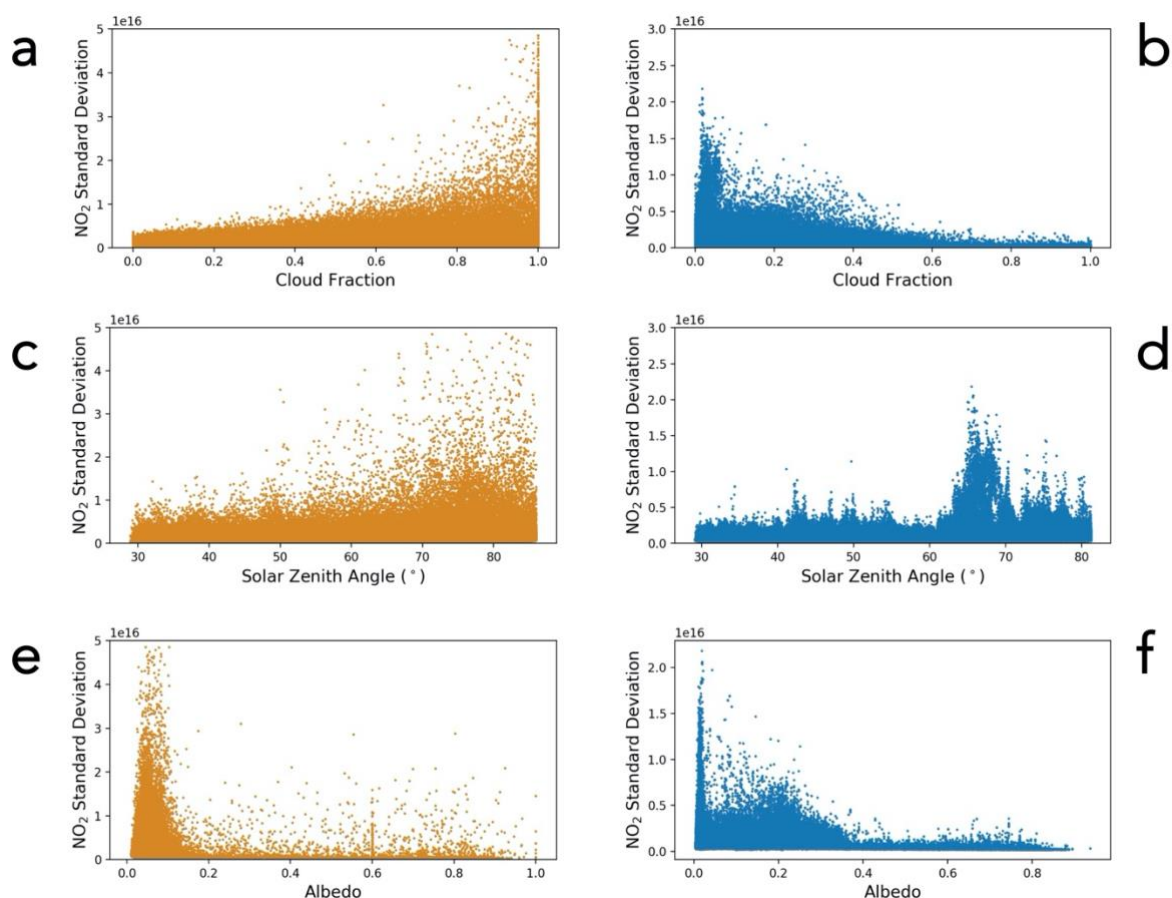


Figure 4: Scatterplots of (left) OMI and (right) TROPOMI NO<sub>2</sub> retrieval uncertainties (molec/cm<sup>2</sup>) and

(top) cloud fraction, (middle) solar zenith angle, and (bottom) surface albedo. These data fields have been filtered using the OMI and TROPOMI data assurance flags.

Figure 5 shows the mean number of good quality observations per month for OMI and TROPOMI complete records (Table 1). We find only a small seasonal variation in filtered observations for both instruments due to seasonal variations in solar zenith angle. OMI typically collects 80,000 to 100,000 raw observations per month over the Scottish zone (defined by the box domain in Figure 2), and the filtering associated with quality assurance remove typically 5-10% of observations. TROPOMI collects between 1-2 million observations per month over the wider Scottish zone. Even after the quality assurance filtering removes 70% of these data, there are 2.5-5 million TROPOMI observations per month over our wider study zone. The advantage of using TROPOMI is its superior horizontal resolution (Table 1), which means it has many more clear-sky scenes than OMI but is also means there are many scenes that are removed due to retrieval fitting issues.

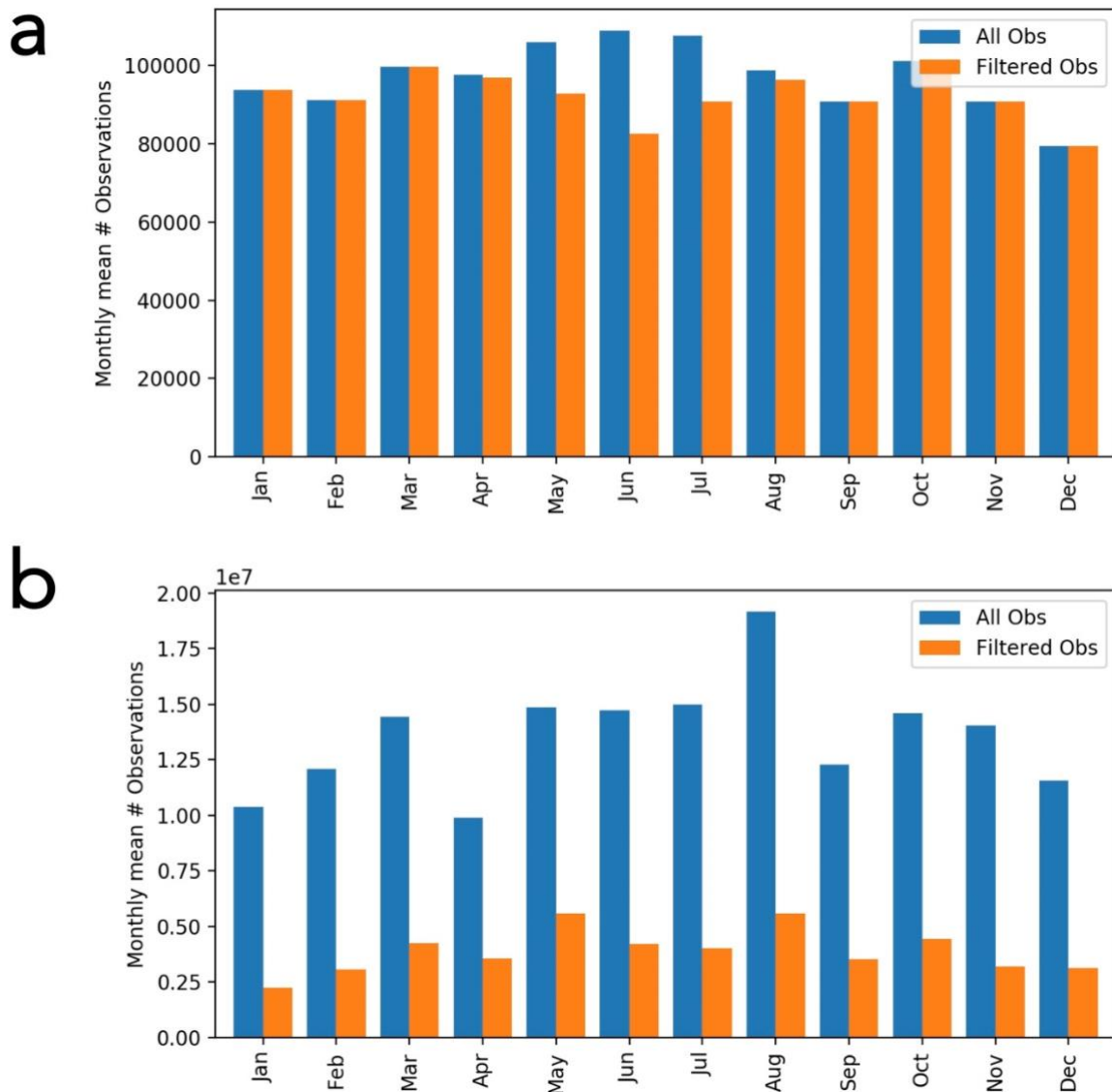


Figure 5: Monthly mean number of observations for a) OMI from 2004 to 2020 and b) TROPOMI from 2018 to 2020 with all available data (blue) and filtered data (orange). Note the difference in y axis upper limit for OMI and TROPOMI.

Using the methods outlined in Section 4 we calculate daily, weekly, monthly and annual weight means and standard deviations for OMI and TROPOMI. We find that variations in good quality vary too much over Scotland to support daily and weekly means. Instead we focus on monthly and annual weighted means.

Figure 6 shows the monthly weighted means over the three Scottish zones from 10/2004 to 9/2020 for OMI and from 4/2018 to 9/2020 for TROPOMI. We find a clear seasonal cycle in tropospheric NO<sub>2</sub> column, peaking during winter months and the lowest values occurring during summer months, reflecting seasonal emission sources. We find good qualitative agreement between OMI and TROPOMI over the different zones. During winter 2018/2019 TROPOMI columns are typically larger by 60%, suggesting they are sampling more of the high emission source regions than OMI by virtue of the instrument's higher spatial resolution which allows it to see between clouds, as expected. We find that a negative bias in the TROPOMI data during winter 2019/2020 is linked with a larger amount of data that has been flagged as poor quality by the TROPOMI team ( $qa\_value < 0.5$ ) compared to similar months in the previous year. During winter 2018/2019 there were ~8 million observations with  $qa\_value \geq 0.5$  but in the following winter there was only ~6 million observations, a reduction of 25%. At the time of writing, we have not found any reports of instrument problems during the 2019/2020 period to explain this finding so we have no reason to believe this will continue.

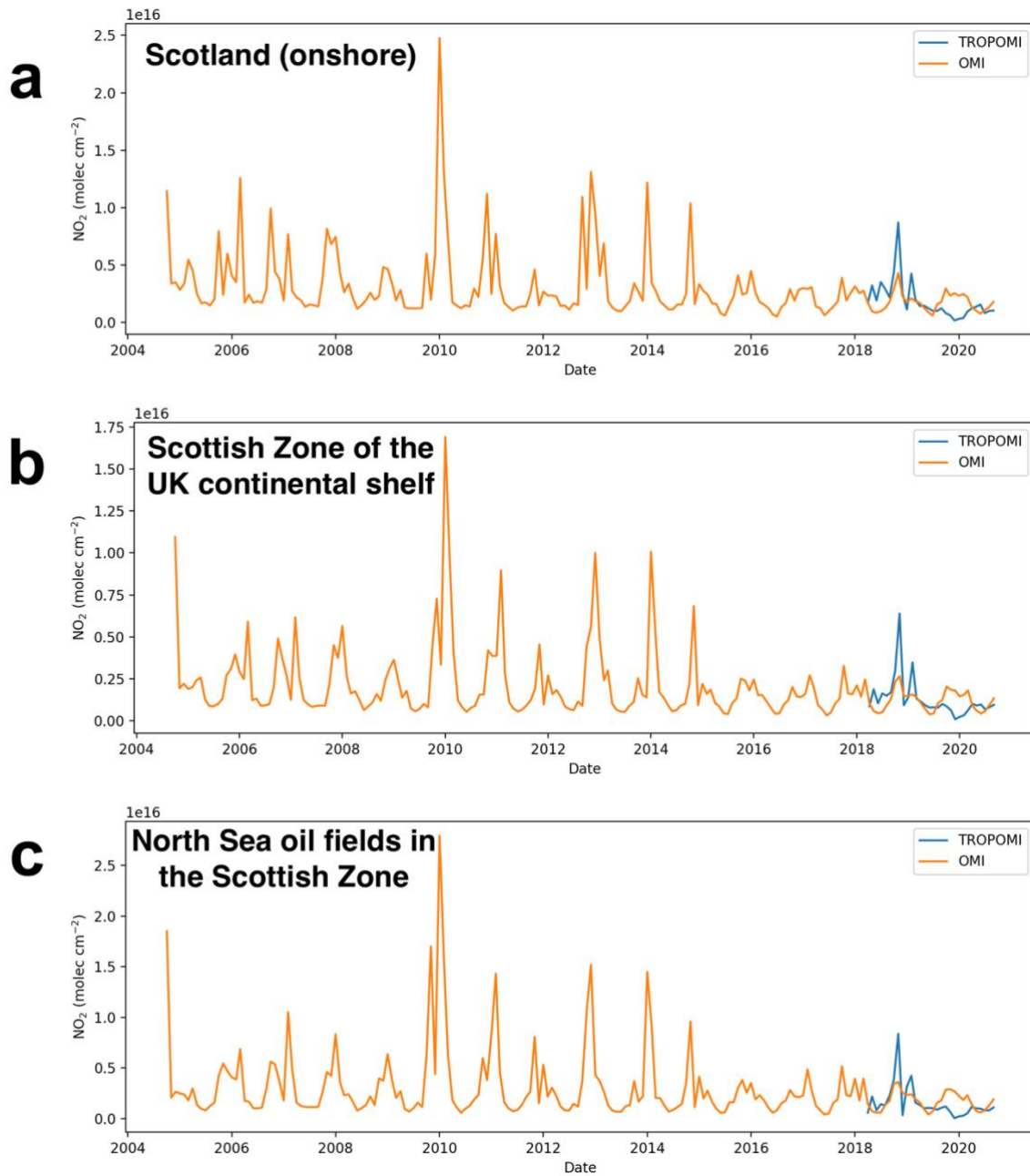


Figure 6: Monthly mean (blue) OMI and (orange) TROPOMI  $\text{NO}_2$  columns ( $\text{molec cm}^{-2}$ ) over a) Scotland (onshore); b) the Scottish zone of the UK continental shelf; and c) subset of the Scottish zone corresponding to the location of oil and gas extraction platforms. Note the difference in y axis upper limit for each zone. The associated weighted mean standard deviations are superimposed but are small compared to the mean values.

Figure 7 shows annual weighted means over the three Scottish zones from 2004 to 2020, keeping in mind that the value for 2020 includes data only up to mid-September. With the exception of the anomalous mean annual value in 2010 reported across all three zones, annual mean values are steadily declining but have a large year-to-year variation. Table 3 report the corresponding linear trends that are fitted to the annual mean values. For all three Scottish zones there is a statistically significant reduction of tropospheric  $\text{NO}_2$  from 2004 to 2020.

Figure 7 also shows the TROPOMI data from 2018 to 2020. During 2018, compared with OMI the TROPOMI NO<sub>2</sub> columns have a positive bias of +38%, +19%, and +6% over Scotland (onshore), the Scottish zone of the UK continental shelf, and the North Sea oil fields in the Scottish Zone, respectively. In contrast, during 2019 and 2020 TROPOMI NO<sub>2</sub> columns have a progressive negative bias against OMI data. During 2019, TROPOMI columns are -30%, -11%, and -29% lower than OMI columns over Scotland (onshore), the Scottish zone of the UK continental shelf, and the North Sea oil fields in the Scottish Zone, respectively. During 2020, these negative biases increase to -65%, -36%, and -71% over Scotland (onshore), the Scottish zone of the UK continental shelf, and the North Sea oil fields in the Scottish Zone, respectively. As mentioned above, the only explanation we can find is that larger amounts of data in 2019 and 2020 have qa\_value flags less than 0.5, indicative of poorer retrievals.

**Table 3** Summary of data volumes associated with OMI and TROPOMI tropospheric data. The asterisk refers to data files corresponding to the files that have been cut to the three Scottish zones.

	<b>Linear annual mean trend fitted to OMI NO<sub>2</sub> data from 2004 to 2020 (molec cm<sup>-2</sup> yr<sup>-1</sup>, %)</b>
<b>Onshore Scotland</b>	-0.13±0.05x10 <sup>15</sup> (-6.1%)
<b>Scottish zone of the UK continental shelf</b>	-0.13±0.04x10 <sup>15</sup> (-6.0%)
<b>North Sea oil fields in the Scottish zone</b>	-0.19±0.07x10 <sup>15</sup> (-6.2%)

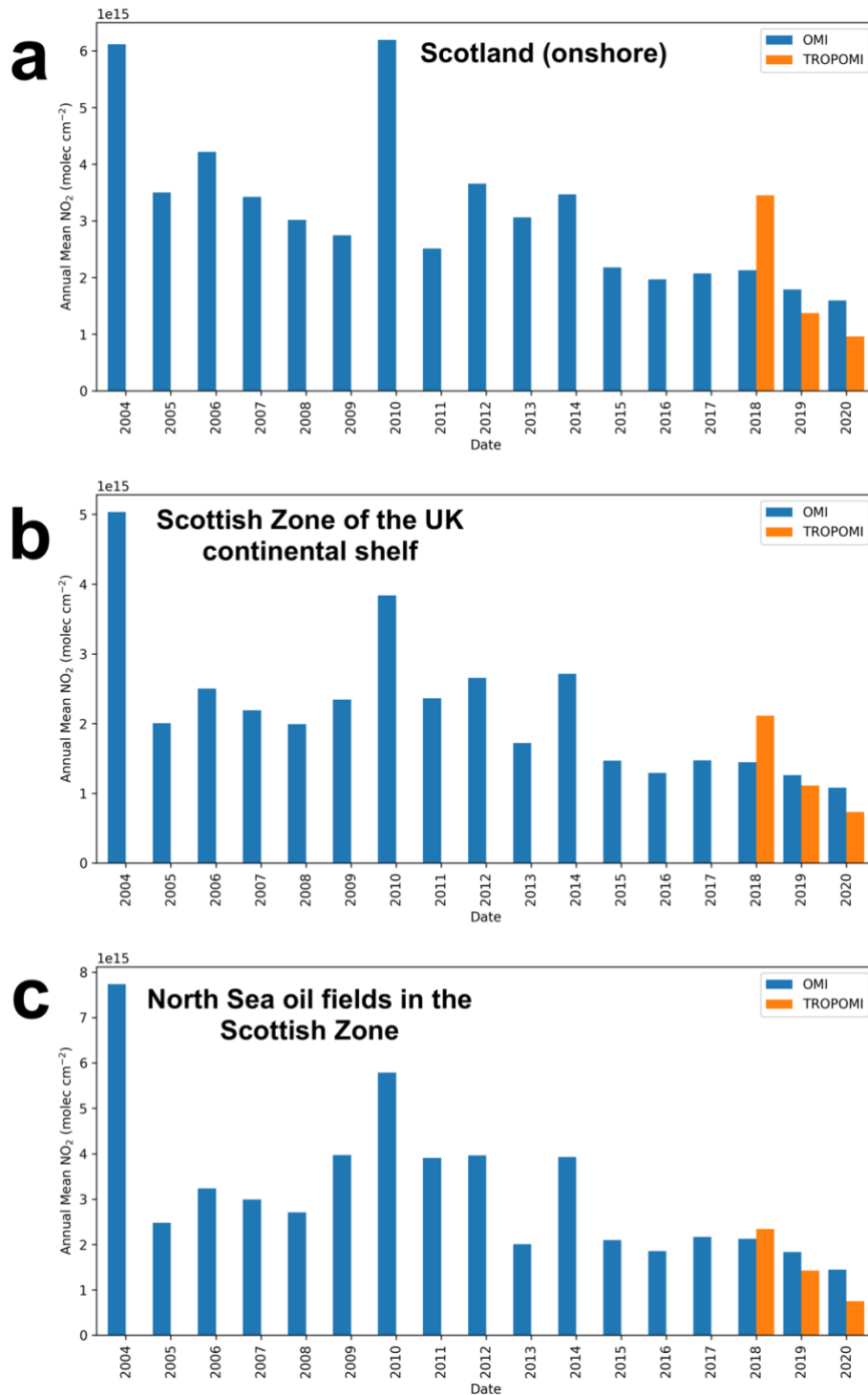


Figure 7: Mean annual observations of tropospheric NO<sub>2</sub> columns (molec cm<sup>-2</sup>) from OMI (blue) and TROPOMI (orange) over a) Scotland (onshore); b) the Scottish zone of the UK continental shelf; and c) subset of the Scottish zone corresponding to the location of oil and gas extraction platforms. Note the difference in y axis upper limit for each zone. The associated weighted mean standard deviations are superimposed but are small compared to the mean values.

### 2.4.3 Activity 3

Figure 8 shows the gradient of OMI and TROPOMI tropospheric NO<sub>2</sub> between the Clyde region (covering Glasgow and the River Clyde) and Midlothian (covering Edinburgh and the surrounding towns). Using OMI and TROPOMI data we find some evidence of higher

values in Glasgow but the difference is neither constant nor seasonal so may reflect changes in atmospheric flow between the two regions.

Figure 8 also show Defra surface NO<sub>2</sub> data for Edinburgh (St Leonards) and Glasgow (Glasgow Townhead) from the Automatic Urban and Rural Network (AURN) that forms the basis of checking nationwide compliance with ambient air quality directives. With the AURN data we find that NO<sub>2</sub> is higher by 10 ppb in Glasgow but acknowledge these sites tend to be located next to roadsides; it is difficult to attach any meaningful interpretation to the comparison since absolute values will reflect local traffic. However, the seasonal cycle of surface NO<sub>2</sub> has a distinct seasonal cycle peaking during winter months, as expected.

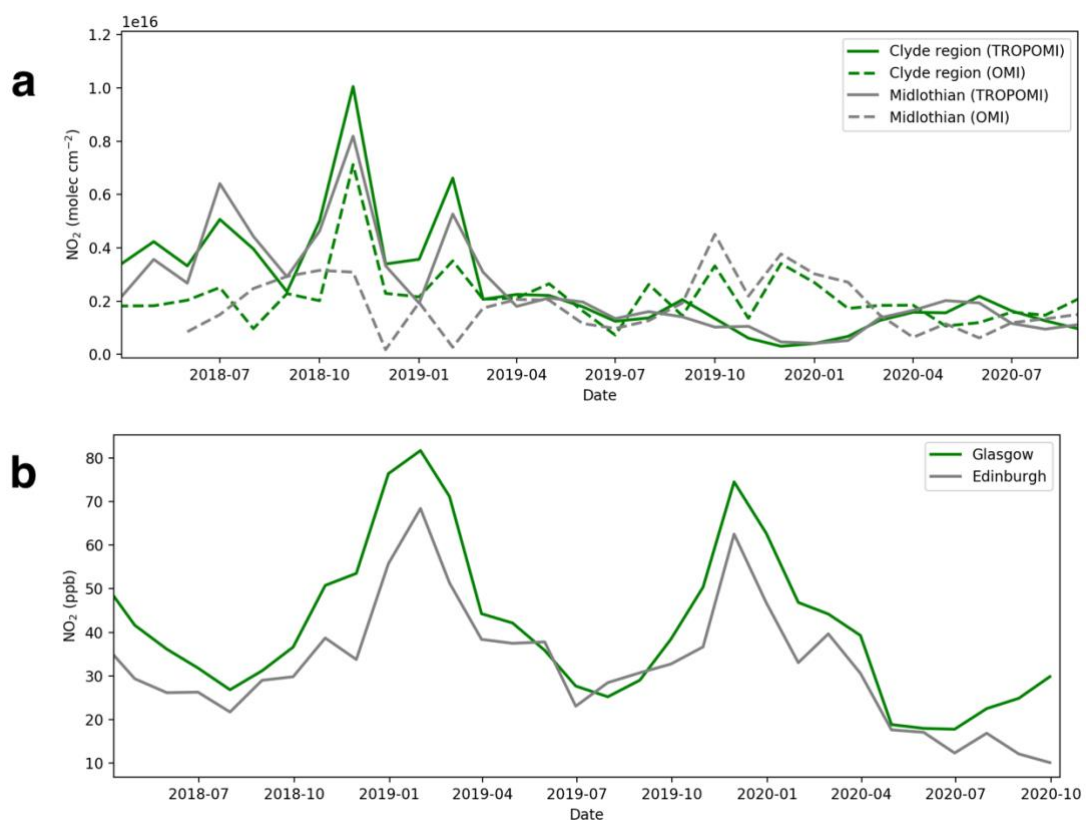


Figure 8: Clyde region (green) vs Midlothian (grey) monthly mean NO<sub>2</sub> (molec cm<sup>-2</sup>) from the TROPOMI satellite. Surface NO<sub>2</sub> concentrations (ppb) from the AURN network in Edinburgh (top) and Glasgow (bottom)

## 4 Conclusion

To the authors' knowledge, this report represents the first study of satellite observations of tropospheric NO<sub>2</sub> over Scotland. A few recent papers have focused on the UK (e.g. Pope et al, 2018), but they tend to focus on hotspots over England, e.g. Midlands and Greater London.

We used variations in tropospheric NO<sub>2</sub> over three Scottish zones, observed by the OMI and TROPOMI satellite instruments, as a proxy for combustion source of CO<sub>2</sub>. The three zones are onshore Scotland, the Scottish zone of the UK continental shelf, and the subset

of the Scottish zone corresponding to the location of oil and gas extract platforms. We use level 2 data that is closely related to measurements collected along the satellite orbits. OMI provides a self-consistent dataset from 2004 onwards, with TROPOMI providing higher spatially resolved data from 2018 onwards.

Despite Scotland being geographically small, cloudy, and at a high latitude (Section 5) the number of high-quality observations from OMI and TROPOMI have enabled robust weighted mean statistics on monthly and longer timescales. This level of cloud-free data coverage is almost exclusively due to the horizontal resolution of the sensors. Other concurrent Earth orbiting sensors, e.g. Global Observing Monitoring Experiment-2 (GOME-2) aboard the Eumetsat Sentinel satellites have a horizontal resolution of 80x40 km<sup>2</sup>, an order of magnitude larger than OMI and two orders of magnitude larger than TROPOMI. However, we found that data available on daily and weekly timescale is too variable to support robust weighted mean values. The OMI and TROPOMI instruments have a similar early afternoon overpass time, minimising any bias due to sampling different times along the diurnal cycle that is influenced by emissions, chemistry and meteorology (e.g. boundary layer height).

We found the retrieval uncertainty of the tropospheric NO<sub>2</sub> data effectively described individual uncertainties associated with changes in cloud cover, surface albedo and the angle of the sun at this latitude, and consequently used it to calculate weighted mean statistics of the data. These uncertainties are available for every observed scene, simplifying the application of weights to determined weighted mean statistics. We also found that it is important to use scene-dependent retrieval quality assurance flags that accompany both OMI and TROPOMI data products. These flags effectively highlight unphysical retrievals so that ignoring this information results in erroneous statistics.

Surprisingly, we find only a small seasonal variation in filtered observations for both instruments due to seasonal variations in solar zenith angle. OMI typically collects 80,000 to 100,000 raw observations per month over the Scottish zone, and the data assurance filtering associated with quality assurance remove typically 5-10% of observations. TROPOMI collects between 10-20 million observations per month over the wider Scottish zone. Even after the quality assurance filtering removes 70% of these data, there are 2.5-5 million TROPOMI observations per month over the wider Scottish zone. The superior horizontal resolution of TROPOMI means it has many more clear-sky scenes than OMI but is also means there are many scenes that are removed due to retrieval fitting issues.

We find that TROPOMI weighted monthly means over our three Scottish zones are, on average, larger than OMI values during 2018 and lower than OMI values in subsequent years. We attribute the positive bias to TROPOMI being able to sample more emission hotspots by virtue of its better horizontal spatial resolution that enables a larger number of observations between clouds. The negative bias is linked to a progressively larger amount of data that has been flagged as poor quality by the TROPOMI data processing team. At the time of writing, we have not found any reports that explain our finding. If, for some unforeseen incident, OMI tropospheric NO<sub>2</sub> data were to become unavailable tomorrow, the OMI data could be extended by TROPOMI but careful attention would be needed to minimize the impact of small-scale NO<sub>2</sub> hotspots on monthly weighted mean statistics.

Taking advantage of the 17-year OMI record of tropospheric NO<sub>2</sub> data over the Scottish zone, we calculated linear trends to weighted annual mean NO<sub>2</sub> values, taking into consideration the weighted standard deviations that are an order of magnitude smaller than the weight mean values. We found significant downward trends in NO<sub>2</sub> of approximately -6%/yr for all the three Scottish zones considered.

While data from the OMI instrument has allowed us to quantify trends and variations across the three Scotland zones, by virtue of the associated length of the dataset, there is much more that can be achieved using TROPOMI data. For example, the amount of data available from TROPOMI could allow the study of emission changes from Scottish counties and from large Scottish cities. Our preliminary analysis of the NO<sub>2</sub> gradient across the central belt suggests this might be possible but on smaller spatial scale more attention will need to be paid to the atmospheric lifetime of NO<sub>2</sub> and atmospheric transport.

If these data were to be pursued, it might be worth investing in ground-based remote sensing instrument that measure tropospheric NO<sub>2</sub> in a similar way to the satellites (but from the ground upwards). These instruments would provide a ground-truth to the satellite observations collected over Scotland and would potentially allow higher spatially resolved trends throughout the sunlit day. The expertise to install and maintain such instruments resides in the Scottish university sector.

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## 6 Annexes

### Annex 1: Diagnostics in OMI and TROPOMI data files

A list of diagnostics provided in the level 2 OMI and TROPOMI data products and included in the processed data files.

OMI Products:	Shape
<b>CloudFraction</b>	50 x 141
<b>CloudFractionStd</b>	50 x 141
<b>CloudPressure</b>	50 x 141
<b>CloudPressureStd</b>	50 x 141
<b>CloudRadianceFraction</b>	50 x 141
<b>ColumnAmountNO2</b>	50 x 141
<b>ColumnAmountNO2Std</b>	50 x 141
<b>ColumnAmountNO2Strat</b>	50 x 141
<b>ColumnAmountNO2StratStd</b>	50 x 141
<b>ColumnAmountNO2Trop</b>	50 x 141
<b>ColumnAmountNO2TropStd</b>	50 x 141
<b>GroundPixelQualityFlags</b>	50 x 141
<b>InstrumentConfigurationId</b>	50 x 141
<b>Latitude</b>	50
<b>LineNumber</b>	50 x 141

<b>Longitude</b>	141
<b>MeasurementQualityFlags</b>	50 x 141
<b>OrbitNumber</b>	50 x 141
<b>PathLength</b>	50 x 141
<b>SceneNumber</b>	50 x 141
<b>SlantColumnAmountNO2</b>	50 x 141
<b>SlantColumnAmountNO2Destriped</b>	50 x 141
<b>SlantColumnAmountNO2Std</b>	50 x 141
<b>SolarAzimuthAngle</b>	50 x 141
<b>SolarZenithAngle</b>	50 x 141
<b>SpacecraftAltitude</b>	50 x 141
<b>SpacecraftLatitude</b>	50 x 141
<b>SpacecraftLongitude</b>	50 x 141
<b>TerrainPressure</b>	50 x 141
<b>TerrainReflectivity</b>	50 x 141
<b>Time</b>	1
<b>TropopausePressure</b>	50 x 141
<b>VcdQualityFlags</b>	50 x 141
<b>ViewingAzimuthAngle</b>	50 x 141

<b>ViewingZenithAngle</b>	50 x 141
<b>XTrackQualityFlags</b>	50 x 141

<b>TROPOMI Products:</b>	<b>Shape</b>
<b>time</b>	1
<b>latitude</b>	1127
<b>longitude</b>	252
<b>qa_value</b>	1127 x 252
<b>nitrogen_dioxide_tropospheric_column</b>	1127 x 252
<b>nitrogen_dioxide_tropospheric_column_precision</b>	1127 x 252
<b>nitrogen_dioxide_tropospheric_column_precision_kernel</b>	1127 x 252
<b>averaging_kernel</b>	1127 x 252
<b>air_mass_factor_troposphere</b>	1127 x 252
<b>air_mass_factor_total</b>	1127 x 252
<b>tm5_tropopause_layer_index</b>	1127 x 252
<b>tm5_constant_a</b>	1127 x 252
<b>tm5_constant_b</b>	1127 x 252
<b>satellite_latitude</b>	1127 x 252

<b>satellite_longitude</b>	1127 x 252
<b>satellite_altitude</b>	1127 x 252
<b>satellite_orbit_phase</b>	1127 x 252
<b>solar_zenith_angle</b>	1127 x 252
<b>solar_azimuth_angle</b>	1127 x 252
<b>viewing_zenith_angle</b>	1127 x 252
<b>viewing_azimuth_angle</b>	1127 x 252
<b>latitude_bounds</b>	1127 x 252
<b>longitude_bounds</b>	1127 x 252
<b>geolocation_flags</b>	1127 x 252
<b>surface_altitude</b>	1127 x 252
<b>surface_altitude_precision</b>	1127 x 252
<b>surface_classification</b>	1127 x 252
<b>instrument_configuration_identifier</b>	1127 x 252
<b>instrument_configuration_version</b>	1127 x 252
<b>scaled_small_pixel_variance</b>	1127 x 252
<b>surface_pressure</b>	1127 x 252
<b>surface_albedo_nitrogendioxide_window</b>	1127 x 252
<b>surface_albedo</b>	1127 x 252

<b>cloud_pressure_crb</b>	1127 x 252
<b>cloud_fraction_crb</b>	1127 x 252
<b>cloud_albedo_crb</b>	1127 x 252
<b>scene_albedo</b>	1127 x 252
<b>apparent_scene_pressure</b>	1127 x 252
<b>snow_ice_flag</b>	1127 x 252
<b>aerosol_index_354_388</b>	1127 x 252
<b>processing_quality_flags</b>	1127 x 252
<b>number_of_spectral_points_in_retrieval</b>	1127 x 252
<b>number_of_iterations</b>	1127 x 252
<b>wavelength_calibration_offset</b>	1127 x 252
<b>wavelength_calibration_offset_precision</b>	1127 x 252
<b>wavelength_calibration_stretch</b>	1127 x 252
<b>wavelength_calibration_stretch_precision</b>	1127 x 252
<b>wavelength_calibration_chi_square</b>	1127 x 252
<b>wavelength_calibration_irradiance_offset</b>	1127 x 252
<b>wavelength_calibration_irradiance_offset_precision</b>	1127 x 252

<b>wavelength_calibration_irradiance_chi_square</b>	1127 x 252
<b>nitrogendioxide_stratospheric_column</b>	1127 x 252
<b>nitrogendioxide_stratospheric_column_precision</b>	1127 x 252
<b>nitrogendioxide_total_column</b>	1127 x 252
<b>nitrogendioxide_total_column_precision</b>	1127 x 252
<b>nitrogendioxide_total_column_precision_kernel</b>	1127 x 252
<b>nitrogendioxide_summed_total_column</b>	1127 x 252
<b>nitrogendioxide_summed_total_column_precision</b>	1127 x 252
<b>nitrogendioxide_slant_column_density</b>	1127 x 252
<b>nitrogendioxide_slant_column_density_precision</b>	1127 x 252
<b>nitrogendioxide_slant_column_density_stripe_amplitude</b>	1127 x 252
<b>ozone_slant_column_density</b>	1127 x 252
<b>ozone_slant_column_density_precision</b>	1127 x 252
<b>oxygen_oxygen_dimer_slant_column_density</b>	1127 x 252
<b>oxygen_oxygen_dimer_slant_column_density_precision</b>	1127 x 252

<b>water_slant_column_density</b>	1127 x 252
<b>water_slant_column_density_precision</b>	1127 x 252
<b>water_liquid_slant_column_density</b>	1127 x 252
<b>water_liquid_slant_column_density_precision</b>	1127 x 252
<b>ring_coefficient</b>	1127 x 252
<b>ring_coefficient_precision</b>	1127 x 252
<b>polynomial_coefficients</b>	1127 x 252
<b>polynomial_coefficients_precision</b>	1127 x 252
<b>intensity_offset_coefficients</b>	1127 x 252
<b>intensity_offset_coefficients_precision</b>	1127 x 252
<b>cloud_fraction_crb_nitrogen_dioxide_window</b>	1127 x 252
<b>cloud_radiance_fraction_nitrogen_dioxide_window</b>	1127 x 252
<b>chi_square</b>	1127 x 252
<b>root_mean_square_error_of_fit</b>	1127 x 252
<b>degrees_of_freedom</b>	1127 x 252
<b>air_mass_factor_stratosphere</b>	1127 x 252
<b>nitrogen_dioxide_ghost_column</b>	1127 x 252

## Annex 2: Technical details of data analysis approach

### Activity 1

We originally planned to use the Open-source Project for a Network Data Access Protocol (OPeNDAP) service provided by NASA to download the OMI data, but we found this service to be unreliable for OMI data and did not work in the way we had expected. Instead, we downloaded the OMI files using data URLs via the Python software. This method downloads entire files at once, including all variables and global coverage. Although it is slower and initially takes up more space than the OPeNDAP method, it is stable and does not rely on as much external software maintenance.

We download and analyse Level 2G OMI data (described in Section 3), which is a Level 2 product that has been resampled by the OMI data onto a regular spatial grid. This gridding step makes it easier to process the data. Each Level 2G data file contains global data for one day, which we download before being generating the Scottish data products for the three zones (Figure 2). The Python code is sufficiently flexible that we can easily reprocess with different data masks.

We download the TROPOMI data that are available in more conventional level 2 files, i.e., each file describes one satellite swath from South to North<sup>1</sup>. There are multiple files per day and each swath covers a different region of the globe. To ensure the downloaded files contain some information relating to the broader Scottish zone, thereby avoiding downloading files that do not cover our study area, we pass our geographical information to the TROPOMI download function. We do this by using a geojson file<sup>2</sup> that contains the coordinates of the study zones.

### Activity 2

We use the following definition to determine the weight (*wtd*) mean of tropospheric NO<sub>2</sub> using:

$$\bar{x}_{wtd} = \frac{\sum_{i=1}^n w_i x_i}{\sum_{i=1}^n w_i}$$

where the *i*th weight  $w_i = 1/\sigma_i^2$  and  $\sigma_i$  corresponds to the Bayesian retrieval uncertainty estimates of an individual measurement. The corresponding standard deviation about the weighted mean value,  $\sigma_{wtd}$ , is given by:

$$\sigma_{wtd} = \frac{\sum_{i=1}^n (x_i - \bar{x}_{wtd})^2 w_i}{\sum_{i=1}^n w_i} \frac{n}{n-1}$$

where all variables are as previously defined. The  $n/n-1$  factor is used to take into account the number of degrees of freedom so the value represents an unbiased estimate of the variance of the population from the individual values were sampled.

---

<sup>1</sup> UV/Vis satellite retrievals of trace gases require sunlight. The South to North direction corresponds to the sunlit hemisphere direction of the orbit.

<sup>2</sup> This is an open standard format designed for representing simple geographical features.

## Annex 3: Technical details of results

Figure 9 shows an overview of the Python code flow structure that we have developed as part of this project.

The repository can be found at [https://github.com/dpfinch/Scottish\\_NO2](https://github.com/dpfinch/Scottish_NO2) and consists of python files containing the main code, geojson files containing information about data masks (within a folder named 'Masks') and a README file describing the project and instructions on how to use the code. It has been formatted and commented so it can be downloaded by Scottish Government scientists.

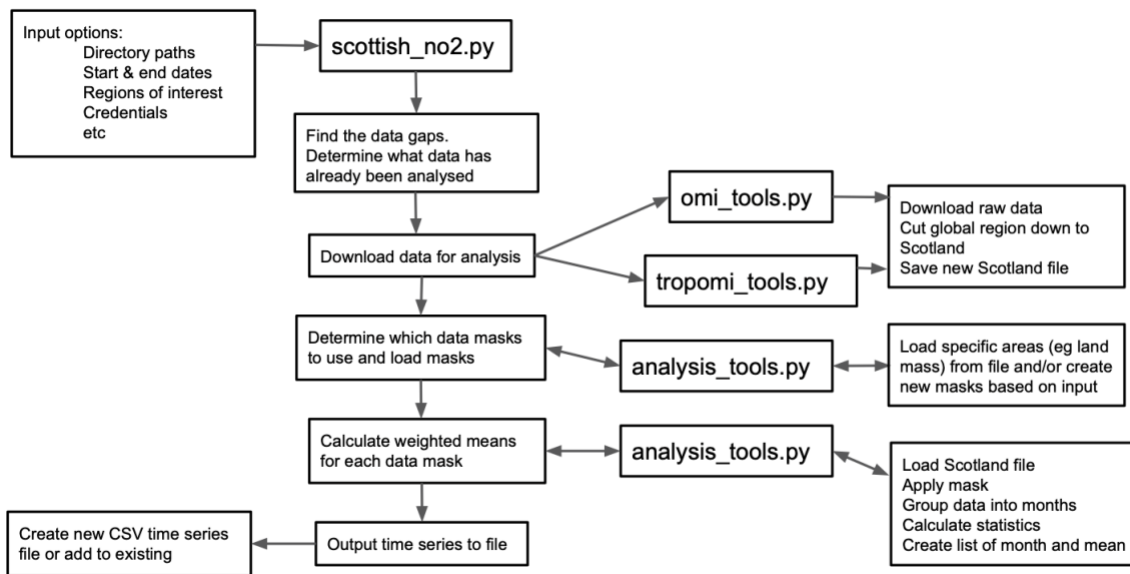


Figure 9: Overview of the code flow developed as part of this project.

### Activity 1

Statistical output is stored in the 'Output' directory and contains comma separated variable (CSV) files. Each of these files have a descriptive name that follows the pattern: '{SATELLITE}\_NO2\_{ZONE}\_{DATE TYPE}\_Mean.csv'

For example:

'OMI\_NO2\_OilGas\_Monthly\_Mean.csv'

Each of these files contain a time series consisted of three columns: Date, NO<sub>2</sub>, and NO<sub>2</sub> standard error. OMI files are about 6 KB in size and TROPOMI files are about 1 KB in size.

Locally stored data subsets will be stored as self-describing NetCDF files that speed up data access in later analysis steps

## Annex 4: Importance of using quality assurance flags to filter data

Figure 10 shows an example case in which unfiltered level 2 TROPOMI data shows a large spike in the tropospheric NO<sub>2</sub> column on the 9th July 2019 that is not present in data on the previous day. The anomalous NO<sub>2</sub> feature that is in excess of  $8 \times 10^{16}$  molecules cm<sup>-2</sup> and two orders of magnitude larger than a typical tropospheric NO<sub>2</sub> column over this region significantly influences the weighted monthly value for July 2019; the resulting weighted monthly values is four times larger than the next highest month. Using the quality assurance flag effectively removes these data from subsequent analysis.

Private communication with the TROPOMI retrieval team suggests this could be influenced by an observable feature. Without further analysis of these data using an atmospheric chemistry transport model it would be difficult to include this day of data in any analysis. Defra AURN data over Aberdeen (not shown) did not report elevated surface NO<sub>2</sub> on the 9<sup>th</sup> July 2019.

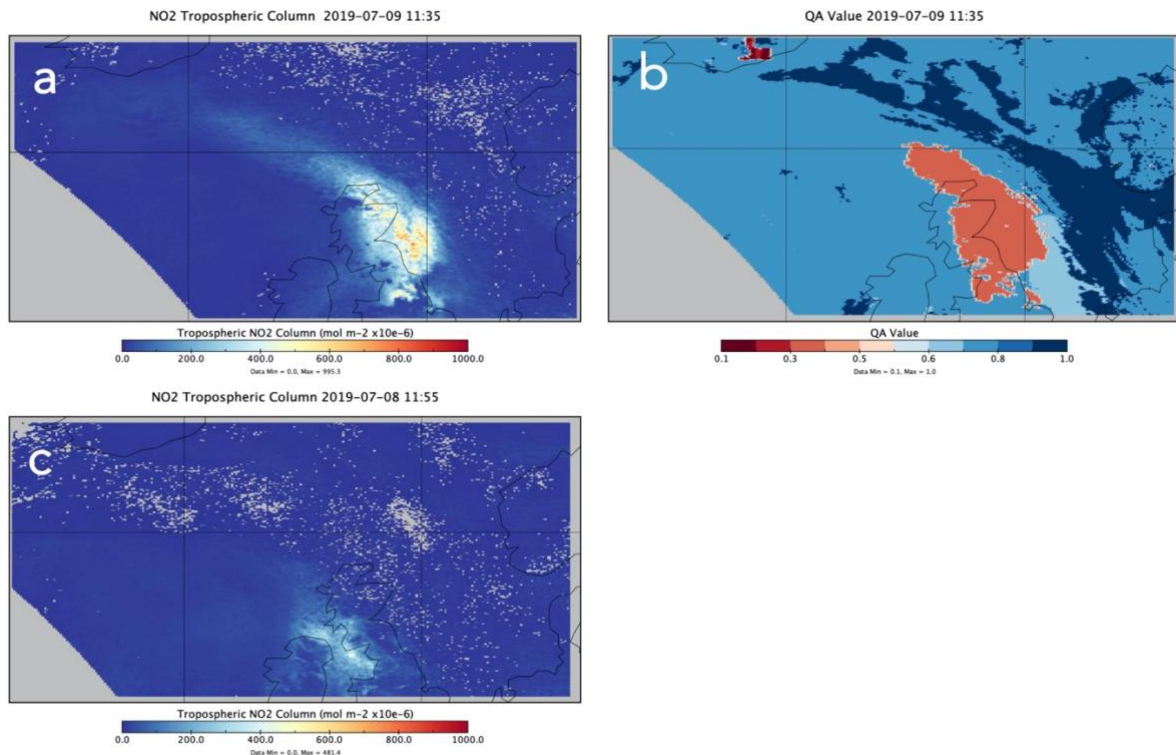


Figure 10: Illustration of anomalous TROPOMI level 2 NO<sub>2</sub> column data (molec cm<sup>-2</sup>) on a) 8th and c) 9th July 2019 over Scotland and the surrounding ocean. The TROPOMI qa\_value corresponding to the 9th July 2019 is shown in panel b.

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