Master Thesis

Retrieval of methane emissions of the oil and gas industry in Romania from TROPOMI aboard Sentinel 5 Precursor

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1. Introduction

1.1. Methane

Methane (CH_4) is an important greenhouse gas emitted anthropogenically from several human activities, including oil and gas production. Methane is less atmospherically abundant than carbon dioxide, which is responsible for the largest share of anthropogenic radiative forcing. Nevertheless, methane has a significantly higher global warming potential, and an atmospheric lifetime of around 12 years, which is technically long, but not in comparison to carbon dioxide's lifetime of centuries. With a lifetime of a dozen years, the earth's radiative budget will be affected in a short time if a reduction in atmospheric methane can be achieved. Nevertheless, methane detection and measurements are demanding, because methane is well-incorporated in the atmosphere, and only small changes within large background concentrations are used for detection. That makes obtaining accurate CH_4 measurements more challenging (Schneising et al., 2019; Hu et al., 2018).

There are anthropogenic and natural sources of atmospheric methane. Agriculture, wastes, fossil fuels (including oil and gas), and biomass burning are considered anthropogenic sources, whereas natural sources include wetlands, freshwaters, geological seepage, and wildfires. The fossil fuel called natural gas consists mainly of methane, and it could have either thermogenic or microbial origins. Thermogenic gas is formed due to heat and pressure in deep geological formations, whereas microbial gas is formed by the microbial consumption of hydrocarbons, in addition to the organic matter breakdown process in the shallow geological formations (Milkov et al., 2020; Lopez et al., 2017).

Methane's chemical composition (CH_4) consists of two elements: carbon and hydrogen. Both elements have two stable isotopes: carbon-12 (^{12}C) and carbon-13 (^{13}C); and protium (^{1}H) and deuterium (^{2}H or D), respectively. The carbon isotopic composition of methane allows a distinction to be made between the exact sources of methane. Thermogenic methane, specifically from oil and gas production, tends to be rich in ^{13}C, while microbial methane is extremely depleted in it (Lopez et al., 2017). By analyzing the isotopic composition of an air sample collected from a certain location, a methane source could be determined by *in-situ* measurements with continuous flow isotope ratio mass spectrometry (CF-IRMS) (Menoud, et al., 2020). Unfortunately, when it comes to satellite measurements of atmospheric methane, differentiating between emissions that come from oil- and gas-production or agriculture requires certain prior knowledge of methane isotopic information, of their different source types and distribution, as well as definite meteorology knowledge. However, this is still not possible to achieve yet from satellite observations (Schneising et al., 2019).

1.2. ROMEO Campaign

Romania is considered one of the countries with the highest atmospheric methane emissions from oil and gas production in Europe, according to UNFCCC statistics in 2015. The current methane emissions reports from Romania are based on estimates, and the emission figures are from standard emission factors, meaning that the accuracy of these figures is questionable (Rockmann et al., 2020).

ROmanian Methane Emissions from Oil & gas (ROMEO) is part of the international Climate and Clean Air Coalition (CCAC's) Methane Science Studies project. It is a campaign that took place in Romania and focused on the oil and gas production sector. ROMEO investigated methane emissions, specifically in areas around Bucharest and in Transylvania, as the largest natural gas fields are found there. Methane and other atmospheric parameters were measured by specific instruments that were placed on aircrafts, drones, and cars. This campaign is extremely important as these measurements will help in producing more accurate methane emissions. Their updated quantification will lead to better emission mitigation measures for the gas and oil sector in Romania. Limiting these methane emissions can reduce the greenhouse gas emissions for the EU, so this possibility is attractive (Rockmann et al., 2020).

The first step of the ROMEO campaign took place in August 2019. It consisted of quantifying methane emissions in Bucharest and Ploiesti, at street level, by using three vehicles. This was then followed by isotopic analysis, and ethane-methane ratio measurements, to determine the sources of the methane. In October 2019, the main campaign took place where eight ground measurement teams carried out methane measurements in more than 1,000 individual facilities. The teams took measurements with both stationary and mobile vehicles by using approaches of tracer release and drone plume mapping. This quantification stage was challenging in low wind speeds. Nevertheless, around 200 quantifications were attempted. In addition, methane emissions from individual facilities, cluster facilities, and extended regions were identified and quantified in order to connect the scales at facility-level and the regional-level. This process was achieved by using two research aircraft from the "Elie Carafoli" National Institute for Aerospace Research

(INCAS), and Scientific Aviation Inc., and it involved more than 20 research flights (Rockmann et al., 2020).

ROMEO’s final goal is to create an approach that combines bottom-up and top-down concerns in order to quantify any methane emissions that are related to the oil and gas sectors in Romania. This includes the distribution of natural gas, its use, as well as oil and gas exploration (Rockmann et al., 2020).

1.3. Sentinel 5-Precursor - TROPOMI

The single-payload satellite, Sentinel 5 Precursor (Sentinel 5P), was launched in October 2017, carrying the TROPOshperic Monitoring Instrument (TROPOMI). TROPOMI is a spectrometer that measures solar radiation reflected by Earth and has spectral bands in UV, VIS, NIR and SWIR. This wide range of wavelengths achieves the goal of observing several crucial atmospheric species, including ozone (O_3), carbon monoxide (CO), methane (CH4), aerosols, and clouds. TROPOMI has a wide swath width of 2,600 km, and a high spatial resolution, in addition to a daily global coverage (Schneising et al., 2019; Veefkind et al., 2012).

Methane total column concentrations can be retrieved by using radiance measurements from the Shortwave Infrared (SWIR) channel of TROPOMI. This is because it shows strong absorption features by methane, with a spatial resolution of 7 km × 7 km for the nadir measurements (Schneising et al., 2019). When it comes to retrieval of methane, scattering by aerosols and cirrus clouds become a major challenge, especially if the scatterers are optically thin to detect, unlike thick clouds that could be easily detected and filtered out.

This drawback can lead to underestimations or overestimations of the true methane column, if not taken into consideration (Apituley et al., 2017). But when taking atmospheric scattering into account, highly accurate measurements can be made. For methane data retrieval, this is possible with a combination of both SWIR and Near Infrared (NIR) channels, which have a spatial resolution of 5.5 km × 7 km (across × along track) since 6 August 2019 (orbit 9,388). The SWIR channel gives the methane concentrations, and the combination of the SWIR and NIR channels limits the atmospheric scattering properties (Butz et al., 2012).

Table 1: NIR and SWIR spectral ranges used for methane retrieval from Sentinel 5P.

Band	Spectral Range
NIR	757 – 774 nm
SWIR	2305 – 2385 nm

Methane column retrieval measurements vary with atmospheric, surface, and viewing conditions. Sentinel 5P instruments provide around 260 measurements per second. However, when it comes to methane retrieval, this number is substantially smaller, as measurements are only considered for those at the dayside of the orbit, over land, and cloud-free, thus leading to a reduction of ~80% in total measurements. All Sentinel 5P Level 2 products have two versions: Near Real-Time (NRT) and Offline (OFFL). In NRT, data is provided within three hours of sensing. In OFFL, data is provided two weeks after sensing, and methane data is only available OFFL. The OFFL products have data from a single orbit, and only for a single hemisphere, as half the earth is always dark (Fehr, 2016).

Two-years' worth of TROPOMI CH4 column average mixing ratio measurements were validated against the Total Carbon Column Observing Network (TCCON), showing an overall good agreement between TCCON data and satellite measurements (±5.6 ppbv standard deviation and -3.4 mean bias). A recent validation found that TROPOMI CH4 column average mixing ratio measurements is also in an overall good agreement with the Network for the Detection of Atmospheric Composition Change (NDACC), as well as corresponding data product from GOSAT satellite (Landgraf et al., 2020).

1.4. Research Objectives

In this master thesis, for the study period of three years, 2019, 2020 and 2021, we will explore the relationship between TROPOMI CH4 column average mixing ratio retrievals and wind speed over Ploiesti, as well as averaging TROPOMI CH4 column average mixing ratio retrievals according to two wind direction categories for the three years-period. This is also conducted for two five-consecutive days periods corresponding to specific wind characteristics, to understand how atmospheric CH4 is impacted by wind activities and using NOAA HYSPLIT model for affirmation.

Finally, the newly retrieved CH4 measurements from the AVIRIS-NG imaging spectrometer flown in Europe in summer 2021, as part of the ROMEO Campaign, and TROPOMI CH4 column average mixing ratio are compared on the one available case.

2. Data

2.1. Study Area and Period

The main study area for this project is Romania. In 2018, it was reported that the total greenhouse gases emissions of Romania are equivalent to 116,115 kt CO2, where 24% of these emissions are made up of CH4. The reported UNFCCC methane emissions indicated a 61.2% decrease between the year 1989 and 2017 (Crosman, 2021).

Within Romania, we focus on one specific city in the south as indicated in figure 1: Ploiesti, Romania. This city is Romania's 'Historical city of oil production'. A study conducted in Romania measuring street-level CH4 emissions, has reported a total of 76 locations that are considered as locations with CH4 leaks in Ploiesti (Crosman, 2021). Additionally, according to the European Pollutant Release and Transfer Register (E-PRTR) database (version 17), up until 2017, there are six facilities in Ploiesti registered as having 'oil and gas refineries' as their main activities (E-PRTR, 2019).

The study period is three years, starting from 1 January 2019 to 31 December 2021.

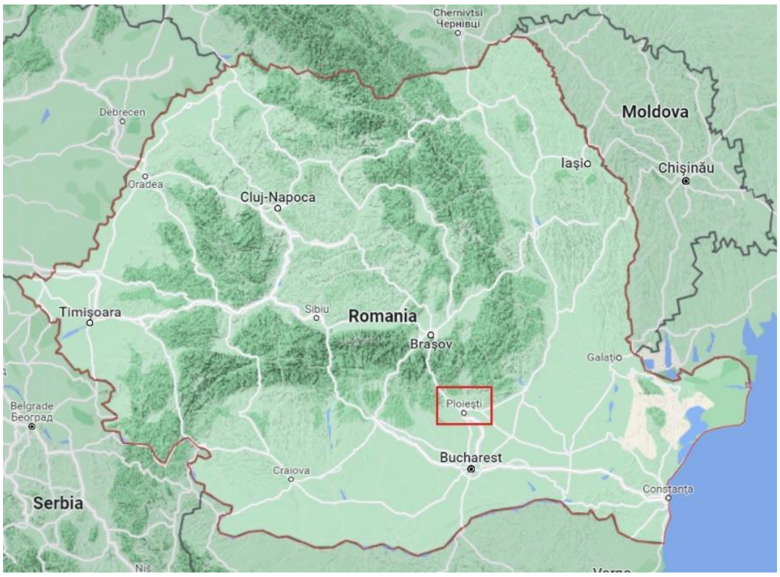


Figure 1: Map of Romania. The red borders indicate the city Ploiesti (Google Maps).

2.2. Sentinel 5P – TROPOMI Methane Observations

The TROPOMI methane product are data files that contain 'the column average dry air mixing ration of methane (XCH4). This is the total atmospheric column between the surface and the top of the atmosphere normalized to the corresponding dry air column. Data on methane total column concentrations is retrieved by using both the SWIR and NIR channels, which have a spatial resolution of 7 km × 7 km, and 5.5 km × 7 km (across × along track), respectfully. They also have a swath width of ~2,600 km on the Earth's surface. Data is retrieved from the Copernicus Sentinel-5P Pre-Operations Data Hub by sensing date, selecting an area that covers all the country Romania, from the Sentinel 5P – TROPOMI Level 2 files. Only pixels with a qa_value above 0.5 are used to ensure no misinterpretation of the data quality, as recommended by the European Space Agency (ESA), where cloud-contaminated and other poor-quality retrievals are eliminated (Landgraf et al., 2020).

2.3. ECMWF – ERA5 Weather Data

Weather data were obtained from the ERA5 reanalysis data developed by the ECMWF. Compared to its predecessor ERA-Interim, ERA5 is significantly improved, as well as being an open access. ERA5 reanalysis has a horizontal resolution of 0.25º × 0.25º, and an hourly temporal resolution. The weather data was downloaded from the C3S Climate Data Storage (CDS) through the ECMWF website.

The two main variables retrieved from the ERA5 reanalysis data are: the 100 m u- component of wind and the 100 m v- component of wind. The 100 m u- component parameter represents the eastward component of 100 m wind, as in the horizontal speed of air blowing towards the east, whereas the 100 m v- component parameter represent the northward component of 100 m wind, meaning the horizontal speed of air moving towards the north, both at a 100 m height above the earth's surface in metres per second (m/s).

2.4. AVIRIS-NG Imaging Spectrometer Measurements – ROMEO Campaign

AVIRIS-NG (Airborne Visible InfraRed Imaging Spectrometer - Next Generation) provides access to high signal-to-noise ration imaging spectroscopy measurements in the reflected sunlight spectral range, and it measures the wavelength range of 380 – 2510 nm.

AVIRIS-NG was flown in summer of 2021 in Europe as part of the ROMEO campaign, where atmospheric CH4 measurements were taken. This data is still a working progress, as a team from EMPA is currently working on determining and listing all the CH4 point sources that are detectable in the campaign data. The EMPA team provided us with their findings so far, where they retrieved three strong CH4 point sources from processing one measurement stripe from the AVIRIS-NG flight.

3. Methods

3.1. Generating S5P CH4 Level 3 Products

The TROPOMI level 2 (L2) methane products are files that are binned by time, and, by default, the pixels of S5P are not equal in size due to the large swath size of 2600 km. So, in order to have files with common temporal and spatial grid, we use the tool HARP from the ESA atmospheric toolbox to generate level 3 (L3) CH4 product. HARP is a toolkit for ingesting, processing and inter-comparing satellite or model data, and is a part of the Atmospheric Toolbox developed specifically for ESA. L3 products are generated by resampling L2 products into a common grid with equal pixel size (latitude – longitude fixed grid). All L2 files are filtered to keep data with qa_value > 0.5, as recommended by ESA. CH4 products are resampled into a grid with a resolution of 0.01°×0.01°.

Once all CH4 L2 files are converted into L3 files with a common spatial grid, we resample the files over the time dimension to have one file per day. Due to the overlap of the orbit, some days have more than one file, and in order to keep one single file per day, we average all files belonging to the same day by using the attribute ‘time_coverage_start’ as time reference, achieving product files with both common temporal and spatial grid.

3.2. Calculating Daily Mean Surface Wind Speed – Ploiesti, Romania

The method used to select the wind components data from ERA5 is by extracting the values for the cells corresponding to the coordinates of the city Ploiesti (44.8, 45.05°N) and (25.8, 26.25°E).

In this study, 100m height wind is used as it tends to be more representative of large-scale flow. The 100m u- and v- components of wind are both required to calculate wind speed. These two separate ERA5 datasets are in hourly steps. Both components could either be positive of negative, where a positive u-component indicates a flow direction of west to east (eastward wind), and a negative indicates an east to west flow (westward wind), whereas a v-component is positive for south to north flow (northward wind), and negative for north to south flow (southward wind). However, since our aim is to calculate wind speed, not direction, absolute values of both u- and v- components are used to find mean values. Both u- and v-components of wind are averaged over 5 time-steps, that correspond to the overpass time of S5P, in order to determine the mean wind speed during the overpass. Mean wind speed (ws) is calculated as follows:

$$\boldsymbol{ws} = \sqrt{(\boldsymbol{u}^2 + \boldsymbol{v}^2)}$$

where u and v are daily mean u- and v- wind components, respectively.

3.3. NOAA HYSPLIT Model

The NOAA Hybrid Single-Particle Lagrangian Integrated Trajecroty (HYSPLIT) model was run using the Global Forecast System (GFS) model at 0.25-degree grid resolution as the meteorological input. It was run three times:

1. HYSPLIT model was run every 24 hours starting from 0900 Universal Time Coordinated (UTC) 25 March 2021 till 0900 UTC 29 March 2021, for a total run time of 3 hours and for 5 trajectories. This was repeated from 0900 UTC 26 September 2021 till 0900 UTC 30 September 2021, for a total run time of 4 hours and for 5 trajectories. Single-particle forward trajectories were initialized from the center of Ploiesti at height 100 m Above Ground Level (AGL).
2. HYSPLIT model was run starting from 0800 UTC 30 July 2021, for total run time of 3 hours, where particles were initialized at 10 m and 100 m AGL, with three starting locations with the coordinates (44.97786°N, 23.73396°E), (44.98455°N, 23.74302°E) and (44.98855°N, 23.73890°E).

4. Results and Discussion

4.1. Sources of Error

Given that Sentinel-5P is rather a newly launched satellite and has been in operation only since October 2017, several algorithm changes and improvements have been applied to the TROPOMI CH4 retrievals, including an increase in surface altitude resolution. Additionally, it was found that TROPOMI measurements are highly sensitive to high and low land surface albedo, and recently a correction for high and low albedo has been implemented. However, the corrected albedo product was not used in this study as it was made available towards the end of the study period.

4.2. TROPOMI Yearly Mean CH4 1 Jan 2019 to 31 Dec 2021 – Romania

The yearly averages (2019, 2020 and 2021) CH4 column average mixing ratio (ppbv) over Romania for the 3-year period extending from 1 January 2019 to 31 December 2021 are shown in figure 2, respectively.

Romania is one of the European countries that The Carpathian Mountain range passes through. This range of mountains forms an arc across Central Europe, where more than half of it lies in Romania. The trend of these mountains is clearly seen in figure 2, as almost no CH4 retrievals are present over the range, creating an arc-shaped gap within the CH4 maps. This TROPOMI data gap caused by topographic effects was also found in a study conducted in the United States (A. de Gouw et al., 2020).

Initially, we can notice that the region that shows the highest average CH4 column average mixing ratio is the southern part of the country. However, we can also see several enhancements in the center and northeast of the country. We can also see an overall increase of CH4 concentrations from year to year.

Figure 3 show the main oil- and gas- exploration regions in Romania. These regions are distributed mainly amongst OMV-Petrom, which is the main oil production company in Romania, and Romgaz, which is the largest producer and main supplier of natural gas in Romania. OMV-Petrom regions are highlighted in dark blue and are found in the southern part of the country, whereas Romgaz regions are highlighted in green, and are located mainly in the center and northeast of Romania. In 2017, it was estimated that there are 13 thousand ‘active’ wells spread across 400 oil fields in Romania. Both oil and gas production are responsible for methane emissions and leaks during the supply chain process. But since methane (CH4) is the primary component of natural gas, less methane emissions are expected during gas production than oil production (CATF, 2021). In figure 2, we can see that the south of Romania, where most of the oil fields are located, shows higher levels of CH4 concentrations, compared to the areas of natural gas production in the center and northeast of the country, for all three years. It is important to note that TROPOMI CH4 column average mixing ratio retrievals include all sources of methane, whether is it anthropogenic sources, including agriculture and oil- and gas-production, as well as non-anthropogenic (natural) sources, such wetlands and freshwaters, as mentioned in section 1.1.

Up until 14 Nov 2021, TROPOMI CH4 retrievals were limited to above-land only. However, recently an algorithm update, processor version 2.3.1, which started operating from 14 Nov 2021, now includes measurements over the ocean under sun glint condition with pixels classified with qa_value > 0.5 (Apituley et al., 2017). Due to the irregularity and unevenness of ocean surfaces, the sunlight bouncing back from the water surface tends to be scattered in multiple directions, causing blurry streaks of light in the data. Retrievals over the ocean are possible under the sun glint condition, where the sunlight reflects off the ocean surface at the same angle in which the sensor on the satellite views it (Copernicus: Sentinel-5P (Precursor - Atmospheric Monitoring Mission), 2012).

In the 2019 and 2020 average TROPOMI CH4 column average mixing ratio figures, we can see that there is not a single pixel over the ocean, as this was prior to the implementation of retrievals over ocean sun glint areas. However, in the 2021 average TROPOMI CH4 column average mixing ratio figure, we can see few retrievals over water along the coastline of Romania and its neighboring countries.

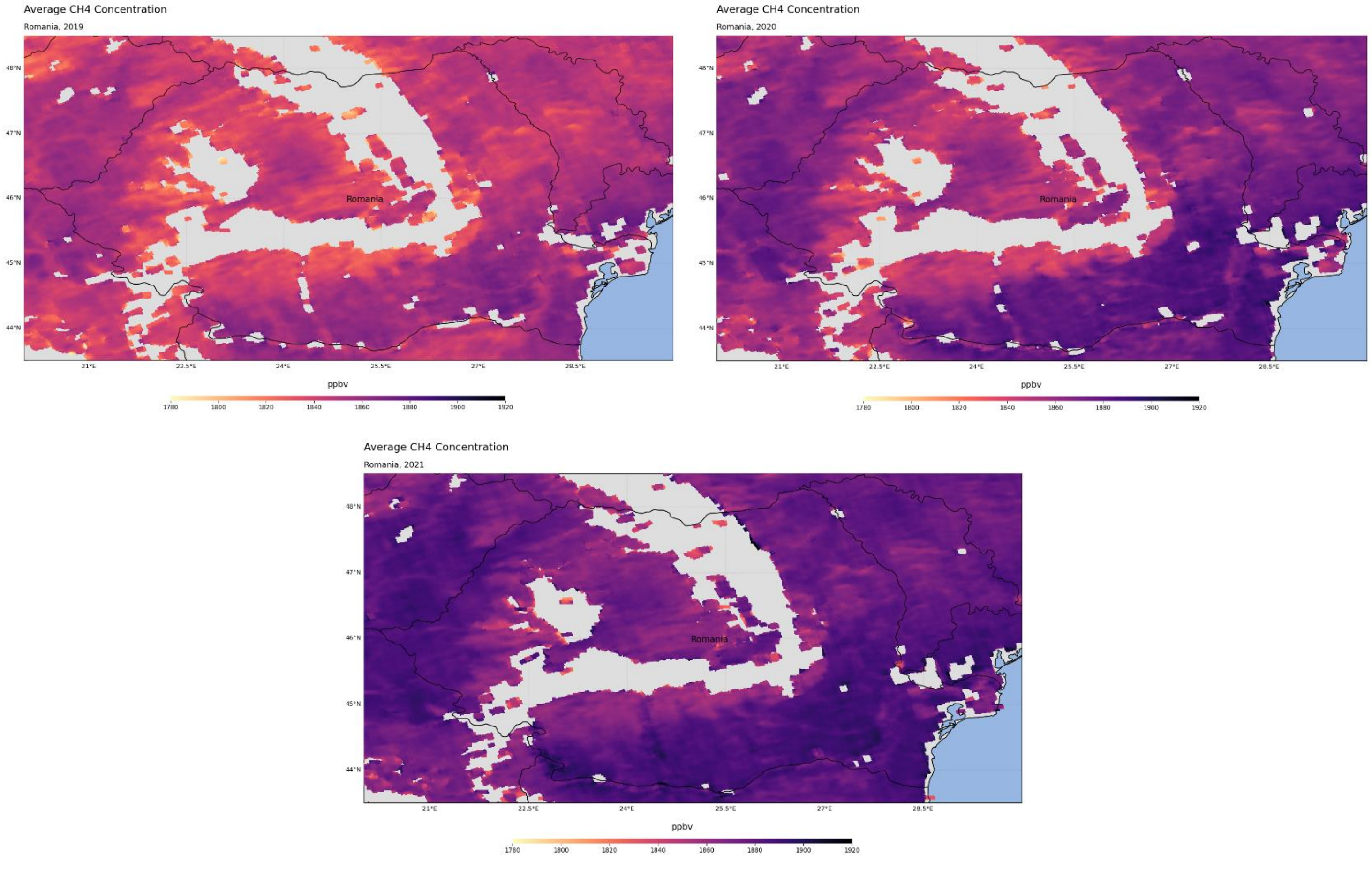


Figure 2: Yearly average TROPOMI CH4 column average mixing ratio (ppbv) over Romania for the year 2019, 2020 and 2021.

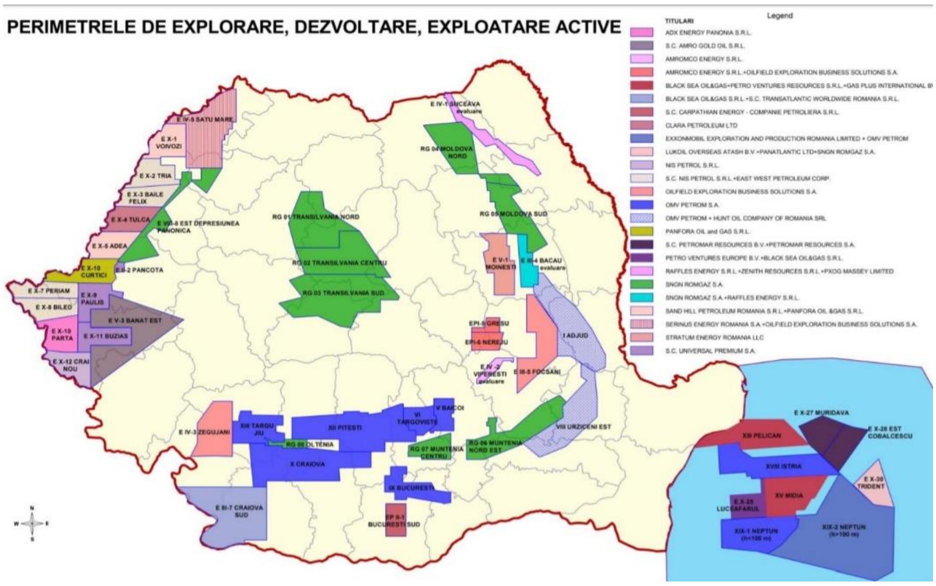


Figure 3: A map of Romania showing the main oil- and gas- exploration regions. Dark blue highlights the regions of OMV-Petrom, and green highlights regions or Romgaz, which are the main oil production company and the main producer and supplier of natural gas in Romania, respectively (Vasalca, 2019).

4.3. Methane Weekly Mean Time Series - Romania

During this 3-year period, a total of 1096 days, only 76.7% (841 days) of the days had CH4 column average mixing ratio retrievals, whereas 23.3% (255 days) of the days showed no retrievals at all over Romania. The months December and January of each year showed the lowest number of days with retrievals, whereas July and August showed the highest.

Working with TROPOMI CH4 retrievals, it is quite evident that the lack of valid CH4 pixels creates a huge limitation. This lack of TROPOMI CH4 retrievals is not only limited to Romania, as three other studies, so far, conducted in the US and Australia have also faced this limitation (Cosman, 2021, A. de Gouw et al., 2020, Sadavarte et al., 2021). One of the three studies was focused on the Permian Basin in the US, and it was found that CH4 are limited, where out of study periods ranging from 7 to 40 days, only 2 to 15 were found to have CH4 observations (Cosman, 2021).

Figure 4 shows the time series of weekly average CH4 column average mixing ratio (ppbv) over Romania for the period 1 January 2019 to 31 December 2021. An upward trend is noticed throughout the years. Weekly CH4 averages ranged between 1831 ppbv and 1896 ppbv. The highest weekly average of 1896 ppbv is recorded on week 148, which corresponds to the last week of October 2021 (25 – 31 Oct 2021). On the other hand, the lowest weekly average of 1831 ppbv was on week 28, corresponding to the second week of July 2019 (8 – 14 July 2019). Two gaps could be seen in the time series, these represent the two weeks during the 3-year period in which not a single measurement was available. These are weeks 55 (13 – 19 Jan 2020) and 103 (14 – 20 Dec 2020).

Due to a couple of uncertainties and limitations, like algorithm changes, the large spatial and temporal gaps in CH4 measurements, and uncorrected albedo CH4 retrievals, it would not make sense to evaluate temporal trends of the 3-year study period. A careful analysis of the frequency and spatial consistency of the satellite observations must be done before attempting to determine data trends.

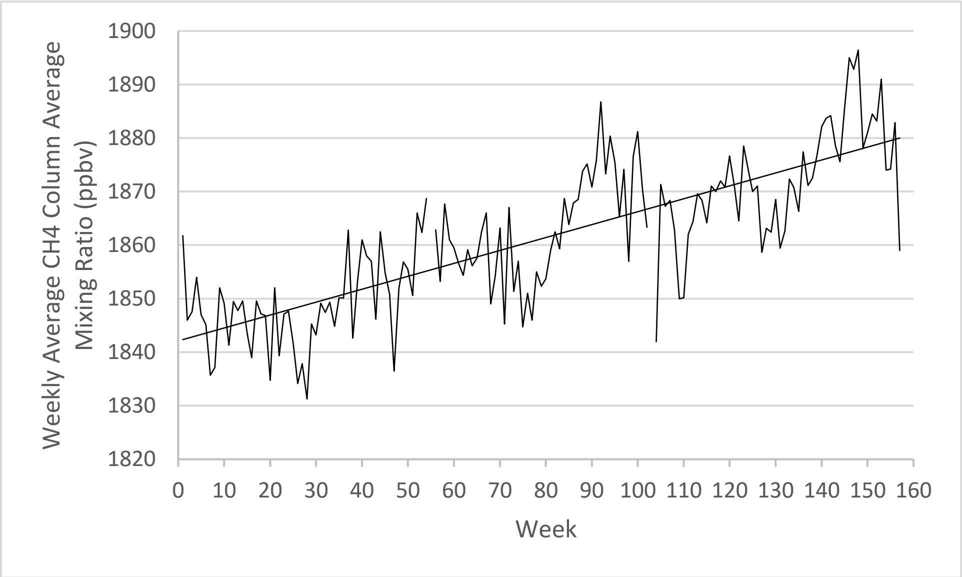


Figure 4: Time series of weekly average CH4 column average mixing ratio (ppbv) for the period 1 Jan 2019 to 31 Dec 2021.

4.4. *Methane Measurements vs. Wind Speed – Ploiesti, Romania*

The city Ploiesti has an active oil- and gas- activities, as it contains wells, storages, processing sites as well as refineries. As mentioned earlier, a study conducted in Romania in 2019 measuring street-level CH4 emissions, has reported a total of 76 locations that are considered as locations with CH4 leaks in the urban city Ploiesti (street level).

It is noticed that as the region of focus gets smaller, CH4 retrievals decrease as well. CH4 column average mixing ratio retrievals over Ploiesti, Romania's historical city of oil production, showed large temporal gaps. Out of the 1096 days (3-year period), a total of 959 days showed no measurements at all, meaning only 137 days had CH4 retrievals over Ploiesti. Daily CH4 column average mixing ratio over Ploiesti ranged between 1820 and 1917 ppbv.

From the ERA5 hourly wind dataset, the mean wind velocity during hours of S5P overpass ranged between 0 m/s and 10.1 m/s. Wind activities affect any gas emission, including CH4 emissions. Strong wind activities tend to rapidly disperse any plume or emission, leading to the dilution of CH4 and lower concentration levels, which can reduce the probability of a plume being detected or measured (Ialongo et al., 2020). However, low wind activities are needed in order for a methane plume to be developed, therefore weak winds are beneficial for plume detection (Føllesdal Brown, 2020, Jacob et al., 2022).

Figure 5 shows a scatter plot between daily CH4 column average mixing ratio (ppbv) and mean wind speed (m/s) calculated over Ploiesti for the period 1 January 2019 to 31 December 2021. CH4 and wind speed did not show a strong correlation, where they were correlated with r = - 0.32 for the days with available TROPOMI data. However, it is worth noticing that 89% of the days with available CH4 measurements fall under wind speeds of equal to or less than 5 m/s, and 50% fall under wind speeds of equal to or less than 2 m/s, and that the highest three CH4 measurements (1917, 1913 and 1903 ppbv) fall within wind speed range of 0.1 m/s and 1.4 m/s.

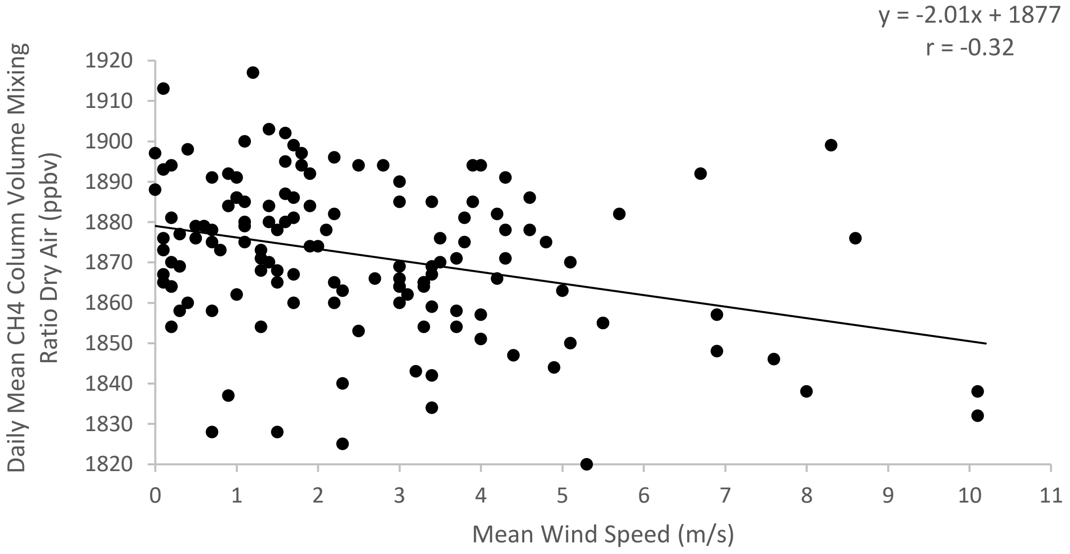


Figure 5: Scatter plot between daily CH4 column average mixing ratio (ppbv) and mean wind speed (m/s) calculated over Ploiesti for the 137 days with CH4 retrievals within the period 1 January 2019 to 31 December 2021.

CH4 measurement on days that witnessed wind speeds of larger than 5 m/s ranged between 1820 ppbv and 1899 ppbv. High CH4 values on these days are unexpected, as stronger wind dilutes the CH4 signal, however, this could be due to the interference of other neighboring CH4 emission sources. Sources of methane emission or leak within or surrounding Ploiesti must be determined in order to confirm or disprove the latter.

4.5. Methane Retrievals Averaged According to Wind Direction – Ploiesti, Romania

For the 137 days that showed CH4 retrieval over Ploiesti, the corresponding hourly 100m wind u-component of the overpass hours of TROPOMI were used to determine the days that had either westward or eastward wind, since the u-component of wind represents the eastward component. The only days that are considered here are those where all hours corresponding to the overpass hours of TROPOMI showed either only westward wind or only eastward wind. Days that had both are eliminated. Due to the limited CH4 retrieval days over Ploiesti, the available dataset was only categorized under westward and eastward wind, as adding other categories, such as northward and southward wind, will diminish the already scarce dataset even more. A total of 59 days were categorized as days with westerly wind, 48 days were listed as easterly wind, and 30 days were eliminated.

4.5.1. Yearly (2019, 2020 and 2021)

TROPOMI CH4 retrievals for days with westerly wind and days with easterly wind for 2019, 2020 and 2021 were averaged separately. A total of six figures were produced as seen in figure 6; three westerly wind figures (left column) and three easterly wind figures (right column).

Averaging available CH4 observations over wind direction (westward or eastward) allows us to analyze and confirm whether methane plumes travel with the wind and whether they are detectable downwind.

From the start we can notice that CH4 column average mixing ratio over Ploiesti and its surroundings show an overall increase throughout the years. Focusing on the CH4 retrievals averaged over westerly wind (left column), which is the movement of wind from west to east, for all three years separately, we can indeed see clear CH4 enhancements towards the right of the plots (downwind), as shown by the white borders. Interestingly, in the 2019 plot (top left), we notice that the strongest enhancement is actually found northwest of Ploiesti, which is opposite the wind direction, and CH4 levels reach up to

1890 ppbv, whereas the CH4 measurements are around 1870 ppbv downwind. Moving on to CH4 retrievals averaged over easterly wind (right column), where wind movement is from the east to the west, we can also see clear enhancements in the direction of the wind as indicated by the white borders left of the plots.

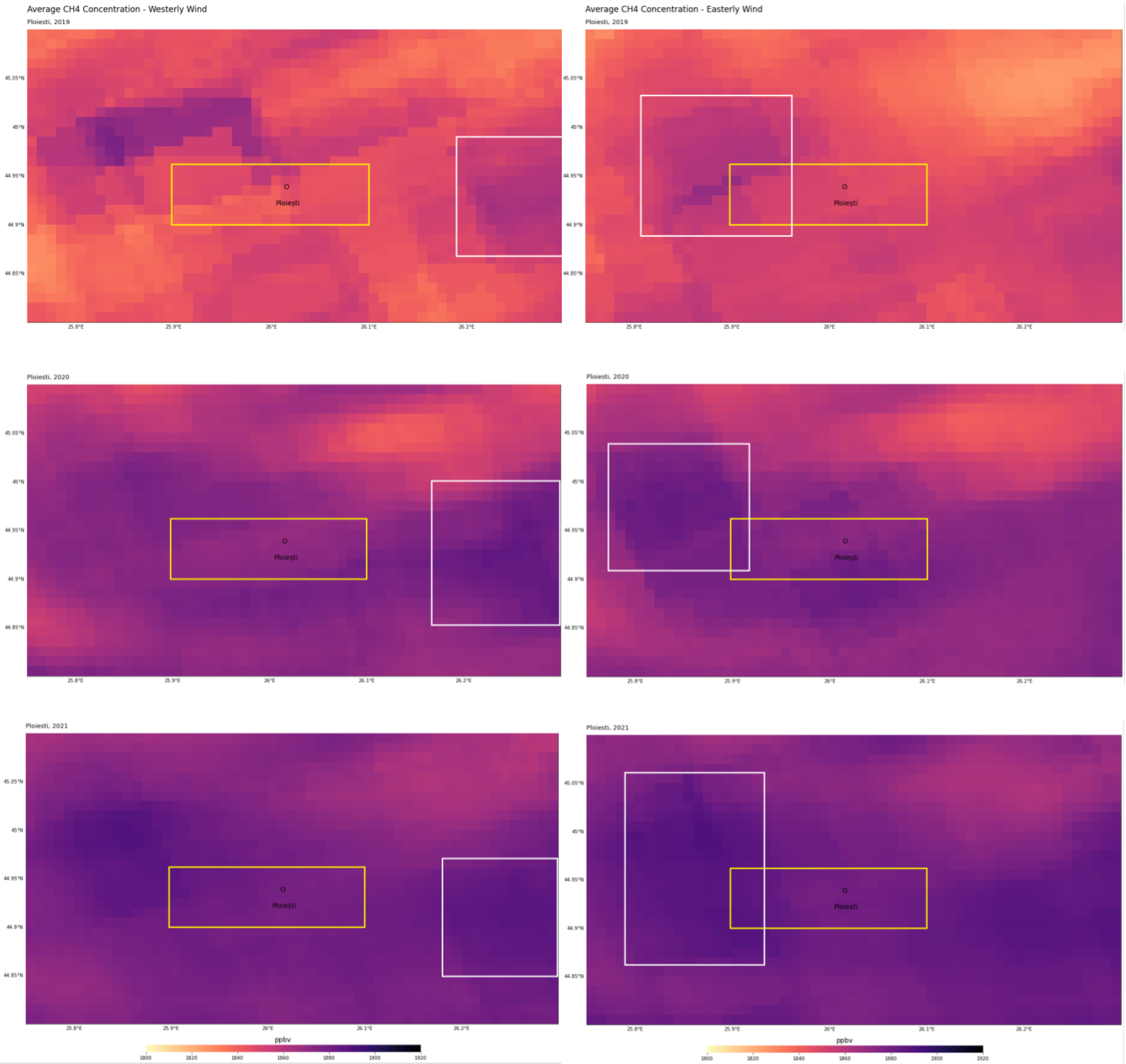


Figure 6: TROPOMI CH4 column average mixing ratio retrievals (ppbv) averaged over wind direction, westerly wind (left column) and easterly wind (right column) for the years 2019, 2020 and 2021. The circle indicated the center of Ploiesti, Romania. The yellow rectangle indicates the bounds of the city. White squares highlight the areas with CH4 enhancements downwind.

CH4 plumes or enhancements are found to be closer to the city (overlaps with city borders) for the days with easterly wind, whereas further from the city for days with westerly wind. This could be related back to wind speed measurements, where the days categorized as days with westward wind showed a mean wind speed of 3.4 m/s, with 11 days out of 59 witnessing wind speeds equal to or larger than 5 m/s. Whereas for the days categorized as days with eastward wind, the mean wind speed is 2.7 m/s, with only 4 days that had recorded wind speeds of equal to or greater than 5 m/s.

In the three plots of westerly wind (2019, 2020 and 2021), and the 2020 and 2021 plots of easterly wind, we cannot only see CH4 enhancements downwind, but also some that are located in the opposite direction. This is likely explained by the interference of other CH4 sources outside of Ploiesti, however, without the knowledge of the location of every single CH4 source and the wind direction in the areas around the city, this cannot be further explored.

When analyzing long time periods, it is hard to be certain of the effect weather systems, including wind direction and speed, have on TROPOMI atmospheric CH4 measurements, as variations in satellite measurements caused by changes in CH4 emissions, seasonal variations in CH4 and other factors are hard to be avoided and dismissed. Additionally, changes in TROPOMI's algorithms and the limited availability of CH4 retrievals could also have impacts on averages for long period (Cosman, 2021).

4.5.2. Five-Consecutive Days of CH4 Retrievals According to Wind Direction

As mentioned above, TROPOMI CH4 retrievals can be affected by several factors including variations in seasonal CH4 concentrations, as well as changes in CH4 emissions due to economic factors or other. By assessing consecutive short time periods that have the same wind categories, the impact of the latter on TROPOMI retrievals could be determined clearer as CH4 seasonal variations and ground CH4 emissions could be ignored to some extent over short time periods. For this chapter, we initially intended to choose four cases to explore:

- Two cases of five consecutive days of westerly wind during the overtime pass of TROPOMI, where one case has mean wind speed of larger the 5 m/s, and the other has a daily mean wind speed of 5 m/s or below during TROPOMI's overpass hours.
- The other two cases are the same as the first two cases, but for days of easterly wind instead of westerly wind.

However, due to the limited number of days with CH4 retrievals over Ploiesti, the availability of five consecutive days meeting the required wind description and having CH4 observations at the same time were very scarce. Therefore, we were obliged to reduce the four cases down to only two cases:

- First case: Five consecutive days of westerly wind, with wind speeds above 5 m/s during TROPOMI's overpass time.
- Second case: Five consecutive days of easterly wind, with wind speeds of 5 m/s or below during TROPOMI's overpass time.

The first case starts on 25 March 2021 to 29 March 2021. CH4 retrievals of these five consecutive days, with westerly wind and mean wind speed of 5.7 m/s, were averaged. Wind speed and CH4 retrievals of the second day (26 March 2021) were eliminated, as wind speed on this day did not meet the criteria and was less than 5 m/s. But since the dataset is limited and there is no other case with five consecutive days as required, an exception was made.

NOAA HYSPLIT model trajectories were conducted using 0.25-degree GFS model output. The trajectories were initialized every 24 hours starting from 0900 UTC 25 March 2021 till 0900 UTC 29 March 2021 for a total run time of 3 hours. This produced 5 trajectories starting at a height of 100m above ground level (AGL) from the center of Ploiesti, where each trajectory represents air flow during the overpass time of TROPOMI.

The second short term period lasted from 26 September 2021 till 30 September 2021. These five consecutive days showed easterly wind activity with mean wind speed of 3.5 m/s, and CH4 observations were averaged over the time period. Just as the first case, HYSPLIT model trajectories were also conducted in the same matter, but for a total run time of 4 hours instead, starting from 0900 UTC 26 September 2021 and ending on 0900 UTC 30 September 2021. It is important to take notice of the different run times between the two cases, as the total TROPOMI overpass hours for the five days of the first case were 3 hours, whereas for the second case it was for 4 hours.

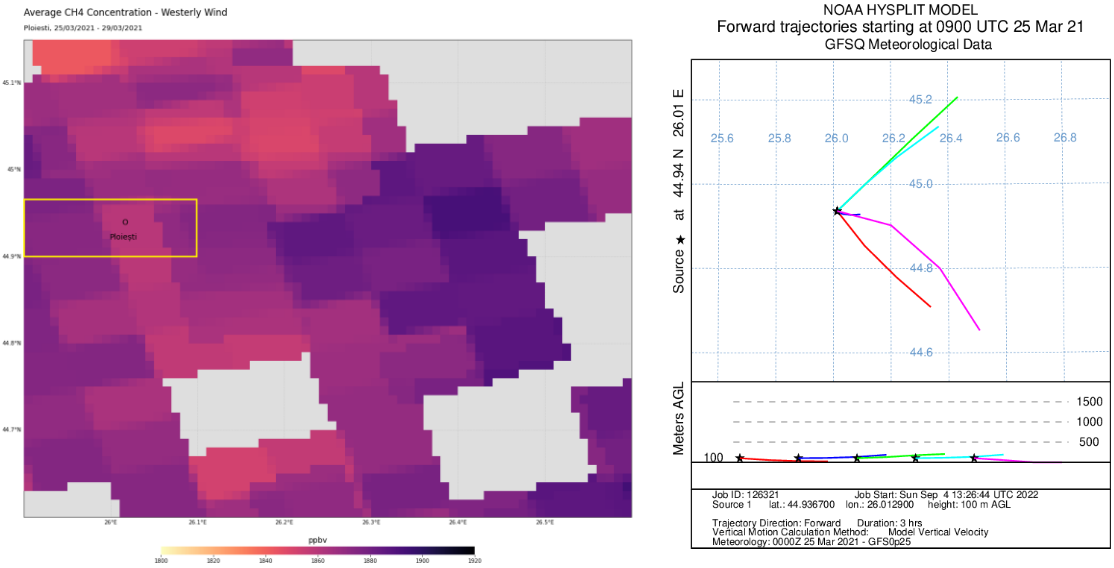


Figure 7: (Left) Average TROPOMI CH4 column average mixing ratio (ppbv) over Ploiesti (yellow border) and its surroundings downwind, for 25 March 2021 to 29 March 2021, categorized as westerly wind with speed of larger than 5 m/s. (Right) NOAA HYSPLIT model trajectories initialized every 24 hours, beginning from 0900 UTC 25 March 2021, for a total run time of 3 hours. Particles are started from the center of Ploiesti (black star) at 100 m AGL. Each colored trajectory represents the air flow during TROPOMI's overpass time for each day (5 days).

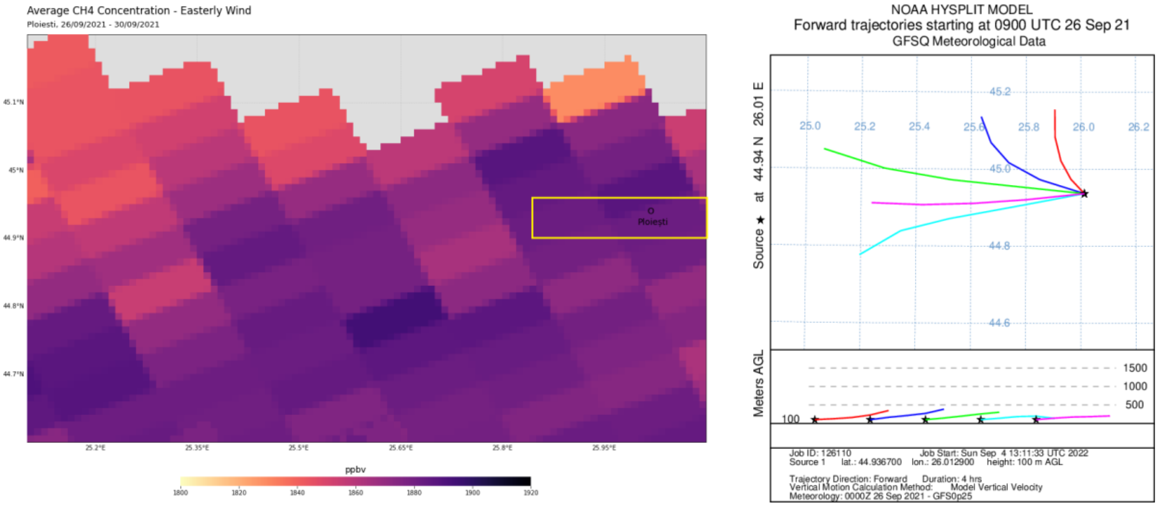


Figure 8: (Left) Average TROPOMI CH4 column average mixing ratio (ppbv) over Ploiesti (yellow border) and its surroundings downwind, for 26 September 2021 to 30 September 2021, categorized as easterly wind with speed of 5 m/s or below. (Right) NOAA HYSPLIT model trajectories initialized every 24 hours, beginning from 0900 UTC 26 September 2021, for a total run time of 4 hours. Particles are started from the center of Ploiesti (black star) at 100 m AGL. Each colored trajectory represents the air flow during TROPOMI's overpass time for each day (5 days).

In the HYSPLIT model, the red, blue, green, sky-blue and purple trajectories are the trajectories of air flow for the five days, respectively. During the time period of the first case (figure 7), the fifth day had the strongest wind activity of 6.9 m/s, and this is mirrored in the HYSPLIT model (purple). Comparing the wind trajectory of the fifth day with the five-day average CH4 retrievals (left), we can see CH4 enhancements along the direction of the trajectory, and a lack of CH4 retrievals towards the end of it. The lack of CH4 retrievals (left) towards the end of the wind trajectory (right) could also be noticed for the third (green) and fourth day (sky-blue).

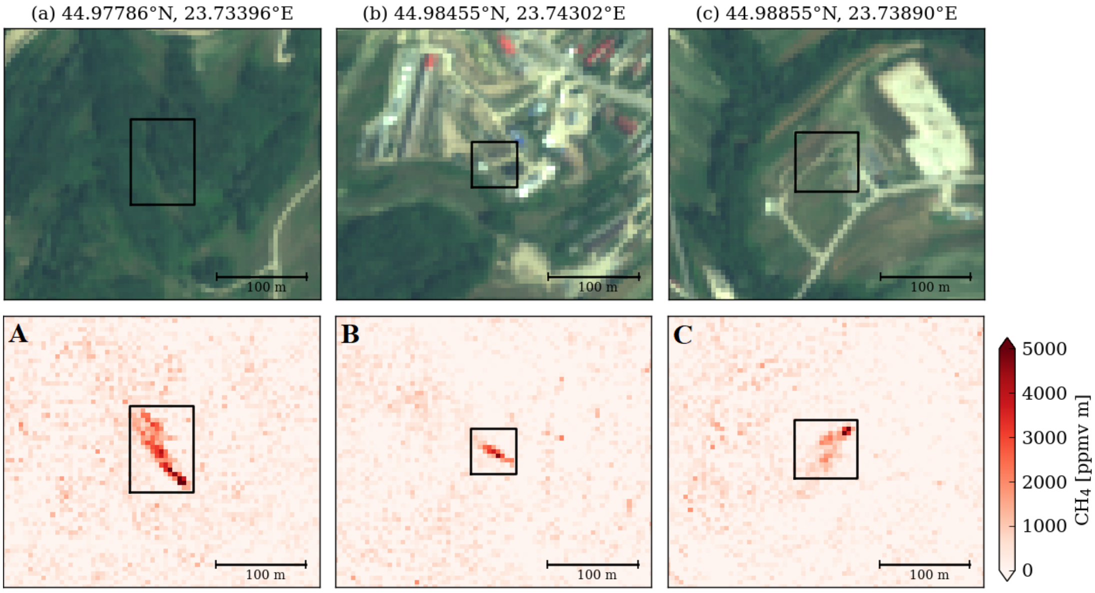
For the second case (figure 8), the third day has the highest wind speed of 4.9 m/s, whereas the first day showed the lowest wind speed of 2.2 m/s. This could also be mirrored with their HYSPLIT (right) wind trajectory (green and red trajectories, respectively). For the first two days of this case, we notice a lack of CH4 retrievals down the end of their wind trajectories in the HYSPLIT model output (right), however, we can still see CH4 enhancements along the direction of the trajectories, represented by the red and blue trajectories, respectively.

Comparing the two average TROPOMI CH4 column average mixing ratio retrievals of the two cases, we can see that in the first case, which has wind speeds larger than 5 m/s, shows lower CH4 concentrations over Ploiesti (yellow border) compared to the second case which has wind speeds of 5 m/s or below. Here we can see a clear indication that stronger wind activities effects CH4 retrievals. The study conducted in the US, focusing on TROPOMI CH4 retrievals over the Permian Basin (Cosman, 2021), has explored the difference in CH4 measurements and retrievals related to wind speed as well. They also found that CH4 measurements are higher for days with lower wind speeds compared to those with higher wind speeds. Additionally, they noticed a drop in CH4 retrievals over the Basin during days with higher wind speeds, however, this was not seen in our two cases over Ploiesti. This could be due to the difference in wind speed ranges between our cases and theirs, where in our first case, which is the one with higher wind speeds, wind activities ranged between 5.1 m/s and 6.9 m/s, whereas in their study, stronger wind ranged between 12 m/s and 16 m/s, and it is established that strong wind activities could lead to the dilution of atmospheric CH4, hence decreasing the probability of its detection. Another reason is that the focus area of our cases (Ploiesti) is much smaller in size compared to the Permian Basin.

4.6. *Comparison Between AVIRIS-NG CH4 Measurements and TROPOMI CH4 Column Average Mixing Ratio Retrievals*

The processing of a single measurement stripe from the AVIRIS-NG flight in Europe in summer 2021 showed the presence of three neighboring CH4 point sources with strong methane emissions. These sources of CH4 emissions and their locations are shown in figure 9. The three CH4 plumes seen in figure 9 (bottom row) were measured on 30 July 2021 at around 0830 UTC.

By plotting TROPMI CH4 measurements taken on the same day, we can see if these plumes are detectable by TROPOMI. Only one single file of CH4 product contained CH4 column average mixing ratio retrievals on 30 July 2021 over the locations of the three detected plumes. The TROPOMI overpass time of this single file start and end at exactly 0932 UTC and 1113 UTC, respectively. This is shown in figure 10, where the exact location of each source of CH4 is marked (x) and labeled (A, B or C) in correlation to labels in figure 9 (bottom row). In figure 10, we see no CH4 enhancement over the exact locations of the plumes. However, we can see clear CH4 enhancements northwest of the locations.


(a) 44.97786°N, 23.73396°E

(b) 44.98455°N, 23.74302°E

(c) 44.98855°N, 23.73890°E

Figure 9: The three sources of CH4 emissions retrieved from a single measurement stripe from the AVIRIS-NG flight data. (Top row) The map locations of the three sources and their corresponding coordinates. (Bottom row) The three detected CH4 plumes on 30 July 2021 at 0830 UTC. The black border in the top and bottom rows highlight the extent of each CH4 plume.

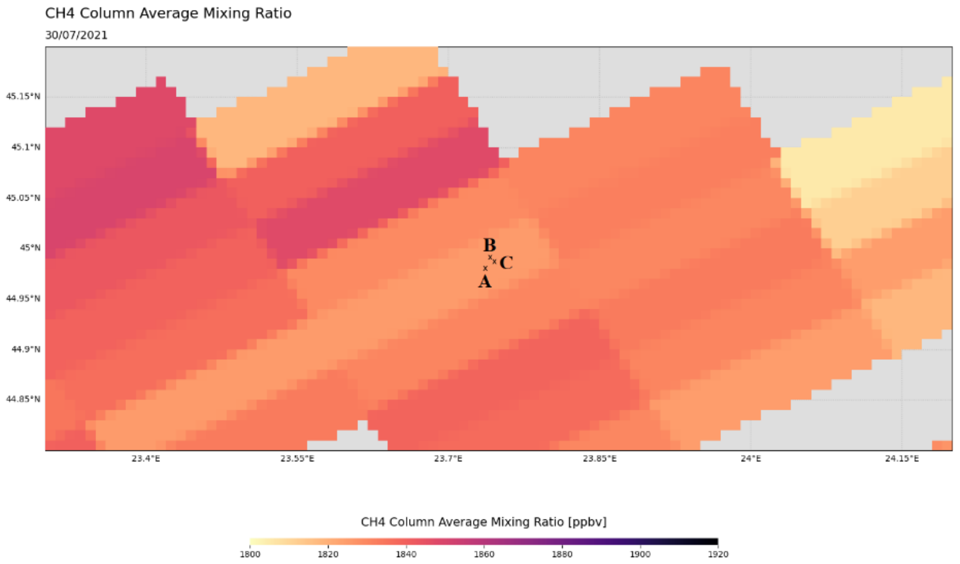


Figure 10: TROPOMI CH4 column average mixing ratio (ppbv) over the three CH4 sources on 30 July 2021. The labels A, B and C indicate the locations of the three detected plumes (figure 9).

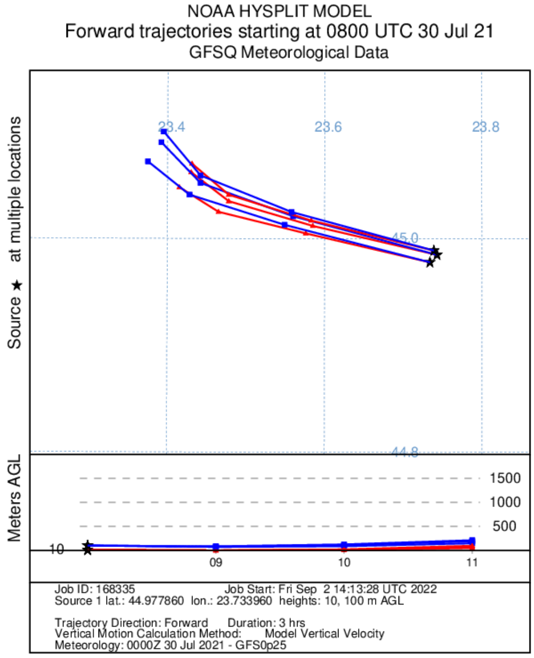


Figure 11: NOAA HYSPLIT model trajectories initialized at the three CH4 sources of emissions (black stars), starting from 0800 UTC 30 July 2021, for total run time of 3 hours, where particles were initialized at 10 m (red trajectory) and 100 m (blue trajectory) AGL.

Using HYSPLIT model, we ran wind trajectories starting from the three locations at 10 m and 100 m height AGL. The trajectories were initialized from 0800 UTC 30 July 2021 for a total running time of 3 hours, which include the overpass time of TROPOMI. Even though the overpass time of TROPOMI starts at 0932 UTC, we chose to start the trajectory at 0800 UTC since the measurement taken by AVIRIS-NG was a 0830 UTC.

The product is shown in figure 11. The three trajectories show a northwest movement of the air. Comparing CH4 retrievals (figure 10) and the HYSPLIT model trajectories (figure 11), we can clearly see a correspondence between the CH4 enhancements seen northwest of the three source locations and the three wind trajectories, as the enhancements are found downwind. And even though this is just a single example, it is still a positive indication that shows that indeed there is correspondence between TROPOMI CH4 retrievals and measurements taken at altitudes closer to the surface, at least in this case. Once more measurement strips are processed and other sources of methane emissions are determined, the correspondence between TROPOMI CH4 retrieval and AVIRIS-NG CH4 measurements could be explored more in depth.

5. Conclusions

Despite being less abundant in the atmosphere than carbon dioxide (CO2), methane (CH4) is still a crucial greenhouse gas, as it is more potent than CO2 and has higher global warming potential. The launch of ESA’s S-5P was a revolutionary achievement, as it provides accurate and high temporal and spatial resolutions of different atmospheric components, including CH4. However, it is clear that the limited availability of valid CH4 retrievals plays as a huge limitation. But hopefully with the consistent algorithm changes, and with the recent implementation of albedo correction, higher retrieval frequency and spatial coherence of TROPOMI CH4 can be delivered soon.

This study used ERA5 wind data, NOAA HYSPLIT model trajectories, a three-year (1 January 2019 – 31 December 2021) dataset of TROPOMI CH4 retrievals, and AVIRIS-NG CH4 measurements to analyze and understand the potential of wind activities to impact spatial and temporal CH4 variability.

By focusing on the different categories of wind activities and on TROPOMI CH4 retrievals over Ploiesti, and despite the extremely limited CH4 retrievals throughout the 3 years, we could still see how wind direction and speed influenced the atmospheric movement of CH4 emissions, concentrations of CH4 and availability of TROPOMI CH4 retrievals. During days of low wind speeds, CH4 measurements are found to be generally higher and CH4 retrievals are more frequent, as stronger wind speeds tend to cause dispersion of CH4 plumes or emissions, leading to the dilution and drop in concentration levels of CH4, and this restrain the probability of CH4 detectability. Not only do wind activities effect the CH4 signal and retrieval, but it also affects the travel of CH4 plumes or emissions, as CH4 enhancements are found downwind, as well as having an effect on the intensity of CH4 plumes depending on the speed of the wind.

Finally, even though Romania is considered one of the countries with the highest atmospheric methane emissions from oil and gas production in all of Europe, it is quite surprising how research and publications about methane emissions from Romania are very scarce.

References

- A. de Gouw, J., Veefkind, J., Roosenbrand, E., Dix, B., Lin, J., Landgraf, J. and Levelt, P., 2020. Daily Satellite Observations of Methane from Oil and Gas Production Regions in the United States. *Scientific Reports*, 10(1).
- Apituley, A., Pedergnana, M., Sneep, M., Veefkind, J., Loyola, D. and Hasekamp, O., 2017. Sentinel-5 precursor/TROPOMI Level 2 Product User Manual Methane. Royal Netherlands Meteorological Institute. (0.11.6). Retrieved from https://doi.org/10.5270/S5P-3p6lnwd.
- Apituley, A., Pedergnana, M., Sneep, M., Veefkind, J., Loyola, D. and Hasekamp, O., 2017. Sentinel-5 precursor/TROPOMI Level 2 Product User Manual Methane. The European Space Agency.
- Butz, André & Galli, Andre & Hasekamp, Otto & Landgraf, J. & Tol, Paul & Aben, I., 2011. TROPOMI aboard Sentinel-5 Precursor: Prospective performance of CH4 retrievals for aerosol and cirrus loaded atmospheres. Rem. Sens. Env., Sentinel-5 special issue. 120. 10.1016/j.rse.2011.05.030.
- *Clean Air Task Force (CATF)*, 2021. It Happens Here Too: Methane Pollution in Europe’s oil and gas network.
- Crosman, E., 2021. Meteorological Drivers of Permian Basin Methane Anomalies Derived from TROPOMI. Remote Sensing, [online] 13(5), p.896. Available at: <https://www.mdpi.com/2072-4292/13/5/896/htm>.
- European Environment Agency. 2019. *European Pollutant Release and Transfer Register (E-PRTR)*. Available at: <https://www.eea.europa.eu/data-and-maps/data/member-states-reporting-art-7-under-the-european-pollutant-release-and-transfer-register-e-prtr-regulation-23/european-pollutant-release-and-transfer-register-e-prtr-data-base/e-prtr_database_v17_mdb>.
- Fehr, T., 2016. Sentinel-5 Precursor Scientific Validation Implementation Plan. *European Space Agency*, (1). https://doi.org/10.5281/zenodo.165739
- Føllesdal Brown, P., 2020. Overview of methane detection and measurement technologies for offshore applications. *Carbon Limits AS*.
- Hu, H., Landgraf, J., Detmers, R., Borsdorff, T., Aan de Brugh, J., Aben, I., Butz, A., and Hasekamp, O., 2018. Toward Global Mapping of Methane With TROPOMI: First Results

and Intersatellite Comparison to GOSAT, Geophys. Res. Lett., 45, 3682–3689, https://doi.org/10.1002/2018GL077259.

- Ialongo, I., Virta, H., Eskes, H., Hovila, J. and Douros, J., 2020. Comparison of TROPOMI/Sentinel-5 Precursor NO2 observations with ground-based measurements in Helsinki. *Atmospheric Measurement Techniques*, 13(1), pp.205-218.
- Jacob, D., Varon, D., Cusworth, D., Dennison, P., Frankenberg, C., Gautam, R., Guanter, L., Kelley, J., McKeever, J., Ott, L., Poulter, B., Qu, Z., Thorpe, A., Worden, J. and Duren, R., 2022. Quantifying methane emissions from the global scale down to point sources using satellite observations of atmospheric methane. *Atmospheric Chemistry and Physics*, 22(14), pp.9617-9646.
- Landgraf, J., Lorente, A., Langerock, B. and Kumar Sha, M., 2020. *S5P Mission Performance Centre Methane [L2__CH4___] Readme*. The European Space Agency. Available at: <https://sentinel.esa.int/documents/247904/3541451/Sentinel-5P-Methane-Product-Readme-File>.
- Lopez, M. & Sherwood, Owen & Dlugokencky, E.J. & Kessler, R. & Giroux, L. & Worthy, Douglas, 2017. Isotopic signatures of anthropogenic CH 4 sources in Alberta, Canada. Atmospheric Environment. 164. 10.1016/j.atmosenv.2017.06.21.
- Menoud, M., van der Veen, C., Maazallahi, H., Fernandez, J., Korben, P., Calcan, A., France, J., Lowry, D., Schmidt, M., and Röckmann, T., 2020. Isotopic characterisation of methane emissions from oil and gas operation in Romania, EGU General Assembly, Online, 4–8 May 2020, EGU2020-13643. https://doi.org/10.5194/egusphere-egu2020-13643.
- Milkov, A.V., Schwietzke, S., Allen, G. et al., 2020. Using global isotopic data to constrain the role of shale gas production in recent increases in atmospheric methane. Sci Rep 10, 4199. https://doi.org/10.1038/s41598-020-61035-w.
- Rockmann et al., 2020 - (ROMEO - ROmanian Methane Emissions from Oil and Gas)
- Sadavarte, P., Pandey, S., Maasakkers, J., Lorente, A., Borsdorff, T., Denier van der Gon, H., Houweling, S. and Aben, I., 2021. Methane Emissions from Superemitting Coal Mines in Australia Quantified Using TROPOMI Satellite Observations. *Environmental Science & Technology*, 55(24), pp.16573-16580.
- Schneising, O., Buchwitz, M., Reuter, M., Bovensmann, H., Burrows, J., Borsdorff, T., Deutscher, N. M., Feist, D., Griffith, D. W., Hase, F., Hermans, C., Iraci, L., Kivi, R., Landgraf, J., Morino, I., Notholt, J., Petri, C., Pollard, D., Roche, S., Shiomi, K., Strong,

K., Sussmann, R., Velazco, V. A., Warneke, T., & Wunch, D., 2019. A scientific algorithm to simultaneously retrieve carbon monoxide and methane from TROPOMI onboard Sentinel-5 Precursor. Faculty of Science, Medicine and Health - Papers: Part B. https://ro.uow.edu.au/smhpapers1/1113.

- The European Space Agency. 2012. *Copernicus: Sentinel-5P (Precursor - Atmospheric Monitoring Mission)*. [online] Available at: <https://www.eoportal.org/satellite-missions/copernicus-sentinel-5p#copernicus-sentinel-5p-precursor---atmospheric-monitoring-mission>.
- Veefkind, P & Aben, I. & McMullan, K. & Förster, H. & Vries, Johan & Otter, G. & Claas, Jacques & Eskes, Henk & Dehaan, Johan & Kleipool, Q. & van Weele, M. & Hasekamp, Otto & Hoogeveen, R.W.M. & Landgraf, J. & Snel, R. & Tol, Paul & Ingmann, P. & Voors, Robert & Kruizinga, Bob & Levelt, P., 2012. 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. 120. 70-83. 10.1016/j.rse.2011.09.027.