

# Response to Interactive comment on “Systematic detection of local CH<sub>4</sub> emissions anomalies combining satellite measurements and high-resolution forecasts” from Anonymous Referee #1

This manuscript introduces a method to systematically detect local CH<sub>4</sub> anomalies by combining total column CH<sub>4</sub> satellite observations from TROPOMI with high resolution CH<sub>4</sub> forecasts produced by CAMS. The manuscript is well written but it is at the edge of the scope of ACP, because it rather demonstrates the theoretical potential of the introduced technical method than having general implications for atmospheric science as it is not analysing how well the method can be applied globally (see general comments). However, as the editor has suggested to stay in ACP (instead of moving to AMT), I recommend publication after the following comments have been addressed.

## General Comments

My main criticism of this manuscript (in the sense of a publication in ACP) is its demonstration character with a lack of general implications for atmospheric science. The advantage is that the method can in principle be applied globally. However, it is not clear if the majority of the detected global anomaly candidates are due to actual unreported or over-reported sources or due to local systematic retrieval biases (e.g. as a consequence of small-scale albedo variations). The presented analysis of the method is limited to a few local case studies (e.g. confirmation of known underreported sources) and does not include an evaluation of the global capabilities to distinguish between missed sources and retrieval biases. Although this would be sufficient for an AMT publication, the global detection statistics and the impact of retrieval biases should be investigated further if possible to better fit the scope of ACP (see also specific comments).

*We firstly thank the reviewer with the review that led to significantly improve the paper. We agree with the reviewer and the editor that the article is at the edge of the ACP scope. The goal of the article is to present the capability we newly developed using part the ECMWF IFS data assimilation system to perform this automatic detection so we were not sure if this would fit the AMT journal scope as well. We are happy to have the article moved to AMT if the editor and the reviewer think that this is more appropriate.*

*We showcase few cases studies (now 5) to show the potential of such methods but also highlight the short comings. Going into details on more case studies to provide a global analysis would significantly lengthen and change the paper. We have however added one more case study to strengthen the point on local persistent biases in response to one specific comment below (see section 4.3 and figure 14).*

*Significant restructuring of the paper would be needed with providing a global evaluation of the capabilities and this would require diagnosing the system for an extended time period (probably several years) to provide a robust analysis. This will be done in further steps as we plan to have this system running routinely soon. But this cannot be done within the time frame of this present review. We do however have changed figures to include the global detection results for few months (now in figure 9) with numbers to show how the system is performing globally. We also now mention this point in the conclusion motivating for a further global analysis once the data set will be available.*

## Specific Comments

Page 2, Lines 56-59: There are also other relevant studies, e.g. Zhang et al. (2020) or Schneising et al. (2020).

*We have added the references to the text.*

Page 4, Lines 115-117: Is it advisable for the presented method to use a model in which satellite data have already been assimilated (IASI and TANSO)? The assimilated satellite data may already correct for under- or overestimations in the emission data bases to some extent and thus complicate the interpretation.

*Our current data assimilation technique does not correct the emissions but only the concentrations. We clarify the text to make this point clearer.*

Page 5, Line 142: Why is Tours visible in Figure 5? What is the origin?

*We have added a brief explanation in the text.*

Page 6, Lines 161-163: Please give a reference for the TROPOMI averaging kernel function as a function of pressure.

*We have now added the reference in the text.*

Equation 6: What does the  $l_p$  stand for?

*We subscript  $l_p$  here was wrong as it should stand for high pass. We changed the subscript to  $h_p$  and clarified the text accordingly.*

Page 8, Line 235: Should "positive" be "negative"?

*This has been corrected.*

Page 9, Lines 252-254: You have identified two candidates, which have not been investigated or documented yet: southern Nevada and northern Baja California. You conclude that the latter may be due to local albedo properties as you were not able to identify a responsible facility. What about the former (Nevada)? Is there are a source or local albedo variations?

*This is likely the case as well for the enhancement seen at the Nevada-Arizona border. We clarified the text accordingly.*

Page 10, Line 284-287: "Consistent shapes over months" sounds longer than it actually is. Figure 12 only spans a time period of 6 weeks. The red Turkmenistan features in Figure 10 look similarly consistent over comparable periods. This exemplarily illustrates the difficulty of distinguishing between underreported sources and retrieval biases. Concerning persistent shapes over time, please also discuss the potential impact of temporally and spatially variable small-scale albedo features (e.g. due to snow).

*We now have improved the Turkmenistan figure to a more zoomed version to show that the shape of the feature is not consistent over months like in the Siberian case. Regarding the Siberian case we can only show it over a reduced span (figure 13 spans over more than 10 weeks as the window length is 30 days i.e. around 4 weeks) as the measurements are only available for a short summer period over these latitudes. To make our point stronger and clearer we have identified an additional case over Australia illustrated with figure 14 where four 30-day windows are displayed over 4 months. We also address the potential impact of temporally and spatially variable small-scale albedo features (e.g. due to snow). We clarified the text accordingly.*

Page 10, Lines 293-295: What is praised as an advantage here (does not only allow for the detection of anomalies but also has the potential of detecting local retrieval errors) is also the main problem: It is not clear if it is possible to reliably distinguish between the two cases in general and there is no global analysis performed to try to approach the answer quantitatively.

*The response to the comment above partially answers this comment. We do agree with the reviewer of the current limitation and that this issue deserves further works as already mentioned at the end of section 4.3. We now have clarified the text accordingly.*

Figure 7: Please specify the time period. Is it 2019-07?

*This has been clarified.*

Figure 8: Please highlight the plot associated to the final choice of parameters (30 days, 2\_) and describe in the caption.

*This has been clarified.*

## **Technical Corrections**

Please replace CH4 by CH<sub>4</sub> in all instances.

*Fixed*

Page 7, Line 193: Please delete "and": "... where  $d_m$  is the average departure ..."

*Fixed*

Page 10, Line 315: Please delete "to": "... emission events could occur ..."

*Fixed*

Caption of Figure 4: Is "next fluxes" correct or should it be "net fluxes"?

## Response to interactive comment on “Systematic detection of local CH<sub>4</sub> emissions anomalies combining satellite measurements and high-resolution forecasts” by Anonymous Referee #2

Received and published: 8 November 2020

The authors have developed a method of combining modeled forecasts and satellite observations of CH<sub>4</sub> to isolate local CH<sub>4</sub> anomalies that might be associated with discrepancies in anthropogenic emissions. Identifying emissions from point sources such as power plants against background CH<sub>4</sub> can be challenging. The manuscript presents a clever approach for doing so that takes advantage of the newly available high-resolution CH<sub>4</sub> retrievals from TROPOMI and the equally high-resolution CAMS forecasts. The confirmation of the previously identified CH<sub>4</sub> source in western Turkmenistan is a nice demonstration of the potential utility of the approach. I recommend the manuscript for publication in ACP after the authors have addressed my comments below.

### Main comments

1) My main concern is that the specification of the filter parameters seems to be subjective. On lines 209-211 the authors explain that specifying a length scale (sigma) of 5 degrees retains large-scale structures in the signal, whereas a length scale of 0.5 degrees results in the loss of too much signal. As a result, they selected a length scale of 2 degrees. How did they decide when too much signal is lost? It would be helpful if the authors could be more quantitative. In addition, what is meant by “large-scale” in this context? For example, the panels in Fig. 8 for sigma of 0.5, 1.0, and 2.0 for both the 10-day and 30-day windows all look similar to me. I can see the three regions of CH<sub>4</sub> enhancements in the southwestern US and Mexico in all six panels, so it is unclear why the case for sigma = 2.0 with a 30-day window is best. If there is a tacit assumption being made about what specific scales are of interest and the type of emissions that are associated with those scales, that assumption should be explicitly stated as it places a constraint in the utility of the approach.

*We thank the reviewer for this helpful comment. We agree that the submitted version of the article do not discuss quantitatively the selection of the filter parameters. We do not have however found a quantitative metric or criteria to find the optimal parameters values for the size of the convolution kernel and the length of the window. Given the answer to the comment 2) and the change to an outlier threshold now based on the measurement precision, we justify the size of the kernel as follows. If the kernel is too small, the filtered signal is getting weaker than the measurement precision and very few to no detections of anomalies is made. Conversely if the kernel is large the signal is strong but to the risk of picking up larger patterns than the targeted features, i.e. which are directly related to local emissions in the CH<sub>4</sub> atmospheric distribution. Regarding the window length, we attempted to explain in the text that a short window could fail to give enough coverage to correctly run the convolution filter. In the opposite a long window will maximise the chances to have to good coverage, but the method would lose its ability to provide temporal variability. For those reasons we chose to run the filtering with a 2 degrees kernel size over a window of 30 days. We have now detailed and clarified the text accordingly.*

2) In a similar vein, on lines 217-221 the authors stated that they selected a three sigma threshold for the outlier classification because it “provides suitable results.” It is unclear how “suitable” is defined here? They stated that a narrower range “starts to fail isolating important anomalies and conversely a wider range might fail to capture useful information.” What are the important anomalies that are not detected with the narrower range? The discussion here should be expanded to give the reader a better sense of what are the implications of this threshold for the type of anomalies that can be detected.

*We thank the reviewer for this very helpful comment. Concerning the three-sigma standard deviation threshold, we have chosen this value because it is what is commonly used to account for outliers. We however agree that this is not an objective criterion. We have then updated and improved the outlier classification and is now based on the measurement precision that is provided with the TROPOMI CH<sub>4</sub> product. When the absolute value of the filtered departure is above the precision value, we consider this as an anomaly to be displayed. We clarified and modified the text and figures of the paper accordingly.*

3) I am also concerned about the lack of discussion about the potential impact of biases. The filtering does remove “large-scale” biases, but it would be helpful if the authors could comment on the impact of spatially and temporally varying biases on smaller scales (i.e., scales between sigma and the “large-scale”). These could arise from the influence of transport errors, for example.

*We are not sure to fully understand the reviewer's concern. Features of scales above sigma (the kernel length scale) are removed by the filter. Thus the 'scales between sigma and the "large-scale"' are not impacting the results and analysis here. We clarified the text accordingly (at the end of section 2.1) by defining large-scale in the paper's context of being the combination of synoptic-scales (2000 km or more) and meso-alpha-scale (between 200 km and 2000 km).*

4) Line 269-270: Is the over-prediction in the Los Angeles area seen only in the Aug-Sept period (i.e., in the right panel)? It doesn't seem to be present in the Jun-Jul plot (left panel). If that is the case, it should be noted in the manuscript. What could be the cause of this temporal variation?

*We have now detailed the section 4.2 to take into account the reviewer's comment. We have added an additional figure (now figure 10) that covers four 30 days window over the southwestern US and northern Mexico area.*

#### **Technical comments**

1) Lines 14, 81, 108, 109, 114, 162, 170, 275: A space is needed between the numbers and units, e.g., "9 km" instead of "9km".

*Fixed*

2) Line 52: I believe the "Pandley et al." reference should be "Pandey et al."

*Fixed*

3) Line 75: Please change "earth" to "Earth".

*Fixed*

4) 4) Lines 336, 341, 343, 354, 360: The line spacing between the references is irregular.

*Fixed*

5) Figure 5: It is difficult to see the features in the plot for Europe. Since the objective here is to show that ability of the model to capture fine scale features, why not plot the European sector with a different scale to better emphasize these features?

*Fixed. We adjusted the color scale.*

6) Figure 7: It is unclear what is the time period for the data shown here.

*Fixed. We added the end of window date in the figure caption.*

7) Figure 9: It is difficult to see the yellow/gold colors for the low forecast category.

*Fixed, we have changed it to darker color scale.*



# Systematic detection of local CH<sub>4</sub> emissions anomalies combining satellite measurements and high-resolution forecasts.

Jérôme Barré<sup>1</sup>, Ilse Aben<sup>2</sup>, Anna Agustí-Panareda<sup>1</sup>, Gianpaolo Balsamo<sup>1</sup>, Nicolas Bousserez<sup>1</sup>, Peter Dueben<sup>1</sup>, Richard Engelen<sup>1</sup>, Antje Inness<sup>1</sup>, Alba Lorente<sup>2</sup>, Joe McNorton<sup>1</sup>, Vincent-Henri Peuch<sup>1</sup>, Gabor Radnoti<sup>1</sup>, Roberto Ribas<sup>1</sup>

<sup>1</sup>ECMWF, European Centre for Medium Range Weather Forecasts, Shinfield Park, Reading, United Kingdom

<sup>2</sup>SRON, Netherlands Institute for Space Research, Utrecht, Netherlands

10 *Correspondence to:* Jérôme Barré (jerome.barre@ecmwf.int)

**Abstract.** In this study we present a novel monitoring methodology to detect local CH<sub>4</sub> concentration anomalies worldwide that are related to rapidly changing anthropogenic emissions that significantly contribute to the CH<sub>4</sub> atmospheric budget. The method uses high resolution (7 km x 7 km) retrievals of total column CH<sub>4</sub> from the Tropospheric Monitoring Instrument (TROPOMI) onboard the Sentinel 5 Precursor satellite. Observations are combined with high resolution CH<sub>4</sub> forecasts (~9 km) produced by the Copernicus Atmosphere Monitoring Service (CAMS) to provide departures (observations minus forecasts) close to the native satellite resolution at appropriate time. Investigating the departures is an effective way to link satellite measurements and emission inventory data in a quantitative manner. We perform filtering on the departures to remove the synoptic-scale and meso-alpha-scale biases on both forecasts and satellite observations. We then use a simple classification on the filtered departures to detect anomalies and plumes coming from CAMS emissions that are missing (e.g. pipeline or facility leaks), under-reported<sup>\*</sup> or over-reported (e.g. depleted drilling fields). Additionally, the classification helps to detect local satellite retrieval errors due to land surface albedo issues.

## 1. Introduction

Atmospheric methane (CH<sub>4</sub>) is the second most important anthropogenic greenhouse gas after carbon dioxide and contributes significantly to changes in radiative forcing and climate change. CH<sub>4</sub> is estimated to account for at least a quarter of the present-day warming (Myhre et al., 2013) and has a near-term global warming potential that is 84 times larger than CO<sub>2</sub> per unit mass (IPCC 2013). There are numerous natural and anthropogenic CH<sub>4</sub> sources, which vary in location and areal extent. The anthropogenic emissions related such as oil and gas production and coal mining and biomass burning tends to be geographically localised, e.g. over a plant facility, a pipeline or a field of extraction. Methane emissions however related to biological fluxes such as livestock, landfills and rice fields which can also be either geographically localised over narrow areas

30 or more widespread. For example, microbial respiration in wetlands showing more extensive patterns over the globe (Sauniois et al., 2016). Atmospheric methane concentrations have more than doubled since the pre-industrial times because of the imbalance between methane sources and sinks (IPCC, 2013), due to an increase of oil and gas production, rice crops, livestock and landfills. Methane has a relatively short atmospheric lifetime (with respect to climate scales) of around 9 years, meaning targeted emission reductions could be an effective way to limit the rate of warming over the upcoming decades (Shoemaker et al., 2013).

Greenhouse gases emission inventories are generated using aggregation and extrapolation of regional and national specific data. These data are reported individually by countries using the guidelines provided by the United Nations Framework on Climate Change (UNFCCC) and the Intergovernmental Panel for Climate Change (IPCC). The reporting follows a bottom-up approach, which utilises activity data and emission factors of individual emissions sectors. Official reporting and processing of this data to build these bottom-up inventories can cause significant lag and information can be out of date for certain sectors once publicly released. This can become an issue in the context of rapidly changing emissions from large point sources, for example in the oil and gas sectors (Alvarez et al., 2018). In the case of atmospheric composition modelling, emission inventories are used for input surface fluxes to simulate atmospheric concentrations. Within the Copernicus Atmosphere Monitoring Service (CAMS) these simulations are used to provide routine real-time forecasts of greenhouse gases concentrations. The CAMS greenhouse gas forecasting system integrates satellite observations (Massart et al., 2014, 2016) to generate initial conditions for high-resolution forecasts at about 10 km (Agusti-Panareda et al., 2019). The lack of up-to-date emission inventories will impact and likely degrade simulated CH<sub>4</sub> concentrations in areas where the local contribution of anthropogenic emissions is significant.

Many studies have shown the rapidly changing and event-based nature of CH<sub>4</sub> anthropogenic emissions, especially in the case of identifying the location of 'super-emitter' point source locations. Conley et al. (2016) used aircraft measurements to characterise a blowout of a well connected to the Aliso Canyon gas storage facility in California from October 2015 to February 2016. Pandey et al. (2019) showcased detection of large methane emission from a gas well blowout in Ohio during February to March 2018 using satellite measurements. More recently, Varon et al. (2019) detected an anomalously large CH<sub>4</sub> source using a combination of satellite instruments over Central Asia (western Turkmenistan) associated with a gas compression station. Those types of suddenly occurring CH<sub>4</sub> emissions cannot be or are not reported/detected in time to be included in the bottom-up inventories but are seen from space. Other studies showed the capability of satellite measurements to detect CH<sub>4</sub> emissions related to extensive drilling and fracking areas. Kort et al. (2014) identified a large methane anomaly over the Four Corners region of the USA and more recently de Gouw et al. (2020), Zhang et al. (2020) and Schneising et al. (2020) showed satellite detection of large and extended enhancements in the San Juan, Uintah and Permian basin in the USA. While these satellite-based studies focused on specific events and locations, none of them systematically detected such anomalies at global scale, nor did they provide a method to do so.

Systematic detection of large point sources of anthropogenic CH<sub>4</sub> emissions using a combination of satellite observations and modelling could enable rapid action to reduce emissions from the oil and gas sectors. Two recent developments allow for

systematic detection of unreported CH<sub>4</sub> atmospheric anomalies linked to small scale and point sources emissions. Firstly, newly available high resolution (7 km x 7 km) satellite observations from the Tropospheric Monitoring Instrument (TROPOMI, Veefkind et al., 2012) on board the Sentinel-5p platform. Secondly, improved real-time forecasting at high-resolution (~9 km) provided by CAMS (Agusti-Panareda et al., 2019). In this paper we present a novel methodology to routinely compare the satellite observations with the model forecasts in order to perform a systematic detection of atmospheric CH<sub>4</sub> anomalies related to emission changes from small scale and point sources emissions that are not reported or lack timely update. The paper is organised as follows: Section 2 describes the setup that includes the TROPOMI observations, the forecasting and monitoring configurations, Section 3 presents the detection method, Section 4 discusses several case studies followed by conclusions where we discuss briefly the benefit of our approach with coarse resolution inverse modelling.

## 2. Setup

### 2.1. TROPOMI CH<sub>4</sub> observations

The TROPOMI (Veefkind et al., 2012) instrument was launched 13 October 2017 onboard the Sentinel-5 Precursor satellite, a low Earth orbiter with a Sun-synchronous orbit that overpasses at 13:30 local solar time. Currently operational since the end of April 2018, the instrument is an imaging spectrometer with a wide spectral range: ultraviolet, visible, near-infrared and shortwave infrared. This allows TROPOMI to measure a variety of atmospheric chemical species such as: ozone, nitrogen dioxide, carbon monoxide, sulphur dioxide, formaldehyde, aerosol and methane (Hu et al., 2018). Current CH<sub>4</sub> observations, which are available for the inner two thirds of the swath and only over land, are vertically integrated columns sensitive to the troposphere (surface to 200 hPa). With a swath of around 1,750 km (normally 2,600 km) wide from the along track position and a ground pixel size of 7 km x 7 km, TROPOMI CH<sub>4</sub> data can provide near global daily coverage at high horizontal resolution over land but is limited by cloud cover and retrieval quality. In this study, we use the bias corrected version of the product and we apply the most stringent quality flagging possible, selecting only pixels that have the *qa\_value* = 1.0 (see Product Readme Methane V01.03.02, <https://sentinel.esa.int/documents/247904/3541451/Sentinel-5P-Methane-Product-Readme-File>). In the later document it is stated that the overall the instrument precision is less than or equal to 2.5% of the total column which is within the mission requirements for both random error and bias (1.0% and 1.5% respectively). Figure 1 illustrates the CH<sub>4</sub> satellite observation coverage that TROPOMI provides over a year, a month and a day.

The measurements show clear geographical variation of the CH<sub>4</sub> column-averaged dry-air mixing ratios (XCH<sub>4</sub>) that are driven by the atmospheric transport but most importantly by the spatial and temporal variability of the surface fluxes and emissions variations. Figure 2 shows the 2019 annual average zoomed over the Middle East region and the western USA regions. Over these regions, spatial variability results in XCH<sub>4</sub> enhancements of up to 50 ppb over emission hotspots. Differences in the average concentrations from region to region are also significant, from approximately 1825 ppb over the USA to 1875 ppb over the Middle East. The strong local enhancements are an indication of strong local surface fluxes and emissions of CH<sub>4</sub> from oil and gas activities, mining, agriculture or wetlands. XCH<sub>4</sub> retrievals can also be prone to some

systematic residual errors especially related to surface albedo (Hasekamp et al., 2019). De Gouw et al. (2020) for instance mentioned the possibility of retrieval biases due to low surface albedo in the short wave infrared spectral bands in the winter. Such retrieval biases, even though mostly reduced by the bias corrected product, need further investigation and are outside the scope of this paper. Nevertheless, the TROPOMI data are sufficiently accurate to show local enhancements linked (but not limited) to oil and gas production. We show in section 3 how to isolate these small-scale signals of interest and how to remove the contribution of synoptic-scale (more than 2000 km) and meso-alpha-scale (between 2000 km and 200 km) biases. In the rest of the paper we define large-scale as the combination of synoptic-scale and meso-alpha-scale.

## 2.2. CAMS high-resolution CH<sub>4</sub> forecasting suite

In this study we use the ECMWF Integrated Forecasting System (IFS), which is used in different configurations for the operational Numerical Weather Prediction (NWP) system as well as for the Copernicus Atmosphere Monitoring Service (CAMS) atmospheric composition analyses and forecasts (e.g. Flemming et al, 2015). As part of the CAMS greenhouse gases services, the IFS is used to provide 5 days CO<sub>2</sub> and CH<sub>4</sub> forecasts (Agustí-Panareda et al., 2019) jointly with other species relevant for air-quality (Flemming et al., 2015).

The IFS model cycle used in this paper is CY45R1 and is run routinely with a TCo1279 horizontal resolution which is a cubic octahedral reduced Gaussian grid at approximately 9 km (Holm et al., 2016) with 137 vertical levels from the surface to 0.01hPa. Details about the transport and meteorological configuration can be found in Agustí-Panareda et al. (2019). The CAMS greenhouse gases (GHG) operational suite is composed of an analysis and forecasts at medium and high resolution (see Fig. 3). The analysis is based on the IFS 4D-Var assimilation system which was adapted to assimilate retrieved column-averaged mole fractions of CO<sub>2</sub> and CH<sub>4</sub> together with all the operational meteorological observations (Engelen et al., 2009, Massart et al., 2014, 2016). The analyses are produced every 12hours (00:00UTC and 12:00UTC). A 4-day forecast is then issued daily after the 00:00UTC analysis on a TCo399, a cubic octahedral grid corresponding to approximately 25 km x 25 km with the same 137 model level configuration. Two satellite observation streams are currently assimilated, the Infrared Atmospheric Sounding Interferometer (IASI) for CH<sub>4</sub> on the MetOp satellites and the Thermal And Near-infrared Sensor for carbon Observations (TANSO) on the GOSAT satellite for both CO<sub>2</sub> and CH<sub>4</sub> (see Massart et al. (2014) for further details). In this configuration only the concentrations are corrected by the assimilation, the emissions and surface fluxes remain unchanged. The processing and acquisition of the level 2 data in 2019 provided the satellite XCH<sub>4</sub> data 4 days behind real time. The high-resolution forecast is then coupled to the analysis experiment by merging the 4-day lower resolution forecast from the CO<sub>2</sub> and CH<sub>4</sub> analysis with the previous 1-day high resolution forecast (Fig. 3) in order to preserve the fine-scale features of the high-resolution forecast. Additionally, the high-resolution forecast coming from the operational NWP runs is used to reset the initial meteorological conditions in order to ensure the best possible accuracy of the transport. In this paper we will focus on using the CH<sub>4</sub> forecasts at high-resolution coming from the setup described above. The high-resolution

forecasts are run on a TCo1279 L137 grid of approximately 9 km x 9 km for a 5-day period and are initialized approximately 4 hours behind **real-time** every day from 00:00UTC.

Both high resolution forecasts and analysis use prescribed **CH<sub>4</sub>** surface fluxes. The anthropogenic emissions including fossil fuel emissions, agriculture and landfill/waste emissions are from the annual EDGARv4.2FT2010 data set (Olivier and G. Janssens-Maenhout, 2012) for 2010 with 0.1°x0.1° resolution and monthly resolution for the rice emissions (Matthews et al., 1991). Monthly mean wetland emissions come from a climatology (1990-2008) based on the LPJ-WHyMe model constrained by SCIAMACHY observations during the HYMN project (Spahni et al., 2011) with a resolution of 1° x 1° degree. The biomass burning emissions are from GFASv1.2 (Kaiser et al., 2012). Other sources and sinks include a monthly soil sink (Ridgwell et al., 1999), annual mean oceanic fluxes (Houweling et al., 1999, Lambert and Schmidt, 1993), and monthly mean fluxes from termites (Sanderson, 1996) and wild animals (Houweling et al., 1999). The chemical sink in the troposphere and the stratosphere is represented by a climatological monthly mean chemical loss rate (Bergamaschi et al., 2009). This is based on OH fields optimised with methyl chloroform using the TM5 model (Krol et al., 2005) with prescribed concentrations of the stratospheric radicals using the 2-D photochemical Max Planck Institute model. Figure 4 shows the geographical and seasonal structure of the surface fluxes. Large-scale and smoother structures are representative of the wetland, soil and agriculture fluxes, whereas the finer-scale and sharper structures are representative of the anthropogenic and fire emissions. Figure 5 shows the capability of the high-resolution forecasts at global and regional scales. Global seasonal cycles and synoptic scale concentrations are represented as well as concentrations at smaller scales such as plumes from point source emissions and orographic effects. Large point sources and associated plumes can be seen over Europe, for example over Madrid, Paris and Tours (western France). Inventory estimates suggest the modelled hotspot region near Tours is probably the result of solid waste landfill emissions. Other possibilities include emissions from both the enteric fermentation and wastewater treatment sectors, all of which may be linked to a landfill site. Over the Middle East region zoom, sharp point sources are seen in Teheran and Southern Iran as well as over Pakistan (Karachi) and also closer to the Himalayan region.

### 2.3. Monitoring suite

To monitor and compare the TROPOMI **XCH<sub>4</sub>** retrievals with the IFS **CH<sub>4</sub>** 9 km forecasts we re-use a part of the IFS assimilation system in a so-called monitoring mode. The system recomputes a high-resolution trajectory at 9 km initialised from the forecasts over a 12-hour monitoring window to calculate so-called first guess departures (difference between the observation and the model forecast) with the observations at the appropriate time. At each observation location the departure can be written as follows,

$$d = y - \mathbf{HM}(x_i) \quad (1)$$

where  $d$  is the departure,  $y$  the observation,  $\mathbf{H}$  the observation operator,  $\mathbf{M}$  the model integration or trajectory and  $x_i$  the initial  $\text{CH}_4$  condition at the beginning of the monitoring window. If we inject the retrieval equation (Rodgers, 2000) the departure becomes,

$$d = \mathbf{A}x_t + (\mathbf{I} - \mathbf{A})x_a + \epsilon - \mathbf{A}\mathbf{M}(x_i) - (\mathbf{I} - \mathbf{A})x_a \quad (2)$$

where  $x_t$  is the true  $\text{CH}_4$  concentration state (which is never exactly known),  $\mathbf{A}$  is the averaging kernel matrix which represents the sensitivity of the retrieval on the vertical profile with respect to the true state,  $\mathbf{I}$  the identity matrix,  $x_a$  the apriori information used in the retrieval and  $\epsilon$  the retrieval error term. The equation then simplifies to,

$$d = \mathbf{A}(x_t - \mathbf{M}(x_i)) + \epsilon \quad (3)$$

which is the difference between the true state and the forecast smoothed by the averaging kernel function plus the retrieval error term. Those departure values are thus strongly dependent on the averaging kernel function shape. For the TROPOMI  $\text{XCH}_4$  retrievals the mean averaging kernel function shows a homogenous sensitivity to the entire troposphere up to 200hPa where the sensitivity decreases in the stratosphere (Schneising et al., 2020). The averaging kernel function is not very variable between pixels or between different regions of the globe (not shown). Figure 6 shows the departures over various time scales (yearly, monthly and daily) for the global domain. Overall the departures (observation minus forecast) show a global positive bias of around 25 ppb (meaning observation values are above the model values) which could be attributed to model biases (Ramonet et al., 2019) and/or observation biases (Langerock et al., 2019). Ramonet et al. (2019) compared the CAMS  $\text{CH}_4$  forecasts with independent total column data. Results showed that the forecasts continuously underestimate the  $\text{CH}_4$  total columns by 5-20 ppb. Langerock et al. (2019) showed that the averaged total column bias for the TROPOMI  $\text{CH}_4$  retrievals bias is -0.32% (i.e. around -5ppb) but with respect to ground-based measurements.

Regional-scale error structures are evident from the observation-model comparison. For example, boreal regions are showing a band of negatives values, potentially attributed to systematic errors caused by surface albedo values during winter (see section 2.1) in the TROPOMI retrieval algorithm. Alternatively, they could be caused by  $\text{CH}_4$  biases at tropopause and lower stratosphere levels in the IFS model. Also, a possible time lag in the wetland emissions, which are calculated offline and provide boundary conditions in the IFS forecasting chain (see section 2.2) could cause such bias. The attribution of this type of large-scale error seen in the departures is not fully understood yet and is beyond the scope of this paper, although an understanding of these biases is crucial to further improve the quality of the CAMS  $\text{CH}_4$  forecasts and TROPOMI retrievals. At finer scales, structures are seen on the yearly average comparison and become more evident on the monthly timescales. Local differences are even stronger on a daily basis but recognising fine scale structures is challenging due to the lack of daily coverage. For those reasons a spatial filtering and temporal averaging of the departures is performed to extract and use the small-scale features seen in the departures.

### 3. Detection method

#### 3.1. Filtering the signal

To remove the large-scale features seen in the departures we have implemented a high pass Gaussian filtering. The filter uses a convolution of a 2D Gaussian kernel on a given averaged and binned departure field. In this study we use a  $0.1^\circ$  latitude-longitude binning. Due to ocean, cloud cover and quality control flagging a number of bins of the departure will show missing values that will jeopardize the convolution. This problem is solved technically by creating two auxiliary matrices that have missing values replaced by 0. The two auxiliary matrices are then defined as

$$\mathbf{D} = \begin{cases} d_m, & \text{if } n > N \\ 0, & \text{otherwise} \end{cases} \quad (4)$$

$$\mathbf{C} = \begin{cases} 1, & \text{if } n > N \\ 0, & \text{otherwise} \end{cases} \quad (5)$$

where  $d_m$  (with the subscript  $m$  standing for mean) is the average departure in the given bin,  $N$  the threshold of minimum number of observations in a given bin. In this study, we have chosen  $N = 2$  in order to avoid smoothing with very isolated pixels that can be faulty but also keep as much data as possible. Replacing the missing values by zeros in  $\mathbf{D}$  introduces an error after convolving (inducing low values due to smoothing out with zeros) in the filtered departures  $d_{hp}$  (with the subscript  $hp$  standing for high pass). This can be compensated by applying the same Gaussian filter on a matrix  $\mathbf{C}$  representing the selected bins for filtering (where number of counts are above  $N$ ) and using the ratio of the two filtered matrices to compensate for the missing value errors. Then a high pass filtering on a given observation space field (here departures) can be formulated as follows,

$$d_{hp} = d_m - \frac{G(\sigma) * \mathbf{D}}{G(\sigma) * \mathbf{C}} \quad (6)$$

where  $G(\sigma)$  is a 2D Gaussian kernel function with a  $\sigma$  length scale. The same filtering is also applied on the observation values  $y$  and the first guess values  $\mathbf{HM}(x_b)$  as this will be used for classification in section 3.2. Figure 7 shows the effect of the filtering on the observation-space data using a 30-day window and a length-scale of  $2^\circ$ . Firstly, we can see that the large-scale features in the departures such as the overall bias and regional variations are removed. Secondly, the departures, observations and first guess distributions are put towards gaussianity, centred around zero and displaying more a symmetrical shape and tails. This then makes the processing and the classification of the data much easier (see section 3.2).

To decide on the appropriate window length and Gaussian kernel length scale we have conducted sensitivity tests with different length scales ( $\sigma = [0.5, 1.0, 2.0, 5.0]$  degrees) and a window length of 10, 30 and 90 days. Figure 8 shows the resulting filtered departures normalized by the instrument precision for the 12 possible sensitivity tests. For tests with Gaussian kernel sizes of 0.5 and 1.0 degree the filtered signal is mostly weaker than the measurement precision and very few to no



detections of local anomalies will be made. Conversely, if the kernel is large the relative signal over the instrument precision is stronger but to the risk of picking up larger patterns than the targeted features, i.e. features that are directly related to local emissions in the CH<sub>4</sub> atmospheric distribution. For these reasons we found that a kernel of 2.0 degrees performs best. If the time window is short, e.g. 10 days, lower coverage could limit a correct detection of outliers especially in the case of isolated data points. Isolated data points that are spotting possible outliers could be filtered out towards 0 as the convolution do not have neighbouring points to use within the kernel range. The shorter the window, the narrower the kernel, the more likely this can occur. Conversely, if the time window is long, i.e. 90 days, will maximise the chances to have a good observation coverage for the convolution filter to run best but the this would reduce the ability to provide information on temporal variability. Also, the sharp spatial structures that correspond to more recent or sporadic emission events are smoothed in the time averaging effect decreasing the filtered departure over instrument precision ratio. For those reasons, we found that a time window of 30 days provides the most reasonable results.

### 3.2. Outlier classification

The final step is an outlier detection of the filtered departures. We choose to retain the values which have a filtered departure absolute value superior to the TROPOMI CH<sub>4</sub> measurement precision. If the filtered departures absolute values are lower than the measurement precision they are then considered as noise and ruled out. Further refinements to the current methodology could be done to find more optimal method for outlier detection using more advanced statistical methodologies. In the present study we found that the provided measurement precision with the satellite product provides suitable results. In addition to the outlier detection we perform a classification given the relative values and sign of the filtered observations and first guess values. This allows, us to define the following four categories:

- **high observations (red in Fig. 9):** where positive filtered observations are higher than filtered first-guess. This class is representative of high XCH<sub>4</sub> values detected by TROPOMI that are not seen as high or at all in the forecasts. These are likely originating from emissions that are not reported or under-estimated in the inventories. However, high observation categorisation may also be caused by poor quality observations category (see section 4.3).
- **high forecasts (green in Fig. 9):** where negative filtered first-guess are higher than filtered observations. This class is representative of high CH<sub>4</sub> values in the forecasts but not seen as strong or at all in the TROPOMI XCH<sub>4</sub> retrievals. High forecasts categorized data points are likely originating from emissions that are over-estimated or no longer being produced or even mis-located in the emission inventory.
- **low observations (blue in Fig. 9):** where negative filtered observations are lower than filtered first-guess. This class is representative of locally low XCH<sub>4</sub> values detected by TROPOMI but are not seen to be as low or at all in the forecasts. Poor-quality observations influenced by low surface albedo likely fall in that category (see section 4.3).



- **low forecasts (gold in Fig. 9):** where positive filtered first guess are lower than filtered observations. This class is representative of low  $\text{XCH}_4$  values in the forecasts but not seen as low or at all in the TROPOMI  $\text{XCH}_4$  retrievals. This category has generally much fewer data points. Orography could be a reason for data points to fall in that category, i.e. model surface height value that are higher than the observation value. Further developments of the method will likely use orography to improve the filtering.

In the maps of Fig 9., shades of the colours indicate the intensity of the offset, i.e. how far from perfectly matching observation versus forecasts the filtered departure is. The size of the points indicates the number of samples. A larger dot indicates more data points within the 30-day window to compute the statistics hence is more robust. Fig 9. gives an overview of such detections globally and cases are many and various. In the next section we will focus on specific cases studies using the under-reported or missed plumes (red) category and the over-reported or under-reported plumes (green) category to showcase the usefulness of the method.

## 4. Case studies

### 4.1. Under-estimation of local sources in the forecasts

**South Western USA and Mexico:** In figure 10, the method detects under-predicted local  $\text{CH}_4$  concentrations (in red) in the forecast system in three areas. This occurs in the Permian Basin region, located around the Texas-New Mexico border, where multiple oil drilling sites are currently operating. Those enhancements have been documented by de Gouw et al. (2020) and Zhang et al, (2020) showing the reliability of the presented method. Two other regions with a smaller bias and extent can be identified around the southern tip of Nevada by lake Meade and northern Baja California close to the US-Mexican border. To our knowledge those two cases have not been investigated or documented yet. Those two cases could be due to local albedo properties that could create local biases in the retrievals (see section 4.3), as we did not identify a facility responsible for those enhancements. This needs further investigation.

**Western Turkmenistan:** To confirm the ability of this methodology for the detection of large point-source emitters we also showcase very strong detection of anomalous concentrations over the western Turkmenistan. Our system detects strong enhancement during most of 2019 (Fig.11) that change in intensity and shape. The filtered departures can be very large (above 50 ppb) with a high number count in the bins (large size of the dots). As mentioned earlier in this paper, anomalously large  $\text{CH}_4$  sources from oil and gas production in this location have been documented and detected using TROPOMI combined with private sector satellite data by Varon et al. (2019).

### 4.2. Over estimation of local sources in the forecasts

**Western Russia:** Our detection system shows two local point sources, that show large first-guess values that are not seen by TROPOMI  $\text{XCH}_4$  (green dots in Fig. 12). The features do not show large sampling (small dots) in time but do exhibit the

shape of plumes, with strong departures near the point sources. One is very close to Moscow and corresponds to the Domodedovo airport surroundings. The other source detection is near the Volga river with a location matching small drilling fields seen in visible satellite images. In these two locations the detection method suggests that emission inventories are over-estimating local sources, which in reality are now producing reduced emissions or are no longer active emitters (at least during the period of monitoring).

**Los Angeles:** Similar features can occur in the area of Los Angeles. Figure 10 shows significant over-prediction of CH<sub>4</sub> (green dots) specifically over San Bernardino and Palmdale. Both towns have industrial facilities and regional airports. The detection is stronger in the 2019-09-01 and 2019-10-01 windows than in the 2019-07-01 and 2019-08-01 windows. Differences in intensity could be attributed to the monthly emission changes but also attributed to seasonal atmospheric transport changes due to different meteorological situation between windows. For example, if the overall windspeed increases near the source less accumulation of CH<sub>4</sub> would be seen leading to smaller departures and less detection.

Such cases in very different locations show the capability of the method to detect not only missing or underreported point sources but also overreported cases. This can only be achieved with combining numerical models forecasts and satellite measurements at close-matching high horizontal resolution (9 km and 7 km respectively). It is also important to mention that the method presented here is subject to uncertainties due to both model transport errors and representation error, although the error associated to emission generally dominate. Further work is needed to account for atmospheric transport and more generally to account for the weather variability in the detection method. Techniques as described in Barré et al., 2020 show interesting potential to be used for this topic.

#### 4.3. Local retrieval issues

The retrieval can be affected by albedo surface issues (see section 2). The filtering is not able to remove features with geographical extent smaller than the size of the Gaussian kernel (see section 3.1). Figure 13 and figure 14 presents examples of such issues where the same pattern is seen repeatedly in the outlier detection. In figure 13, the persistent pattern in shape and intensity corresponds to a land surface feature seen in visible satellites images that produces consistently higher TROPOMI XCH<sub>4</sub> values than its surroundings. The inverse can be true as well displaying local areas with consistently lower local XCH<sub>4</sub> retrievals (not shown). Thus, great care should be taken when diagnosing such filtered departures. Features with a consistent shape and intensity are retrieval error artefacts, as atmospheric plumes would show more variability and not a consistent shape over months as illustrated in Fig. 13 and in Fig. 14. In Fig. 13 the pattern clearly corresponds to a change in the land surface type in visible satellite images (e.g. google maps) and the same shape is seen for the span of more than 10 weeks (four 30-days windows that are around 6 weeks apart). In Fig. 14 the shape of the anomaly is also consistent over 4 months using 30-days windows. No clear visible change in the land surface is seen in that case. More investigation is needed by the retrieval provider to understand this systematic local bias. Further improvements of the method could be implemented with pattern shape recognition to automatically discard or classify persistent shapes over time. Satellite retrieval providers could be notified about of such biases, in order to improve the quality of the satellite product. However, there is still potential impact of temporally

310 and spatially variable small-scale albedo features (e.g. due to snow cover). Such transient features would still go undetected  
by a possible detection of persistent shape features.

## 5. Conclusions

In this paper we have shown the potential of systematic detection of anthropogenic CH<sub>4</sub> point and local source emissions  
relative to known emission inventory data using the TROPOMI satellite measurements in combination with high resolution  
315 CH<sub>4</sub> forecasts. While many studies have shown detailed analysis of a few case studies using TROPOMI observations, this is  
the first time that a systematic way to detect strong anthropogenic local emitters of CH<sub>4</sub> and to compare results with emission  
inventories is presented. The method presented here does not only allow for the detection of unreported or missing sources but  
also targets over-reported sources in the inventories. The method also has the potential of detecting systematic local retrieval  
errors which can help to improve the satellite product. We demonstrated the potential of the methodology by focusing on  
320 several case studies but further work is required to provide a global assessment using several years from this dataset.

Our method is novel by combining information from multiple sources (emission inventory, modelled surface fluxes, and  
observations) in a data assimilation framework to detect and analyse observed anomalies. We have used global emission  
inventories and fluxes that were the best possible global estimates we had available at the time when running our system. Using  
different emission inventories from research specific activities that are more specific to local regions, for instance, could  
325 provide different answers. In that way our methodology could provide an efficient way to validate improvements in sector-  
specific emission inventories. For example, using revised CH<sub>4</sub> inventories such as presented by Maasakers et al. (2016) over  
the USA or more recently by Scrapelli et al., 2020 globally could lead to different detection patterns. Bottom up inventories  
will always lag in time and therefore cannot track rapid emission changes such as pipeline and gas facility blowouts. Satellite  
measurements have a clear added value for timely detection in the case of large emissions.

330 Combining satellite measurements, forecasts and emission inventories partially using a data assimilation system paves the  
way to estimate the emissions themselves. Inverse modelling studies to estimate CH<sub>4</sub> emissions have been done with  
SCIAMACHY and GOSAT CH<sub>4</sub> satellite data generally performed at rather low resolution and focus specific study sites (e.g.  
Jacob et al., 2016). To our knowledge no published studies showed global inversions using TROPOMI data updating emissions  
close to the 10 km scale globally. Inverse modelling is computationally expensive and in the case of running operations beyond  
335 10 km scales to close-match satellite observations is a challenge that needs to be overcome over the next decade. Efforts are  
underway to implement a sector-specific inverse high-resolution modelling monitoring system as part of the CAMS service  
evolution at ECMWF and the future Copernicus CO<sub>2</sub> service at global and regional scales (e.g. Barré et al., 2019, Bousserez  
et al., 2019, Pinty et al., 2019, Janssens-Maenhout et al., 2020). Approaches combining global and regional modelling could  
be adopted to perform inversion at fine scales but at the cost of missing fine-scale detection outside the regional domains.  
340 Large and local CH<sub>4</sub> emissions events could occur in very remote areas, which are typically not considered in regional  
modelling setups (e.g. West Turkmenistan, Varon et al., 2019). Systematic detection will then require setting up many

regional subdomains leading again to computational burden for a single monitoring entity. We have demonstrated that monitoring of satellite XCH<sub>4</sub> departures at high resolution at global scale using already existing parts of a forecasting chain remains an affordable solution to develop a much needed capability: tracking rapidly changing CH<sub>4</sub> sources across the world and support the urgently needed effort on developing climate policies for reducing anthropogenic CH<sub>4</sub> emissions.

## References

- Agustí-Panareda, A., Diamantakis, M., Massart, S., Chevallier, F., Muñoz-Sabater, J., Barré, J., Curcoll, R., Engelen, R., Langerock, B., Law, R. M., Loh, Z., Morguí, J. A., Parrington, M., Peuch, V.-H., Ramonet, M., Roehl, C., Vermeulen, A. T., Warneke, T., and Wunch, D.: Modelling CO<sub>2</sub> weather – why horizontal resolution matters, *Atmos. Chem. Phys.*, 19, 7347–7376, <https://doi.org/10.5194/acp-19-7347-2019>, 2019.
- Alvarez, R. A., Zavala-Araiza, D., Lyon, D. R., Allen, D. T., Barkley, Z. R., Brandt, et al. (2018). Assessment of methane emissions from the U.S. oil and gas supply chain. *Science*, 361, 186– 188. <https://doi.org/10.1126/science.aar7204>
- Bergamaschi, P., Frankenberg, C., Meirink, J. F., Krol, M., Villani, M. G., Houweling, S., Dentener, F., Dlugokencky, E. J., Miller, J. B., Gatti, L. V., Engel, A., , and Levin, I. (2009). Inverse modeling of global and regional CH<sub>4</sub> emissions using SCIAMACHY satellite retrievals. *J. Geophys. Res.*, 114.
- Barré J., Massart S., Ades M., Jones L., and Engelen R. (2019)., Emission optimisations first attempt based on Ensemble of DA for atmospheric composition, ECMWF technical memoranda, <https://doi.org/10.21957/4grkg5ga0>
- Barré, J., Petetin, H., Colette, A., Guevara, M., Peuch, V.-H., Rouil, L., Engelen, R., Inness, A., Flemming, J., Pérez García-Pando, C., Bowdalo, D., Meleux, F., Geels, C., Christensen, J. H., Gauss, M., Benedictow, A., Tsyro, S., Frieze, E., Struzewska, J., Kaminski, J. W., Douros, J., Timmermans, R., Robertson, L., Adani, M., Jorba, O., Joly, M., and Kouznetsov, R.: Estimating lockdown induced European NO<sub>2</sub> changes, *Atmos. Chem. Phys. Discuss.*, <https://doi.org/10.5194/acp-2020-995>, in review, 2020.
- Bousserez, N., 2019. Towards a Prototype Global CO<sub>2</sub> Emissions Monitoring System for Copernicus. *arXiv preprint arXiv:1910.11727*.
- Conley, S., Franco, G., Faloona, I., Blake, D. R., Peischl, J., & Ryerson, T. B. (2016). Methane emissions from the 2015 Aliso Canyon blowout in Los Angeles, CA. *Science*, 351, 1317– 1320. <https://doi.org/10.1126/science.aaf2348>

- de Gouw, J.A., Veefkind, J.P., Roosenbrand, E. et al. Daily Satellite Observations of Methane from Oil and Gas Production Regions in the United States. *Sci Rep* 10, 1379 (2020). <https://doi.org/10.1038/s41598-020-57678-4>
- Engelen, R. J., Serrar, S., and Chevallier, F. ( 2009), Four-dimensional data assimilation of atmospheric CO<sub>2</sub> using AIRS observations, *J.* 114, D03303, doi:[10.1029/2008JD010739](https://doi.org/10.1029/2008JD010739).
- 380 Flemming, J., Huijnen, V., Arteta, J., Bechtold, P., Beljaars, A., Blechschmidt, A.-M., Diamantakis, M., Engelen, R. J., Gaudel, A., Inness, A., Jones, L., Josse, B., Katragkou, E., Marecal, V., Peuch, V.-H., Richter, A., Schultz, M. G., Stein, O., and Tsikerdekis, A.: Tropospheric chemistry in the Integrated Forecasting System of ECMWF, *Geosci. Model Dev.*, 8, 975–1003, <https://doi.org/10.5194/gmd-8-975-2015>, 2015.
- 385 Hasekamp, O., Lorente, A., Hu, H., Butz, A., aan de Brugh, J., and Landgraf, J., Algorithm Theoretical Baseline Document for Sentinel-5 Precursor Methane retrieval, SRON-S5P-LEV2-RP-001, <https://sentinel.esa.int/documents/247904/2476257/Sentinel-5P-TROPOMI-ATBD-Methane-retrieval>, 2019
- Holm, E., Forbes, R., Lang, S., Magnusson, L., and Malardel, S.: New model cycle brings higher resolution, ECMWF Newsletter, No. 147, available at: <https://www.ecmwf.int/en/elibrary/16299-newsletter-no-147-spring-2016> (last access: 30 May 2019), 2016. a, b
- Houweling, S., Kaminski, T., Dentener, F., Lelieveld, J., and Heimann, M.: Inverse modeling of methane sources and sinks using the adjoint of a global transport model, *J. Geophys. Res.*, 104, 26137–26160, doi:[10.1029/1999JD900428](https://doi.org/10.1029/1999JD900428), 1999
- 395 Hu, H., Landgraf, J., Detmers, R., Borsdorff, T., Aan de Brugh, J., Aben, I., et al. ( 2018). Toward global mapping of methane with TROPOMI: First results and intersatellite comparison to GOSAT. *Geophysical Research Letters*, 45, 3682– 3689. <https://doi.org/10.1002/2018GL077259>
- IPCC. Observations: Atmosphere and Surface (2013).
- 400 Jacob, D. J., Turner, A. J., Maasakkers, J. D., Sheng, J., Sun, K., Liu, X., Chance, K., Aben, I., McKeever, J., & Frankenberg, C. (2016). Satellite observations of atmospheric methane and their value for quantifying methane emissions. *Atmospheric Chemistry and Physics*, 16( 22), 14,371– 14,396. <https://doi.org/10.5194/acp-16-14371-2016>
- 405 Janssens-Maenhout, G., B. Pinty, M. Dowell, H. Zunker, E. Andersson, G. Balsamo, J.-L. Bézy, T. Brunhes, H. Bösch, B. Bojkov, D. Brunner, M. Buchwitz, D. Crisp, P. Ciais, P. Counet, D. Dee, H. Denier van der Gon, H. Dolman, M. Drinkwater, O. Dubovik, R. Engelen, T. Fehr, V. Fernandez, M. Heimann, K. Holmlund, S. Houweling, R. Husband, O. Juvyns, A. Kentarchos, J. Landgraf, R. Lang, A. Löschner, J. Marshall, Y. Meijer, M. Nakajima, P.I. Palmer, P. Peylin, P. Rayner, M.

- ✖ Scholze, B. Sierk, J. Tamminen, P. Veeffkind, Towards an operational anthropogenic CO<sub>2</sub> emissions monitoring and verification support capacity, Bulletin of the American Meteorological Society (accepted), 2020, doi: 10.1175/BAMS-D-19-0017.1
- Kaiser, J. W., Heil, A., Andreae, M. O., Benedetti, A., Chubarova, N., Jones, L., Morcrette, J.-J., Razinger, M., Schultz, M. G., Suttie, M., and van der Werf, G. R.: Biomass burning emissions estimated with a global fire assimilation system based on observed fire radiative power, Biogeosciences, 9, 527–554, <https://doi.org/10.5194/bg-9-527-2012>, 2012.
- ✖ 415 Kirschke, S., Bousquet, P., Ciais, P. *et al.* Three decades of global methane sources and sinks. *Nature Geosci* 6, 813–823 (2013). <https://doi.org/10.1038/ngeo1955>
- Kort, E. A., Frankenberg, C., Costigan, K. R., Lindenmaier, R., Dubey, M. K., and Wunch, D. ( 2014), Four corners: The largest US methane anomaly viewed from space, *Geophys. Res. Lett.*, 41, 6898– 6903, doi:10.1002/2014GL061503.
- ✖ 420 Krol, M., Houweling, S., Bregman, B., van den Broek, M., Segers, A., van Velthoven, P., Peters, W., Dentener, F., and Bergamaschi, P.: The two-way nested global chemistry-transport zoommodel TM5: algorithm and applications, *Atmos. Chem. Phys.*, 5, 417–432, doi:10.5194/acp-5-417-2005, 2005.
- ✖ 425 Lambert, G. and Schmidt, S.: Reevaluation of the oceanic flux of methane: uncertainties and long term variations, *Chemosph. Global Change Sci.*, 26, 579–589, 1993
- Langerock B., Kumar M., Lambert J., Lorente A. and Landgraf J., First comparison results for the S5P CH<sub>4</sub> product based on correlative reference measurements acquired by FTIR instruments contributing to NDACC and TCCON networks. [http://mpc-vdaf.tropomi.eu/ProjectDir/reports//pdf/S5P-MPC-VDAF-VWA-L2\\_CH4\\_20190301.pdf](http://mpc-vdaf.tropomi.eu/ProjectDir/reports//pdf/S5P-MPC-VDAF-VWA-L2_CH4_20190301.pdf)
- ✖ 430 Maasakkers, Joannes D., Daniel J. Jacob, Melissa P. Sulprizio, Alexander J. Turner, Melissa Weitz, Tom Wirth, Cate Hight, Mark Degrep ueiredo, Mausami Desai, Rachel Schmeltz, Leif Hockstad, Anthony A. Bloom, Kevin W. Bowman, Seongeun Jeong, and Marc L. Fischer: Gridded National Inventory of U.S. Methane Emissions *Environmental Science & Technology* 2016 50 (23), 13123-13133, DOI: 10.1021/acs.est.6b02878
- ✖ 435 Massart, S., Agustí-Panareda, A., Aben, I., Butz, A., Chevallier, F., Crevoisier, C., Engelen, R., Frankenberg, C., and Hasekamp, O.: Assimilation of atmospheric methane products into the MACC-II system: from SCIAMACHY to TANSO and IASI, *Atmos. Chem. Phys.*, 14, 6139–6158, <https://doi.org/10.5194/acp-14-6139-2014>, 2014.
- ✖ 440 Massart, S., Agustí-Panareda, A., Heymann, J., Buchwitz, M., Chevallier, F., Reuter, M., Hilker, M., Burrows, J. P., Deutscher, N. M., Feist, D. G., Hase, F., Sussmann, R., Desmet, F., Dubey, M. K., Griffith, D. W. T., Kivi, R., Petri, C., Schneider, M.,

and Velazco, V. A.: Ability of the 4-D-Var analysis of the GOSAT BESD XCO<sub>2</sub> retrievals to characterize atmospheric CO<sub>2</sub> at large and synoptic scales, *Atmos. Chem. Phys.*, 16, 1653–1671, <https://doi.org/10.5194/acp-16-1653-2016>, 2016.

- 445 Matthews, E., Fung, I., and Lerner, J.: Methane emission from rice cultivation: geographic and seasonal distribution of cultivated areas and emissions, *Global Biogeochem. Cy.*, 5, 3–24, doi:10.1029/90GB02311, 1991

Myhre G. et al., “Anthropogenic and natural radiative forcing” in *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*, T. F.

- 450 Stocker et al., Eds. (Cambridge University Press, Cambridge, United Kingdom, 2013), pp. 659–740.

Olivier and G. Janssens-Maenhout, *CO<sub>2</sub> Emissions from Fuel Combustion -- 2012 Edition*, IEA CO<sub>2</sub> report 2012, Part III, Greenhouse-Gas Emissions, ISBN 978-92-64-17475-7

- 455 Pandey S., Ritesh Gautam, Sander Houweling, Hugo Denier van der Gon, Pankaj Sadavarte, Tobias Borsdorff, Otto Hasekamp, Jochen Landgraf, Paul Tol, Tim van Kempen, Ruud Hoogeveen, Richard van Hees, Steven P. Hamburg, Joannes D. Maasackers and Ilse Aben, Satellite observations reveal extreme methane leakage from a natural gas well blowout, *Proceedings of the National Academy of Sciences*, <https://doi.org/10.1073/pnas.1908712116>, (201908712), (2019).

- 460 Pinty B., P. Ciais, D. Dee, H. Dolman, M. Dowell, R. Engelen, K. Holmlund, G. Janssens-Maenhout, Y. Meijer, P. Palmer, M. Scholze, H. Denier van der Gon, M. Heimann, O. Juvyns, A. Kentarchos and H. Zunker (2019) An Operational Anthropogenic CO<sub>2</sub> Emissions Monitoring & Verification Support Capacity – Needs and high level requirements for in situ measurements, doi: 10.2760/182790, European Commission Joint Research Centre, EUR 29817 EN.

- 465 Ramonet, M., A. Wagner, M. Schulz, Y. Christophe, H. J. Eskes, S. Basart, A. Benedictow, Y. Bennouna, A.-M. Blechschmidt, S. Chabrillat, E. Cuevas, A. ElYazidi, H. Flentje, K.M. Hansen, U. Im, J. Kapsomenakis, B. Langerock, A. Richter, N. Sudarchikova, V. Thouret, T. Warneke, C. Zerefos, Validation report of the CAMS near-real-time global atmospheric composition service: Period June - August 2019, Copernicus Atmosphere Monitoring Service (CAMS) report, CAMS84\_2018SC1\_D1.1.1\_JJA2019\_v1.pdf, November 2019, doi:10.24380/def9-na43.

470

Ridgwell, A. J., Marshall, S. J., and Gregson, K.: Consumption of atmospheric methane by soils: a process-based model, *Global Biogeochem. Cy.*, 13, 59–70, doi:10.1029/1998GB900004, 1999

- Sanderson, M. G.: Biomass of termites and their emissions of methane and carbon dioxide: a global database, *Global Biogeochem. Cy.*, 10, 543–557, doi:10.1029/96GB01893
- 475



Saunois, M., Bousquet, P., Poulter, B., Peregon, A., Ciais, P., Canadell, J. G., Dlugokencky, E. J., Etiope, G., Bastviken, D., Houweling, S., Janssens-Maenhout, G., Tubiello, F. N., Castaldi, S., Jackson, R. B., Alexe, M., Arora, V. K., Beerling, D. J., Bergamaschi, P., Blake, D. R., Brailsford, G., Brovkin, V., Bruhwiler, L., Crevoisier, C., Crill, P., Covey, K., Curry, C., Frankenberg, C., Gedney, N., Höglund-Isaksson, L., Ishizawa, M., Ito, A., Joos, F., Kim, H. S., Kleinen, T., Krummel, P., Lamarque, J. F., Langenfelds, R., Locatelli, R., Machida, T., Maksyutov, S., McDonald, K. C., Marshall, J., Melton, J. R., Morino, I., Naik, V., O'Doherty, S., Parmentier, F. J. W., Patra, P. K., Peng, C., Peng, S., Peters, G. P., Pison, I., Prigent, C., Prinn, R., Ramonet, M., Riley, W. J., Saito, M., Santini, M., Schroeder, R., Simpson, I. J., Spahni, R., Steele, P., Takizawa, A., Thornton, B. F., Tian, H., Tohjima, Y., Viovy, N., Voulgarakis, A., vanWeele, M., van der Werf, G. R., Weiss, R., Wiedinmyer, C., Wilton, D. J., Wiltshire, A., Worthy, D., Wunch, D., Xu, X., Yoshida, Y., Zhang, B., Zhang, Z., & Zhu, Q. (2016). The global methane budget 2000–2012. *Earth System Science Data*, 8( 2), 697–751. <https://doi.org/10.5194/essd-8-697-2016>

Scarpelli, T. R., Jacob, D. J., Maasakkers, J. D., Sulprizio, M. P., Sheng, J.-X., Rose, K., Romeo, L., Worden, J. R., and Janssens-Maenhout, G.: A global gridded ( $0.1^\circ \times 0.1^\circ$ ) inventory of methane emissions from oil, gas, and coal exploitation based on national reports to the United Nations Framework Convention on Climate Change, *Earth Syst. Sci. Data*, 12, 563–575, <https://doi.org/10.5194/essd-12-563-2020>, 2020.

Schneising, O., Buchwitz, M., Reuter, M., Vanselow, S., Bovensmann, H., and Burrows, J. P.: Remote sensing of methane leakage from natural gas and petroleum systems revisited, *Atmos. Chem. Phys.*, 20, 9169–9182, <https://doi.org/10.5194/acp-20-9169-2020>, 2020.

Shoemaker J. K., D. P. Schrag, M. J. Molina, V. Ramanathan, Climate change. What role for short-lived climate pollutants in mitigation policy? *Science* 342, 1323–1324 (2013).


Spahni, R., Wania, R., Neef, L., van Weele, M., Pison, I., Bousquet, P., Frankenberg, C., Foster, P. N., Joos, F., Prentice, I. C., and van Velthoven, P.: Constraining global methane emissions and uptake by ecosystems, *Biogeosciences*, 8, 1643–1665, <https://doi.org/10.5194/bg-8-1643-2011>, 2011.

Varon, D. J., Jacob, D. J., McKeever, J., Jervis, D., Durak, B. O. A., Xia, Y., & Huang, Y. (2018). Quantifying methane point sources from fine-scale satellite observations of atmospheric methane plumes. *Atmospheric Measurement Techniques*, 11, 5673–5686. <https://doi.org/10.5194/amt-11-5673-2018>



510 Varon, D. J., McKeever, J., Jervis, D., Maasakkers, J. D., Pandey, S., Houweling, S., et al. (2019). Satellite discovery of anomalously large methane point sources from oil/gas production. *Geophysical Research Letters*, 46, 13507–13516. <https://doi.org/10.1029/2019GL083798>

Veefkind, J. P., Aben, I., McMullan, K., Förster, H., de Vries, J., Otter, G., et al. (2012). TROPOMI on the ESA Sentinel-5  
515 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. <https://doi.org/10.1016/j.rse.2011.09.027>

 Zhang, Y., Gautam, R., Pandey, S., Omara, M., Maarakkers, J. D., Sadavarte, P., et al, Quantifying methane emissions from the largest oil-producing basin in the United States from space, *Science Advances*, doi: 10.1126/sciadv.aaz5120 (2020)

520

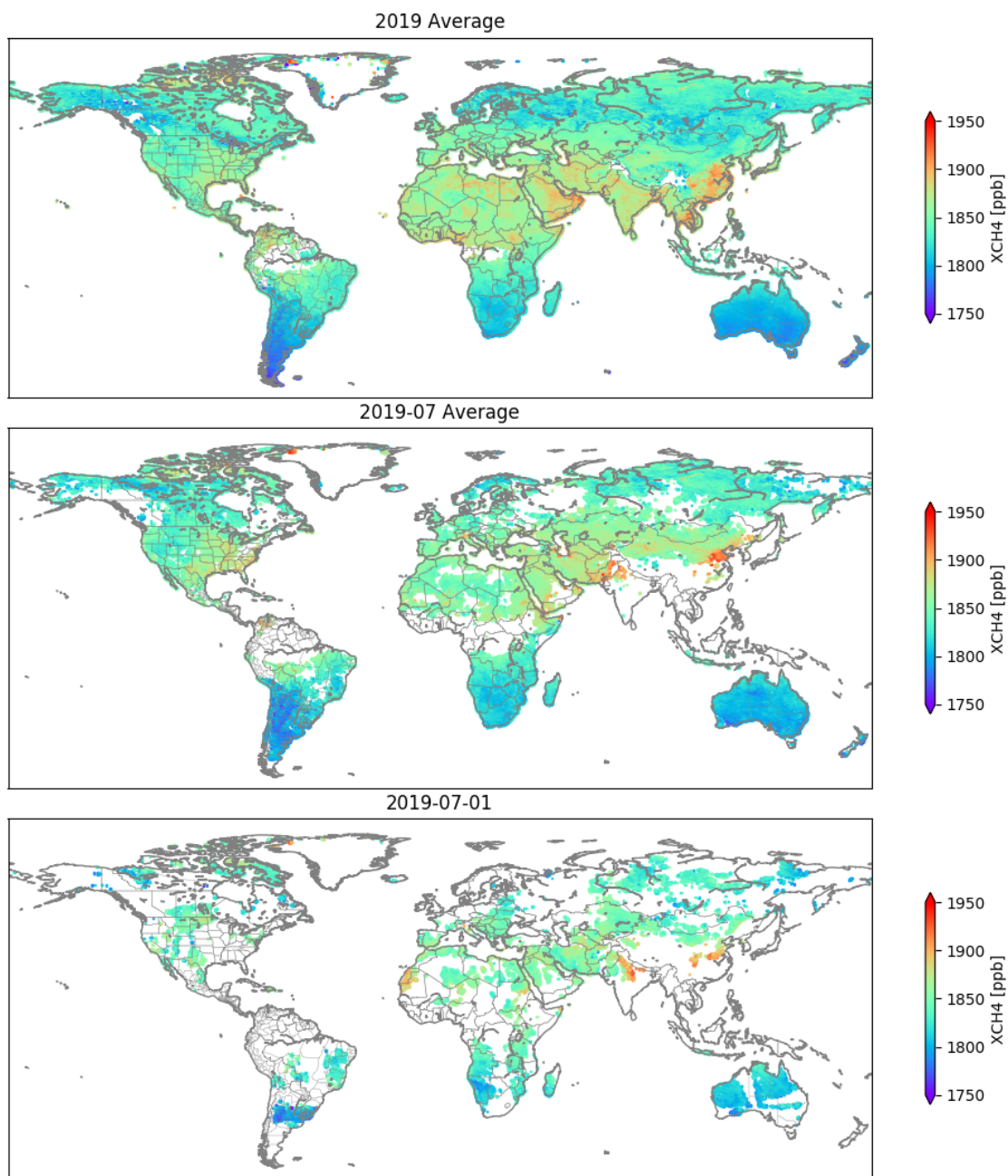


Figure 1. Global average of TROPOMI XCH<sub>4</sub> column-averaged dry-air mixing ratios for the full year 2019, July 2019 and July 1st, 2019 (top to bottom).

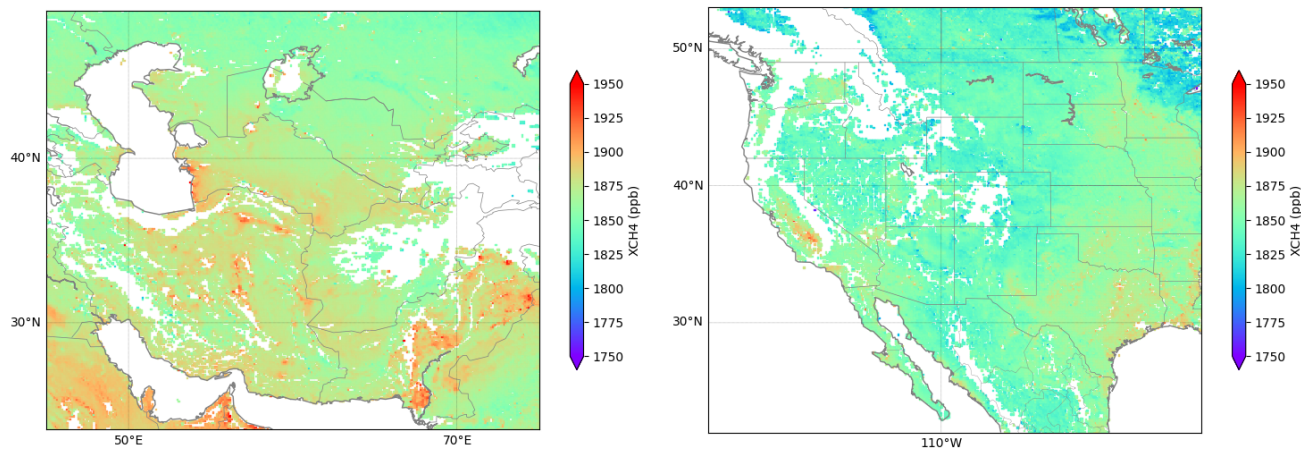


Fig 2. Regional zooms of TROPOMI  $\text{XCH}_4$  columns for the full year 2019. Middle East (left) and Central-Western North America (right).

530

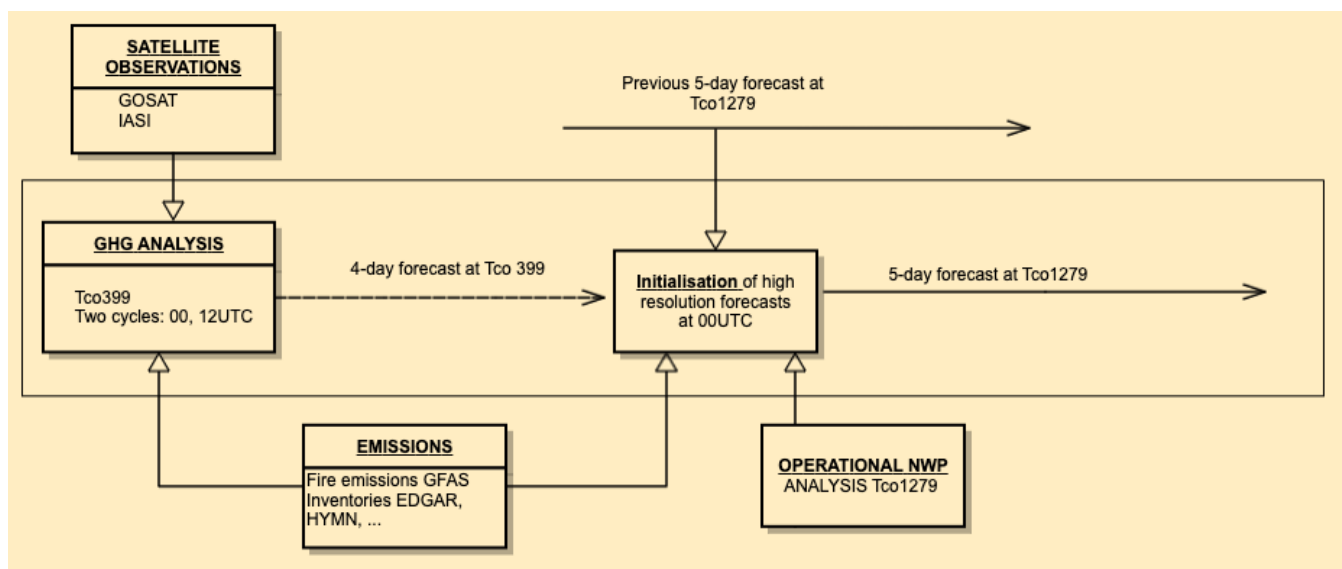
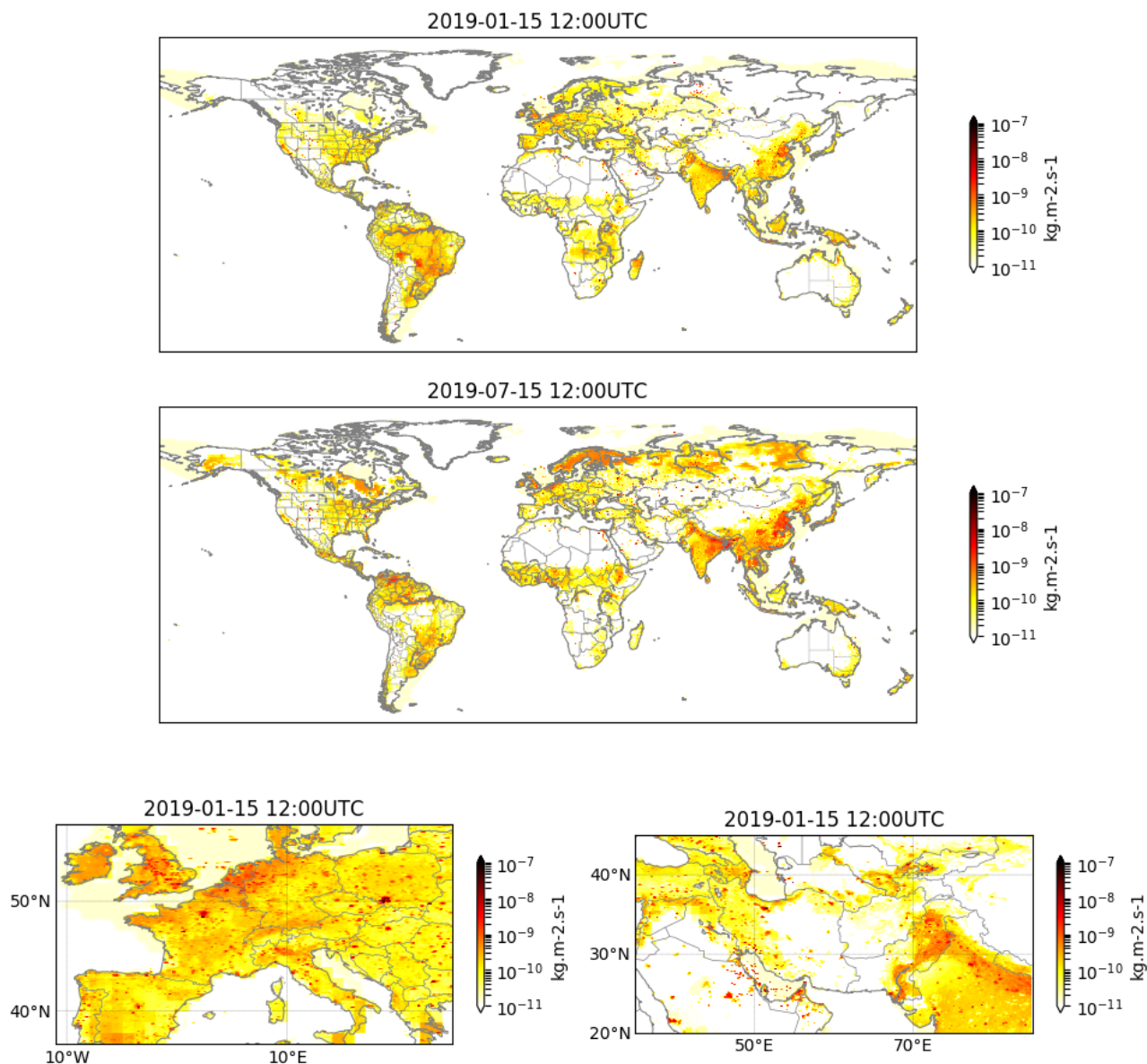


Figure 3. Flow chart of the CAMS greenhouse gas analysis and forecast system.



535 **Figure 4. Examples of combined net fluxes (positive only shown due to the logarithmic scale) that constitute the surface boundary conditions of the IFS high resolution CH<sub>4</sub> forecast. Global and regional scale examples for 2019-01-15 and 2019-07-05 at 12:00 UTC.**

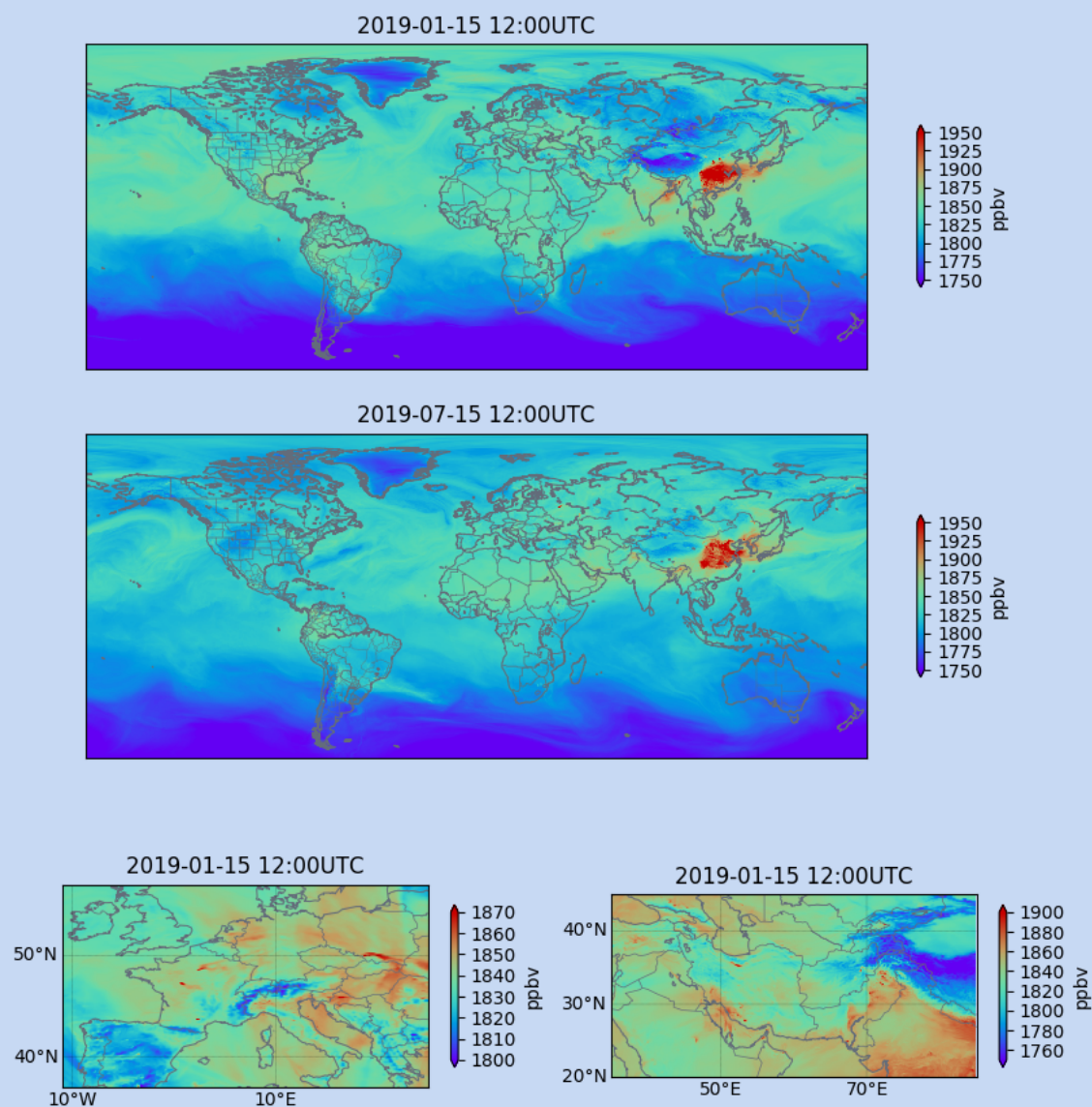


Figure 5. Examples of outputs of the IFS high resolution  $\text{CH}_4$  forecasts displaying snapshots at global and regional scale of the total column mean molar fractions for 2019-01-15 and 2019-07-15 at 12:00 UTC. Lower panels show parts of Europe (left) and Middle East (right).



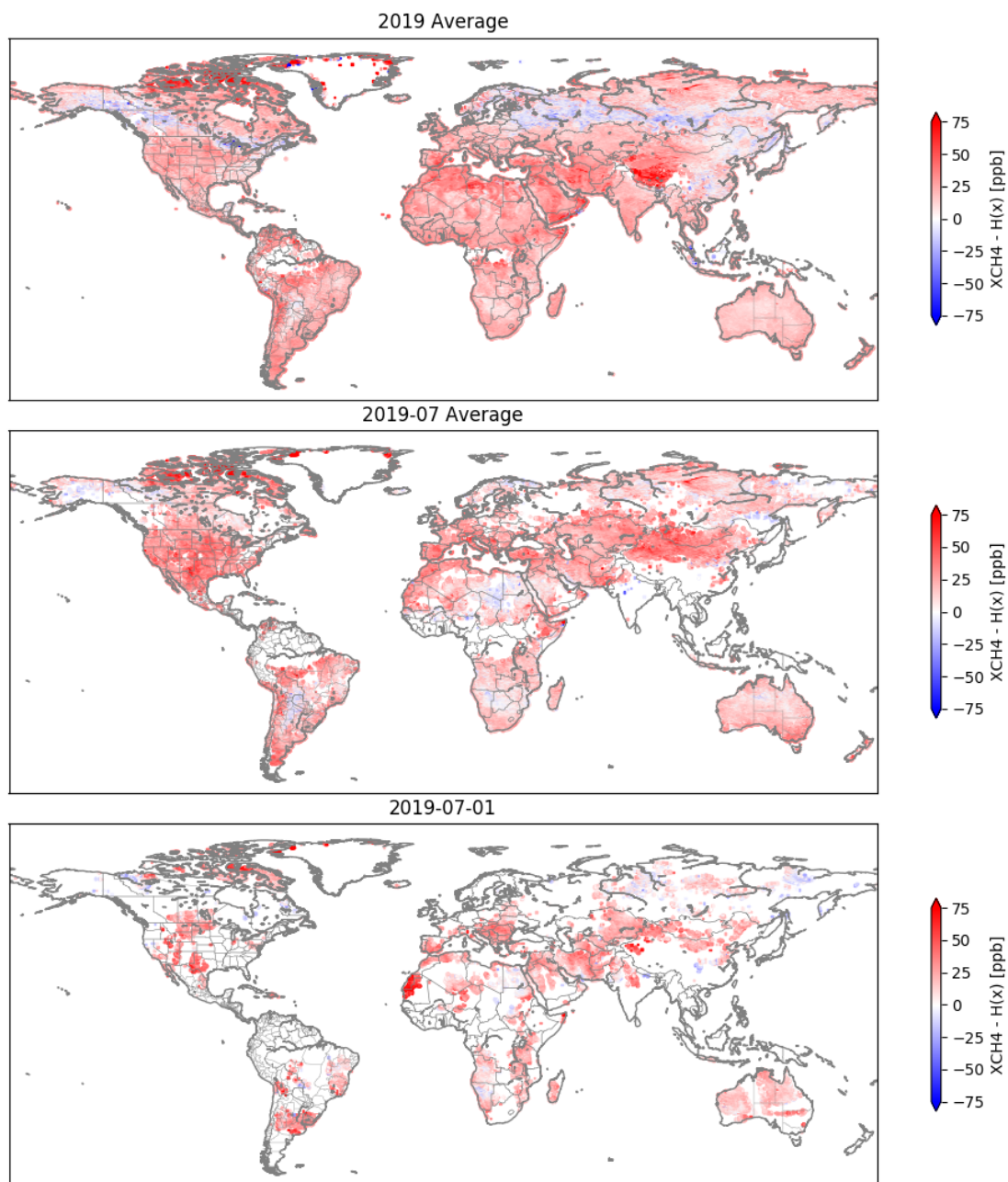


Figure 6. Departures values computed with the observation displayed in figure 1, for the full year 2019, July 2019 and July 1st, 2019.

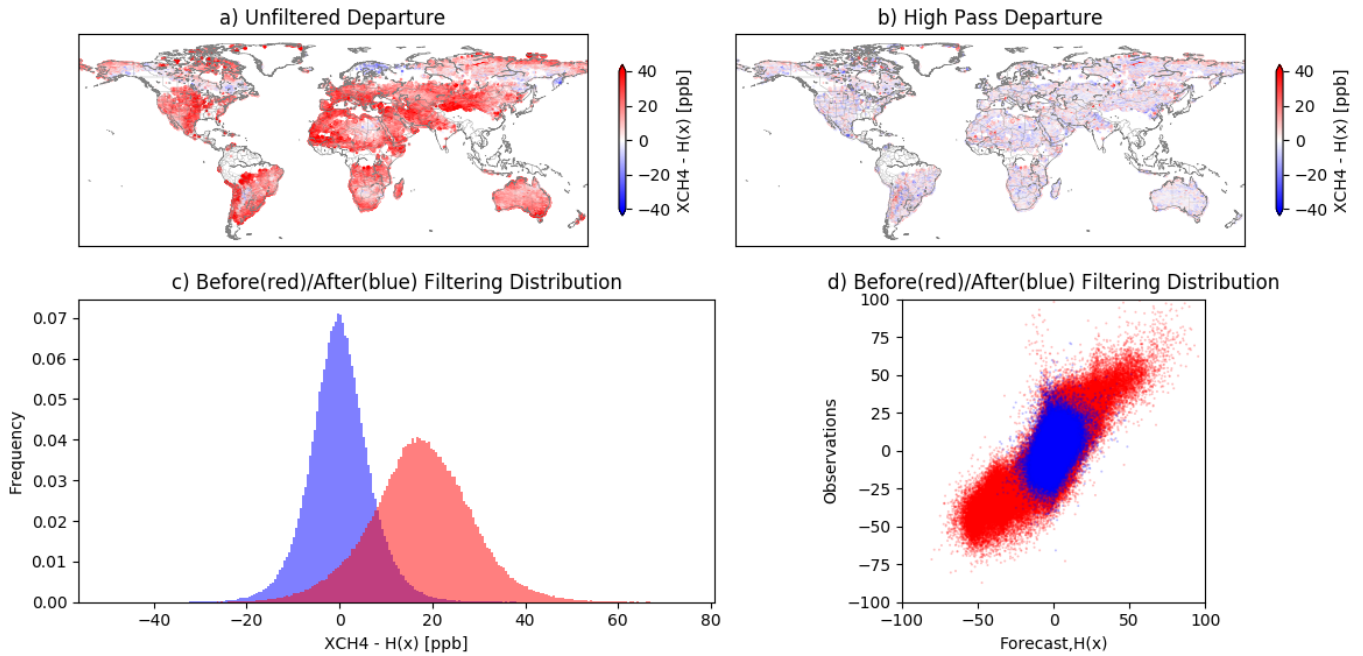
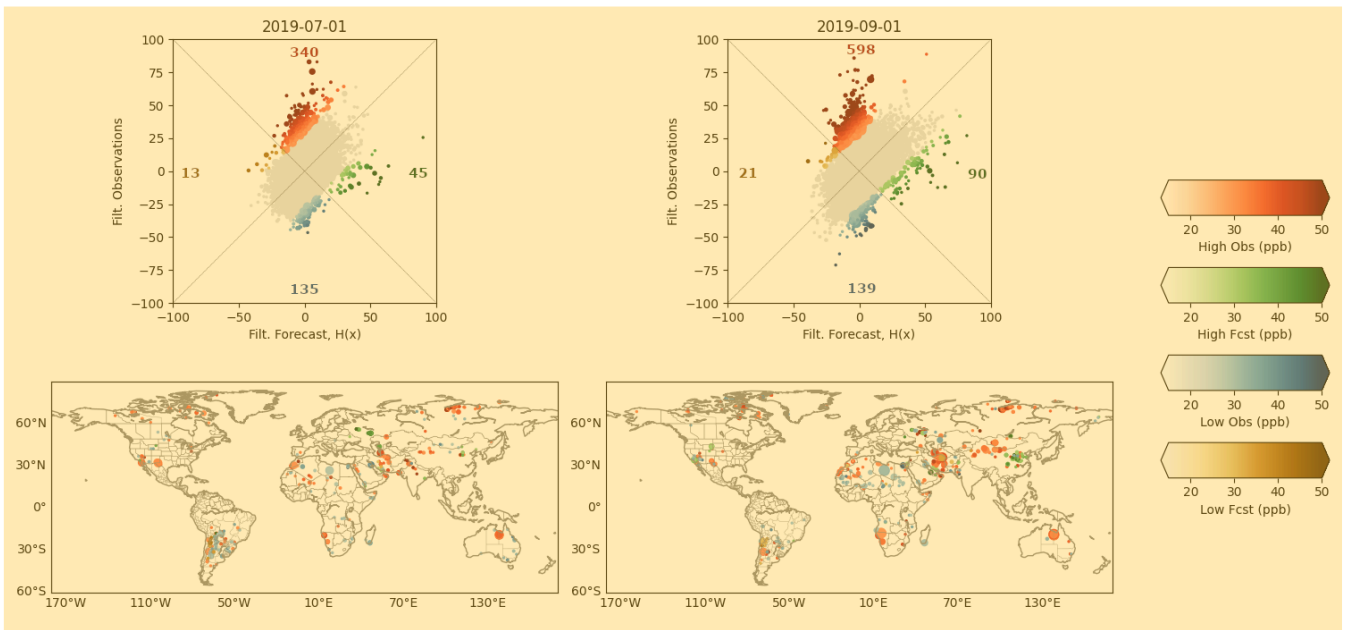


Figure 7. Example of the high pass filtering effect over a 30-day window ending 2019-07-01 with a  $2^\circ$  Gaussian kernel length scale. a) The unfiltered departures, b) the filtered departures, c) histograms comparing unfiltered (red) versus the filtered (blue) departures and d) 2D distributions in the observation and first-guess space for unfiltered (red) and filtered (blue) data points. Note that the unfiltered data points have been centred around the mean for this plot to make it comparable to the filtered distribution.







**Figure 9. Examples of the outlier classification. Top panels: global distributions in the observation-first guess space for two different end dates of 30-day window (July 1st 2019 and September 1st 2019). Colours illustrate the four different data classes with number indicating the amount of outliers. Bottom panels: outlier classes localisation example over the globe. Darker dots show larger departures. Larger dots indicate that more occurrences have been detected in the bin and time window.**

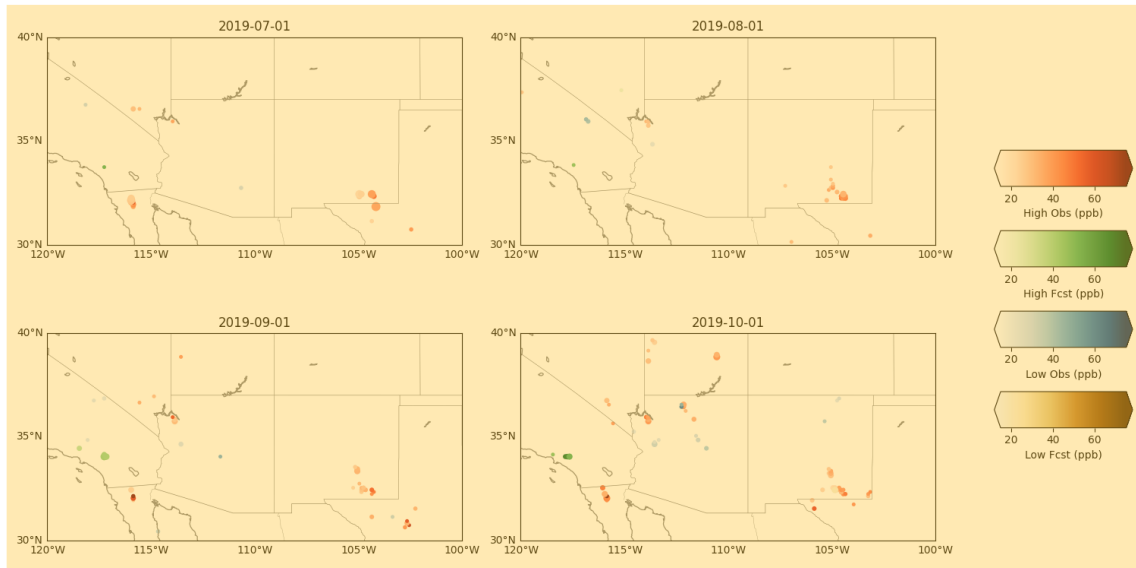
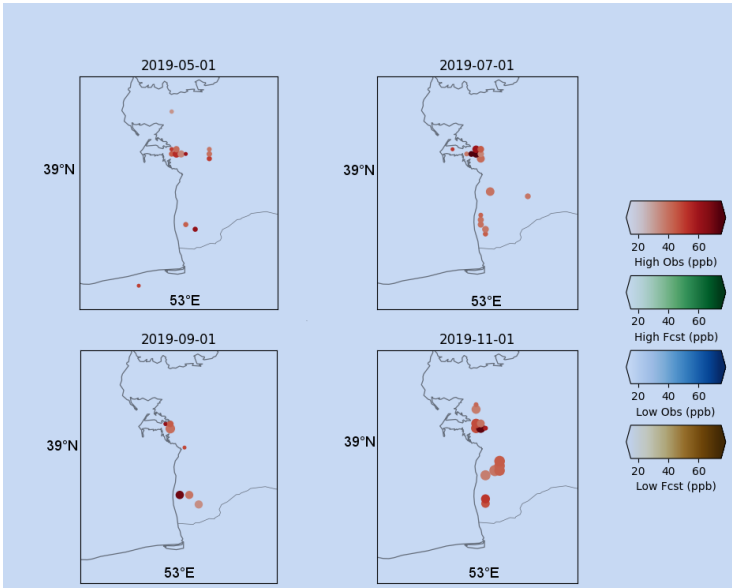
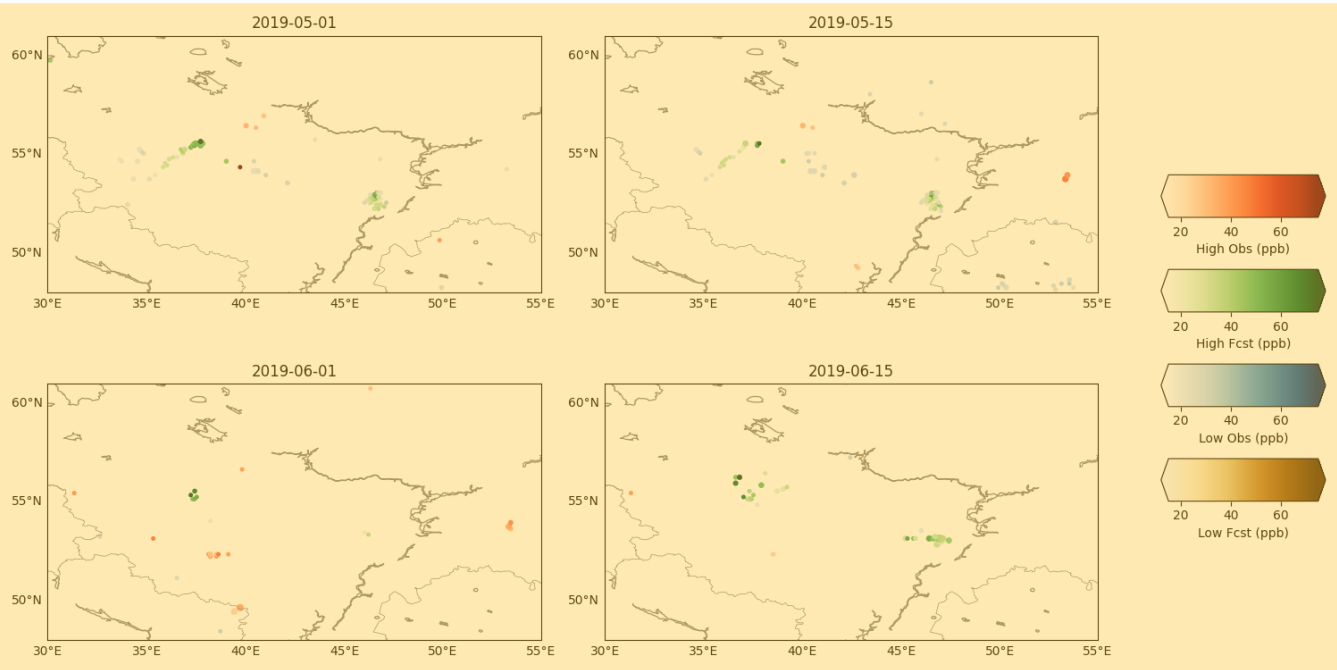


Figure 10. Outlier detection and classification over south western US region. Dates indicate the end date of the 30-day time window.

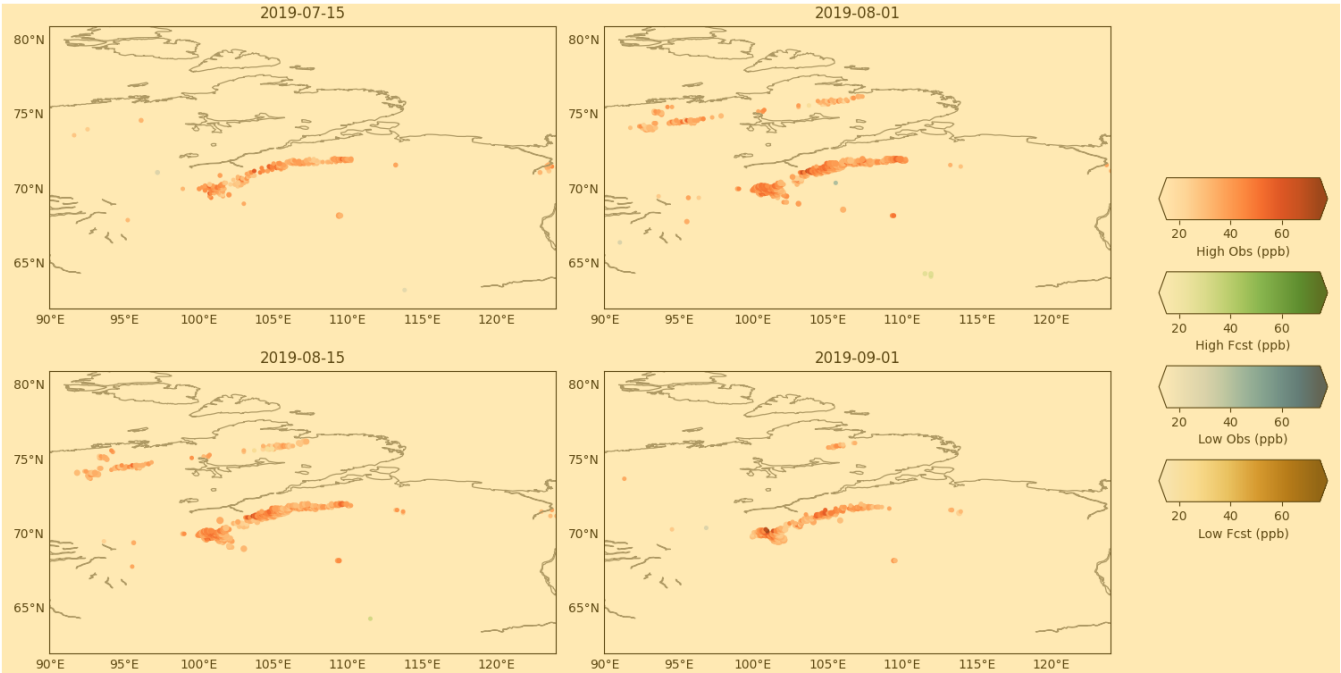


570

Figure 11. Outlier detection and classification over Turkmenistan. Dates indicate the end date of the 30-day time window.

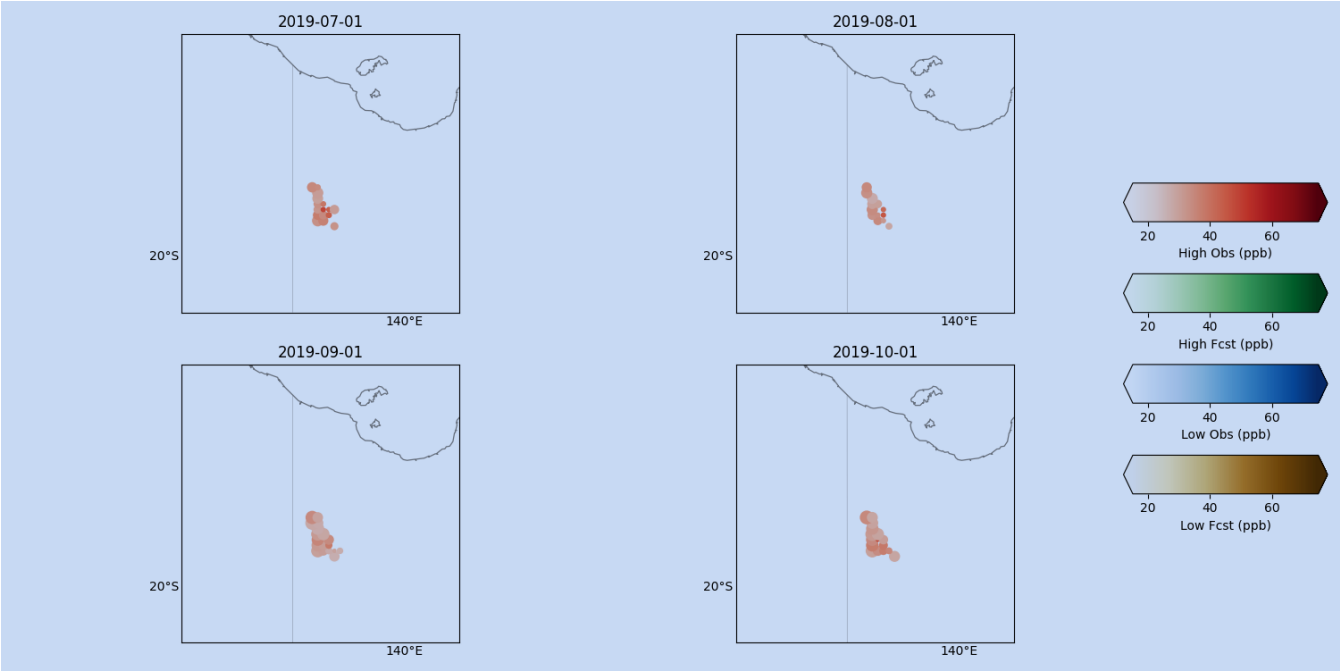


**Figure 12. Outlier detection and classification over western Russia. Dates indicate the end date of the 30-day time window.**



575

**Figure 13. Outlier detection and classification over Siberia. Dates indicate the end date of the 30-day time window.**



**Figure 14. Outlier detection and classification over Australia. Dates indicate the end date of the 30-day time window.**

