

Authors response to referee comments on manuscript “Satellite-derived methane hotspot emission estimates using a fast data-driven method” of Michael Buchwitz et al., MS No.: acp-2016-755

This document includes our point-by-point response to the reviews, a list of all relevant changes made in the manuscript, and a marked-up manuscript version.

Point-by-point response:

Our point-by-point response to the reviews has been submitted via the ACP website and is already online, see <http://www.atmos-chem-phys-discuss.net/acp-2016-755/#discussion> :

AC1: 'Authors response to comments from reviewer No. 1', Michael Buchwitz, 08 Feb 2017:

http://editor.copernicus.org/index.php/acp-2016-755-AC1.pdf?_mdl=msover_md&_jrl=10&_lcm=oc108lcm109w&_acm=get_comm_file&_ms=54349&c=118994&salt=554266951424103570

AC2: 'Authors response to comments from reviewer No. 2', Michael Buchwitz, 08 Feb 2017:

http://editor.copernicus.org/index.php/acp-2016-755-AC2.pdf?_mdl=msover_md&_jrl=10&_lcm=oc108lcm109w&_acm=get_comm_file&_ms=54349&c=118995&salt=621007627688187377

Nevertheless, these 2 documents with our answers are attached to this document (see following pages).

Marked-up manuscript version:

The Marked-up manuscript version is also attached to this document (at the end).

List of all relevant changes:

We have aimed at carefully addressing all referee comments (see our Point-by-point response). This resulted in several major and minor modifications which have been implemented for the revised version of our manuscript (see Marked-up manuscript version). The most relevant changes are:

- We have used an additional methane data set to address several of comments of the referees. This new high-resolution methane data set has been provided by Alexander J. Turner from the Harvard University, who has been added as a co-author. The new data set and its analysis is now described in a new Section 3.1, which also contain several (new) figures (Figs. 8-13).
- Based on the recommendation from one of the referees we have removed the Annex and moved the corresponding text and the figure to Sect. 3.

- Based on the comments from both referees we have added more information on our analysis w.r.t. the potential use of mean wind speed from meteorological data to improve the accuracy of our inversion method. This resulted in additional text in Sect. 3 and a new figure 7.
- Furthermore, there are many other changes: For example, we have improved text and figures at various places to eliminate typos and to improve explanations (as requested by the referees). We have also added an additional co-author from Univ. Leicester who is one of the data providers and helped to significantly improve the manuscript.

We conclude that addressing the referee comments resulted in a significantly improved version of our manuscript. We hope that the revised version of the manuscript meets the high standards of ACP.

Michael Buchwitz on behalf of all co-authors

On the following pages please see our response to the two referees and the marked-up manuscript version.

Reply to reviewer No 1

We thank the referee for carefully reading our manuscript and for providing a critical review. Below we are giving our point-by-point answers to each of the referee's comments and concerns. Addressing these comments, concerns and questions helped us to prepare a significantly improved version of our manuscript.

Q1: Referee:

Larger concerns: Methodological concerns: The authors need to explicitly state the necessary conditions for their approach to produce robust emissions estimates. What is the size of the region, size of xch4 signal, isolation from other sources, meteorological conditions, and emissions magnitude are necessary for the approach works?

Author's reply:

In the revised version of our manuscript we present additional investigations concerning the performance of our method. These investigations are based on a simulated high-resolution methane data set. Furthermore, we now better highlight already in the abstract limitations of our method. We show that our method typically provides a conservative estimate of the emissions, i.e., our emissions are typically underestimated. We better explain why our method tends to underestimate emissions. The large uncertainty of our method is reflected in quite large uncertainty estimates which are typically on the order of 100% for the source regions discussed in our manuscript. Our fast and simple method has been developed to obtain a reasonable estimate of the annual methane emission of a region which shows elevated methane relative to its surrounding region in maps of annually averaged satellite-derived XCH₄. When applying our method to multiple years of satellite data, the results will show, if elevated atmospheric methane is present in all years or not. If the methane is elevated in all years than this is very likely due to an "underlying" methane emission source (assuming that the satellite data do not have a "local bias"). We would not apply our method to situations, where this is not the case (although our method can be applied also to methane fields which are spatially constant/flat but in this case our method will deliver an estimated emission of zero together with a large error bar). There are no limitations w.r.t. the size of the region (as size is (approximately) considered by parameter L) or the size of the XCH₄ signal (as explained above) or the magnitude of the emission (which is unknown as the satellite only provides XCH₄). As shown in our manuscript we have not identified any conditions, where the method is shown to fail entirely but we recommend to be careful if the targeted source region is known to exhibit "pooling overnight" (more details on this aspect are given below) and/or for regions with complex topography (where, for example, methane can accumulate in valleys; these are however situations where all inversion methods will likely have severe difficulty). Our method assumes that the emission sources are homogeneously distributed in the targeted source region. We show in our manuscript that the estimated emissions are underestimated if this is not the case. Underestimation also results from sources located in the surrounding region. From all this we conclude that our method can be applied to all situations but that typically the emission will be underestimated, i.e., our estimates are conservative estimates. If the resulting emission is unexpectedly high, then this is a strong indication that the true emission is in fact higher than expected. In this case we recommend additional investigations, e.g., using a much more advanced (and computationally much more expensive) model than our simple mass balance approach (as explained in our paper).

Q2: Referee:

The method the authors employ is essentially a very simple mass balance approach where the elevated methane levels are attributed to a necessary flux assuming a constant wind speed (ventilation time). (note supplemental figure A1 is actually very helpful in explaining the method and should really be in the main text). However, I was quite surprised that the author's determined one single wind speed for use around the globe in this technique. In essence, this states the size of the XCH₄ enhancement seen in any hotspot is driven entirely by emissions, as wind speed is taken as globally constant. This would require significant justification, as we know this is not the case, and in particular, we know the manifestation of 'hot-spot' signal is often a consequence of meteorological conditions as well as emissions. For example, the Four Corners region discussed in the manuscript is known to exhibit pooling overnight, and part of what a midday satellite observations sees such an elevated signal is this meteorological dynamic (which is why in analyses such as the Kort et al., 2014 paper the winds are explicitly modeled). A region like North Dakota (discussed later), would have much higher wind speeds, and thus low XCH₄ enhancements would actually be linked with higher emissions. There is much more justification needed to justify a single wind speed for all regions, as this would be expected to produce answers that are strongly biased at each individual region.

Author's reply:

We are not assuming a constant wind speed. Wind speed is a parameter of our inversion model. However, we show that the consideration of (regionally varying) annual mean wind speed (as obtained from meteorological data) does not help to reduce systematic errors of our annual emissions as obtained from annually averaged XCH₄ (which is the goal of our method). We therefore use a constant wind speed but this is not because we assume this but because this results from our analysis, which shows that the use of spatially resolved annual mean wind speed (from meteorological data) does not help to improve our method. In the revised paper we present more details on this.

We removed Appendix A and present figure A1 now directly in our methods section.

“Pooling overnight” is in fact a concern for our method as this could result in a significant overestimation of the estimated emissions, which is what we aim to avoid as this would result in “false alarm” in comparison to emission inventories. For Four Corners we have no indication for overestimation of the Four Corners emissions as estimated with our method. In the revised version of our manuscript we investigated this using high-resolution methane simulations and found underestimation in line with the general characteristics of our method, which tends to underestimate emissions. We also present new results for several other regions (incl. California) and never found significant overestimation.

Q3: Referee:

The comparison with the global model at 6x4 does not really provide a satisfactory answer as to why one wind speed would be appropriate – this analysis would suggest that integrating globally using one wind speed does not produce a biased estimated, but for individual regions (the whole point of the analysis) there can and will be large bias errors. Furthermore, calibrating with a model that is at 6x4 degrees would then restrict the conclusion to analyses that are of the same resolution, as wind speeds in this type of box model setup will be rather different at a 6x4 degree region compared to a 1 or 1/2 degree region.

Author’s reply:

In the revised version of our manuscript we address this aspect by presenting additional results based on high resolution (< 1 deg) methane simulations.

Q4: Referee:

How can you justify applying the analysis on such different spatial scales – small in CA and Four Corners and large areas in Turkmenistan and Azerbaijan? All of which are different scales than the 4x6 degree model used for calibration?

Author’s reply:

In the revised version of our manuscript we address this aspect by presenting additional results based on high resolution (< 1 deg) methane simulations applied to small regions such as Four Corners and large (country-scale) regions such as large parts of California.

Q5: Referee:

Why are these four regions chosen only? There should be some discussion of what selection bias may be present and the reasoning behind the choice.

Author’s reply:

We selected these four regions because they show up as regions of elevated methane in the satellite data products (e.g., our Fig. 1) and because they are extensively discussed in the peer-reviewed literature (Central Valley, CA, and Four Corners) or other data sets exist which can be used for comparison (e.g., EDGAR for Turkmenistan and Azerbaijan). Initially our main motivation to develop our method was to at least roughly estimate Turkmenistan’s emissions as this country prominently shows up as a region of elevated methane in the satellite data. We also studied Azerbaijan because it is located close to Turkmenistan primarily to see how the estimated emissions of these two counties compare (as typically relative accuracy is better than absolute accuracy).

Q6: Referee:

Why have the author’s ignored two other regions in the US which they have published on previously (Schneising et al., 2014 for North Dakota and Texas)? It is true the recent publication by Peischl et al., 2016 JGR collected aircraft data in North Dakota and showed the Schneising 2014 paper was physically inconsistent with the atmospheric observations and emissions estimates (and that it is implausible that emissions all of a sudden declined in the face of increasing production between the Schneising and Peischl studies) but the authors here do not acknowledge that in citing the Schneising paper. One would suspect the discrepancy is because the Schneising paper relied on data from SCIAMACHY post-2009, which the author’s have deemed not robust in this analysis.

Given that this paper is discussing methane hotspots and an approach for quantifying a region and cites the Schneising paper, the North Dakota and Texas regions analyzed and published on previously by this group need to be addressed.

Author's reply:

As explained in our manuscript our method has been developed to obtain emission estimates for regions where satellite XCH₄ is clearly elevated compared to their surrounding areas. This condition is not met for the areas studied in Schneising et al., 2014 (see their Fig. 3). Furthermore, Schneising et al., 2014, used a method to minimize the potential impact of systematic errors of the used satellite product in later years by analysing differences of the satellite product between two 3-year time periods including years we are not analysing in our manuscript for reasons explained in our manuscript. We have therefore not ignored the two areas studied in Schneising et al., 2014, but we do not study them here because our method is not optimized to deal with them, in contrast to the method of Schneising et al., 2014.

We may misunderstand you but it appears that your comment suggests that it is “true” that the Schneising et al., 2014, results are “physically inconsistent” with other published observations. We do not agree with this as these other observations have not been made during the time period analysed by Schneising et al., 2014, but later and because such a statement needs to consider the uncertainty estimates as reported in Schneising et al., 2014. The uncertainty estimates as reported in Schneising et al., 2014, are large (nearly 70% 1-sigma) and statements w.r.t. consistency or inconsistency need to consider this. If one would do that one would find out that there is no inconsistency at a 5% (or even much higher) significance level. Concerning “that it is implausible that emissions all of a sudden declined” please see Schwietzke et al., Nature, 2016, showing that leakage rates of fugitive emissions decline with time.

Reference:

Schwietzke et al., Upward revision of global fossil fuel methane emissions based on isotope database, Nature, Vol. 539, 88-91, doi:10.1038/nature19797, 2016.

Q7: Referee:

What emission model underlies the model runs used for simulation? Is that also EDGAR?

Author's reply:

Several emission data bases are used as input as explained in Bergamaschi et al., 2009, but the anthropogenic a priori emissions are based on EDGAR. These emissions are however not used directly for the data set we used as this data set is based on forward modelling using optimized (a posteriori) emissions.

Q8: Referee:

Representation problems: The abstract reads as if the paper provides a satellite estimate for emission in different regions that are statistically significantly different from best-estimate inventories for different regions (for example lines 26-27 about the central valley in CA). This is actually quite misleading. This oversells the utility and robustness of the conclusions compared to the rather heavily caveat-ed discussion in the main text.

Author's reply:

For the revised version of our manuscript we have modified the abstract to also highlight the limitations of our method. We tried to eliminate all potential misunderstandings and clearly do not want to oversell our method and results.

Q9: Referee:

Firstly, the authors imply through much of the text the uncertainty in their approach is often 100% or greater, and this is neglected in the abstract. Secondly, the central valley CA result is much larger than EDGAR, but is rather close to the best estimates made in the literature from both other top-down studies, but also from other bottom-up inventories specifically made for California! The authors cite and acknowledge this in the main text, but the abstract sensationalizes a 6-9x discrepancy with EDGAR, which is known to fail at these spatial scales and really does not mean reported or inventoried emissions are too low. In general, comparisons with EDGAR are fine to do, but should not be overemphasized as being thought of as an accurate representation of emissions on small spatial scales (or representative of government reported inventories on this scale).

Author's reply:

For the revised version of our manuscript we have modified the abstract to also highlight the limitations of our method, e.g., by explicitly stating that uncertainty is often around 100%.

Q10: Referee:

Also, what is the overall utility of this method?

Author's reply:

The overall utility of our method lies in the fact that it provides at least rough estimates of emissions of source regions from large amounts of satellite data. As explained in our paper, we recommend further studies using more complex (and therefore computationally much more expensive) methods in case our method indicates significantly higher emissions compared to emission inventories. We write in our "Summary and conclusions" section: "More detailed assessments likely require the use of much more complex approaches compared to the simple method used in this study. Nevertheless, simple and fast approaches also have a role to play as they permit to perform quick assessments on possible discrepancies with respect to emission inventories or other data sets and can also be used for plausibility checks for more complex approaches".

Q11: Referee:

Where around the world can it be used?

Author's reply:

We have not identified any region where it cannot be used but please see also our detailed response to your first concern Q1.

Q12: Referee:

Which regions satisfy the criterion for usage (and what is the criteria)?

Author's reply:

We have not identified any region where our method cannot be used but please see also our response to your earlier questions but in particular our detailed response to your first concern Q1.

Q13: Referee:

What percentage of emissions can be tracked or observed this way? Need to see these numbers to understand the utility and impact of the approach.

Author's reply:

This question is difficult to answer but in general (as shown in more detail in the revised version of our manuscript) the local or regional emission sources must be quite strong, on the order of several 100 ktCH₄/yr. As also shown in the revised version of our manuscript there are many of these source regions in the USA and therefore likely also in many other parts of the world. However, we cannot give a reliable number in terms of percent of total methane emissions at this stage as this answer also depends on the spatial distribution of the sources (as our method requires relatively well isolated sources).

Q14: Referee:

Page 1 line 26-30: these concluding sentences in the abstract are misleading and overstated as discussed above.

Author's reply:

We have modified the abstract to highlight also limitations of our method and we explicitly mention that our uncertainty is on the order of 100%.

Q15: Referee:

Page 2 line 24-26: This is where the question of selection bias and why these regions comes into play.

Author's reply:

Please see our answers as given above on these aspects.

Q16: Referee:

Page 7: This would be where defining the location requirements (ie XCH₄ signal, size of area, wind speeds, emissions rate) would be valuable

Author's reply:

Please see our answers as given above on these aspects.

Q17: Referee:

Page 7 line 22: This is where the single wind speed is defined – see above for the concerns related to this approach.

Author's reply:

Please see our answers as given above on these aspects.

Q18: Referee:

Page 8 Line 8-9: This claim is really not robust. My assessment of these tests suggest that integrating globally the single, constant wind speed does not lead to a (large) bias, but for individual regions it will be strongly biased and this must be addressed and fixed.

Author's reply:

Please see our answers as given above on these aspects and please note that in the revised version of the manuscript we present additional investigations using high-resolution methane simulations and we apply our method to these simulations to obtain a better understanding of the performance of our method.

Q19: Referee:

Page 10 Line 17-18: Agreement here does not indicate the approach is sound and robust and therefore can safely be applied. There could easily be errors that cancel and lead to a coincidental agreement, or it could be that this one region is particularly good for this method.

Author's reply:

Yes, it is true that this could be a coincidental agreement. Therefore, we added for the revised version of the manuscript additional investigations using high-resolution methane simulations and we also added Four Corners to this extended assessment.

Q20: Referee:

Page 10 Line 23-26: This type of comparison is misleading – the under representation of EDGAR on this small regions is well known and defined previously, and emphasizing this gives an inaccurate impression that these high emissions are not accounted for properly in inventories (on this spatial scale EDGAR does not agree or match even the US inventory).

Author's reply:

EDGAR is an important, frequently used and carefully constructed data base (which does not mean that EDGAR is perfect) and we have not found statements in the peer-reviewed literature that EDGAR is inaccurate and therefore should not be used for applications like this.

Q21: Referee:

Page 12 line 29: This is a prime example of why the assumed constant velocity globally is of concern. Four Corners experiences even more pooling of emissions than the Central Valley, yet that isn't discussed. This problem or wind speed representation gives great concern to this approach.

Author's reply:

Please see our response as given above.

Q22: Referee:

Page 12: Far too much discussion and emphasis on the comparison to EDGAR for the central valley. The emissions being higher there than in EDGAR is well understood and documented from top-down and bottom-up emissions estimates in citations referenced, and is more an illustration of the failure of EDGAR on small, sub-national scales.

Author's reply:

Please see our response w.r.t. EDGAR as given above.

Q23: Referee:

Page 13 Line 21-22: If this is not a well-defined emission hotspot, why focus this study on this region?

Author's reply:

Please see our response w.r.t. Turkmenistan as given above.

Q24: Referee:
Page 14 Line 28: typo, “toinvestigate”

Author’s reply:
Many thanks. This has been corrected.

Q25: Referee:
Page 15 Line 7-9: This type of statement about concern about errors/problems in the approach needs to be addressed more explicitly in the abstract, and also should be addressed more quantitatively in sections such as this in the manuscript – what are the possible magnitudes of bias errors?

Author’s reply:
For the revised version this comment has been considered by modifying the abstract and by providing additional investigations using high-resolution methane simulations and more detailed discussion at several places.

Reply to reviewer No 2

First of all, we would like to thank the referee for carefully studying our manuscript and for providing critical comments and questions. Below we provide answers to each of these comments and questions. The referee's comments have resulted in a significantly improved version of our manuscript.

Q1: Referee:

First, the word “hotspot” in the title is somewhat misleading because the main result of this study is a regional or subregional (relatively large area) estimate of CH₄ emissions although pixels (but at coarse resolution) with large enhancements relative to surrounding pixels are identified. I strongly suggest that the authors remove the word “hotspot” from the title because this work essentially estimates emissions for source “regions”, for which many studies have already been doing using data from ground tower sites or aircrafts or remote sensing. The more accurate bottom-up inventories the authors cites (e.g., Jeong et al., 2014) can now identify hotspots with a much finer resolution. At the global scale, the source regions in this study may be considered hotspots, but those areas are really regions or subregions as shown in many previous studies.

Author's reply:

A hotspot does not have to be a very small area. It can be a large area, e.g., a country, see Oxford dictionary (<https://en.oxforddictionaries.com/definition/hotspot>): Definition: “A place of significant activity, danger, or violence.” Example sentence: “Madagascar is considered a biodiversity hot spot, an area that is home to great numbers of species and that is under constant assault from human activity”.

As we apply our method to areas of very different size and to areas emitting large amounts of methane, the term hotspot seems appropriate for this manuscript.

Q2: Referee:

Second, the authors try to match their satellite-based XCH₄ to another assimilated product. This is disappointing because the value of those satellite products for XCH₄ is significantly diminished as they are supposed to be used as independent retrievals of XCH₄. The authors need a clear justification for this. Please see the related specific comments below.

Author's reply:

In the revised version of the paper we will improve the description of how the methane data product used for “matching” has been generated. This product uses optimized emissions (obtained via assimilation) which are then used to generate the atmospheric methane concentration (via forward simulation). Therefore, the atmospheric concentrations are consistent with the emissions and this is exactly what we need for our purpose. The correctness of the emissions is not relevant for our application but what is relevant is that the link between emissions and concentrations is modelled as good as possible.

Q3: Referee:

Third, it looks like that the proposed method ends up with a simple linear scaling of satellite-derived XCH₄ to CAMS, in particular with a single parameter of V, which seems to be estimated as one value for the whole globe (as written it sounds like that; if not please clarify it).

Author's reply:

Yes, this understanding is correct. Please see also our detailed answer to the concerns of the other referee. In the revised version of our manuscript we will explain this better and will also add more details on our efforts to use meteorological data to improve on this. We also present an additional investigation using another model, which simulates methane at much higher spatial resolution compared to the used CAMS data set.

Q4: Referee:

Also, given the too large uncertainty for individual annual emission estimates, I wonder what value from this study can be added to the scientific community for regional GHG modeling.

Author's reply:

The purpose of our method is not to improve regional GHG modelling but to obtain very quickly (rough) methane emission estimates from (large amounts of) satellite data. The results can be used to identify regions where methane emissions are potentially higher than existing emission data bases suggest. We write in the “Summary and conclusions” section: “More detailed assessments likely require the use of much more complex approaches

compared to the simple method uses in this study. Nevertheless, simple and fast approaches also have a role to play as they permit to perform quick assessments on possible discrepancies with respect to emission inventories or other data sets and can also be used for plausibility checks for more complex approaches”.

Q5: Referee:

Page 4, Lines 26 - 28, the sentence needs to be revised because the authors are trying to say two conflicting things in the sentence, making it confusing. Also, I would recommend that the authors be more quantitative instead of saying "agree reasonably well". In terms of data gap, how SCIAMACHY and GOSAT are different, e.g., available data points/pixels at the annual scale?

Author's reply:

It is not entirely clear for us why this sentence is confusing. Taking into account the sampling of GOSAT and the fewer number of observations (a factor of 2-3 depending on product) compared to SCIAMACHY we were also surprised about the reasonably good agreement of the emissions as obtained from SCIAMACHY and GOSAT. At present this is a finding based on our end results. We have not aimed at explaining this in detail in terms of number of observations and required precision, accuracy and sampling as this is a complex topic requiring additional assumptions, e.g., on error correlations, and because we think that this is a bit out of scope and not mandatory for our study. Concerning "agree reasonably well": We have added more specific information in the revised version of the manuscript by adding in brackets: "(e.g., in terms of mean value and scatter of the resulting annual emission estimates)". The difference in terms of data gaps is addressed in our manuscript as we show for each investigated target region XCH₄ maps for SCIAMACHY and GOSAT in Sect. 4.

Q6: Referee:

Page 5, Line 10, I wonder if the authors considered the data scarcity (i.e., small number of data) for the annual averages in terms of uncertainty. For certain pixels, the # of available data would be too small while others have enough for averaging.

Author's reply:

We consider this by visual inspection of annual XCH₄ maps for each target region (examples are shown in Sect. 4 of our manuscript) and quasi-automatically by varying the size and shape of the surrounding region and by considering the standard deviation of the resulting emissions in our error estimate. We are confident that this is better than explicitly using the number of individual data points for our error estimate as this would require knowledge on error correlations (please note that from previous studies we know that improvement upon averaging will not follow a square root law).

Q7: Referee:

Page 5, Line 17, I wouldn't use the term "enhancement" because the surrounding region is not equal to the CH₄ "background" region, e.g., the Pacific region for the western US.

Author's reply:

In that paragraph we are not using the term "background". "Enhancement" is defined as source region XCH₄ minus surrounding region XCH₄. If this difference is positive then we have a positive enhancement, i.e., XCH₄ is higher over the source region compared to its surrounding area. In this context, it does not matter if the surrounding is equal to a true background or not. This only matters in terms of the accuracy of our method (e.g., additional sources in the surrounding region). In the revised version of our manuscript we have added more information on this accuracy aspect.

Q8: Referee:

Page 6, Lines 4 - 9, Looking at Eq. (1), the authors are trying to estimate emissions (flux) for the source region using ΔXCH_4 . But ΔXCH_4 is not exactly the local enhancement, but only the relative enhancement to the surrounding region, which itself has some local enhancements. This will lead to underestimation of the emissions for the source region.

Author's reply:

Yes, this is true and in the revised version of our manuscript we will highlight this aspect more prominently and provide more details.

Q9: Referee:

Page 7, Lines 9 - 10, The authors confirm my point about the underestimation when using Eq. (1). The authors state that "we aim at quantifying the impact of the choice of the surrounding region by varying its size and shape."

This makes it very hard to adopt the proposed method in other regions because it involves adjustments of size and shape, likely yielding multiple estimates and subsequently expanding the uncertainty.

Author's reply:

Yes, this expands the uncertainty as explained in our paper. As our emission result depends on the chosen surrounding region our uncertainty estimate contains an error term which reflects this. Please note that it is not hard to adopt our method to other regions. For the four source areas discussed in our manuscript we vary the surrounding region using a pre-defined automatic procedure which is the same for all four source regions (see page 10, top).

Q10: Referee:

Page 7, Lines 20 - 22, There are two important concerns about the method. First, I expected from the title that the satellite products would provide independent observations as in most of the top-down studies. It is not very satisfactory to try to match estimates from another product, i.e., CAMS. Also, from what is written here, I find that a single value for V needs a serious justification. Also, I am not convinced why CAMS should provide "true" estimates. Can the CAMS estimates be truly representative of any of the study sites/regions? How well are they compared with the estimates from previous studies for those source regions (maybe the word "true" may not be appropriate here; otherwise needs clarification).

Author's reply:

In the revised paper we will show additional results using another model which provides methane simulations at much higher spatial resolution. We also provide more details on why we are using a single value of V. If we apply our method to real satellite data, then the true emissions are not known. However, if we apply our method to simulations the underlying emissions are known. We refer to these emissions as "true emissions", meaning "known emissions". In the revised version of our manuscript we will explain this better.

Q11: Referee:

With respect to the optimization of V, this parameter optimization would be the key to this study. However, it seems that there is no explanation or consideration of the errors between the relationship between CAMS and XCH₄, which can be defined as: $CAMS = f(XCH_4, V) + err$, where the function f is likely a linear one and err is the irreducible error (e.g., mean 0, normal error). Here for correct estimation of V, we need some independent estimates for err, similar to a linear regression case with errors.

Author's reply:

It is true that parameter V is very important as the estimated emissions are directly proportional to it. It is also true that it would be good to have an independent assessment of the error of our estimated emissions. Therefore, we have added in the revised version of our manuscript additional assessment results using another model to compute emission biases for several source regions and we use the results to present more details on the performance of our method.

Q12: Referee:

Page 10, Lines 18 - 21, I differ with the authors. The too large uncertainty suggests that the method is not powerful. I would conclude that the only value of the satellite products used in this study is to provide auxiliary information derived from the columnaveraged XCH₄ which is linearly scaled to match another model product (rather than independent measurements).

Author's reply:

It is not clear for us what is wrong with what we write in lines 18 – 21. Our emission estimates are independent as they are derived from independent satellite retrievals. However, we agree that our large uncertainty limits the power of our method. In this context please see our response given above related to Q4.

Q13. Referee:

Page 11, 33-34, Again, the uncertainty is too large. When we think about hotspots, we expect relatively unambiguous isolation of emissions. The papers cited in this work already estimated emissions for the region with much better uncertainty. What policy makers need is identification of hotspots at the level of km scales and emission estimates for those small regions to mitigate sources from them. However, in this study, even the regional annual total yields very large uncertainty. Is there any way to reduce the uncertainty, even at the annual scale?

Author's reply:

Please see our response to your concern for Q12 and Q4. Our method is not accurate enough for “policy applications”. This would require a much more powerful method. As explained above (and in our manuscript) the main purpose of our fast method is to obtain rough estimates of emissions for source regions of interest using large amounts of satellite data. Via our method, source regions can be identified where emissions are potentially significantly underestimated in emission inventories. These regions can then be studied in detail using more powerful (but also computationally much more demanding) procedures.

Q14: Referee:

Table 3. EDGAR v4.2 happens to estimate the same Mt CH₄ for both Four Corners and the Central Valley?

Author’s reply:

We have checked this for both source regions and found that the correct value for the Central Valley is 0.19, not 0.17. Many thanks for pointing to this! We have corrected Tab. 3.

Q15: Referee:

Figure 1. The region needs to be defined more accurately. For example, the region defined as the Central Valley of California in Figure 1 includes Southern California, and is different from that in Table 2.

Author’s reply:

The purpose of Fig. 1 is to present an overview about the entire globe and to show where the investigated source regions are located and how XCH₄ looks like in these areas but also in their surrounding area. It is not the purpose of Fig. 1 to define exactly the source regions. The exact definitions of the source regions is given in Tab. 2.

Q16: Referee:

Figure 8 needs some improvements. First, the data points (circles) should match the years on the X-axis label that are represented. Is the “standard deviation” the standard deviation of 7 annual estimates, e.g., for the 2003 - 2009. If this is the case, standard deviation is not very useful. I would be more interested in knowing the overall mean estimate for the multi-year period and the uncertainty about the mean, e.g., during 2003 - 2009. When individual annual estimates have huge uncertainties associated, I don’t see the benefit of using standard deviation.

Author’s reply:

We have improved this figure by changing the annotation of the x-axis (Year -> Time[year]). We use standard deviation as this is a precisely defined quantity in contrast to the computation of the uncertainty about the mean as this would require sufficiently good knowledge of error correlations.

Q17: Referee:

Also, the 1-sigma uncertainty in estimated emissions for individual years overlap with the EDGAR estimate, making it hard to statistically evaluate EDGAR. Looking at this at face value, I am not sure if there is any statistical power in the proposed method to say about the regional emission, even at the annual scale.

Author’s reply:

Please see our response to these aspects (large uncertainty, power of our method) as given above.

Satellite-derived methane hotspot emission estimates using a fast data-driven method

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Abstract. Methane is an important atmospheric greenhouse gas and an adequate understanding of its emission sources is
20 needed for climate change assessments, predictions and the development and verification of emission mitigation strategies. Satellite retrievals of near-surface-sensitive column-averaged dry-air mole fractions of atmospheric methane, i.e., XCH₄, can be used to quantify methane emissions. [Maps of time-averaged satellite-derived XCH₄ show regionally elevated methane over several methane source regions. In order to obtain methane emissions of these source regions we use](#) ~~Here we present a~~ [a simple and fast data-driven method to estimate emissions of methane hotspots from satellite derived XCH₄ maps to](#)
25 [estimate annual methane emissions and corresponding 1-sigma uncertainties directly from maps of annually averaged satellite XCH₄.](#) ~~From theoretical considerations we expect that our method tends to underestimate emissions. When applying our method to high-resolution atmospheric methane simulations, we typically find agreement within the uncertainty range of our method (often 100%) but also find that our method tends to underestimate emissions by typically about 40%. To what extent these findings are model-dependent needs to be assessed.~~ We apply ~~our~~^{this} method to an ensemble of [satellite](#)
30 XCH₄ data products consisting of two products from SCIAMACHY/ENVISAT and two products from TANSO-FTS/GOSAT covering the time period 2003-2014. We obtain annual emissions of the source areas Four Corners in the southwestern USA, for the southern part of Central Valley, California, and for Azerbaijan and Turkmenistan. We find that our estimated emissions are in good agreement with independently derived estimates for Four Corners and Azerbaijan. For the Central Valley and Turkmenistan our estimated annual emissions are higher compared to the EDGAR v4.2
35 anthropogenic emission inventory. For Turkmenistan we find on average about 50% higher emissions with our annual

emission uncertainty estimates overlapping with the EDGAR emissions. For the region around Bakersfield in the Central Valley we find a factor of ~~56-89~~ higher emissions compared to EDGAR albeit with large uncertainty. Major methane emission sources in this region are oil/gas and livestock. Our findings corroborate recently published studies based on aircraft and satellite measurements and new bottom-up estimates reporting significantly underestimated methane emissions

5 of oil/gas and/or livestock in this area in EDGAR inventories.

1 Introduction

Methane (CH₄) is the second most important human-emitted greenhouse gas ~~—directly after carbon dioxide—~~ and increases in its atmospheric abundance contribute significantly to global warming (IPCC, 2013). Accurate knowledge of its sources and sinks and the origins of any changes are needed for the accurate prediction of future climate change, the attribution of change, and the development of mitigation strategies. ~~;~~ ~~However, but~~ our current knowledge about the various natural and anthropogenic methane sources and sinks ~~is inadequate~~~~has significant gaps~~ (e.g., Rigby et al., 2008; Dlugokencky et al., 2009; IPCC, 2013; Kirschke et al., 2013; Houweling et al., 2014, 2017; Nisbet et al., 2014; Jeong et al., 2014; Alexe et al., 2015; [Jacob et al., 2016](#); Schaefer et al., 2016; Miller and Michalak et al., 2016).

Near-surface-sensitive satellite observations of atmospheric methane have been used in recent years to obtain quantitative information on methane emissions (e.g., Alexe et al., 2015; Bergamaschi et al., 2007, 2009, 2013; Bloom et al., 2010; Turner et al., 2015, 2016; Fraser et al., 2013; ~~;~~ ~~Monteil et al., 2013; Cressot et al., 2014; Wecht et al., 2014a, 2014b; Kort et al., 2014;~~ [Jacob et al., 2016](#); [Houweling et al., 2017](#)). Nevertheless, there are still many important aspects, ~~;~~ which need further investigation. For example, ~~aspects related to~~ the recent renewed methane growth (e.g., Houweling et al., 2014) ~~needs to be~~ ~~an~~ ~~unambiguously~~ ~~explained~~ ~~ration~~, and better knowledge ~~of about aspects related to specific but~~ evolving man-made emission sources (e.g., Schneising et al., 2014) is required.

Several important issues for the future management and mitigation of methane emissions ~~are not yet resolved~~ ~~have not yet~~ ~~been addressed~~ adequately, e.g., ~~the methods such as verification to verify of~~ emission inventories and reported emissions per region (country down to city scale) (e.g., Ciais et al., 2014). The latter aspect ~~has been addressed for future satellite missions, especially for CO₂ in the context of the proposed was studied in the development of the~~ CarbonSat mission (Bovensmann et al., 2010; Velazco et al., 2011; Buchwitz et al., 2013; Pillai et al., 2016) ~~for CO₂~~ using performance assessments based on simulated satellite observations (ESA, 2015) but so far only few studies have been published using real satellite data (e.g., Wecht et al., 2014a; Turner et al., 2015, 2016, for USA methane emissions). ~~In this study we report an approach~~ ~~Here we report on an attempt~~ to use satellite methane retrievals to estimate ~~the~~ methane emissions of ~~the two~~ ~~countries~~ Azerbaijan and Turkmenistan, which are both important oil and gas producing countries, and also apply our method to two regions in the USA. ~~All four studied regions show methane enhancements relative to their surrounding area in satellite derived XCH₄ maps.~~

This manuscript is structured as follows: In Sect. 2 we introduce briefly the satellite data which have been used in this study. In Sect. 3 we describe the analysis method developed to derive methane emissions of (relatively) well localized areas

~~emission hot spots~~ from time-averaged satellite XCH₄ retrievals. The results as obtained from the satellite retrievals are presented and discussed in Sect. 4 and a summary and conclusions are given in Sect. 5.

5

2 Satellite data

During recent years the retrieval of near-surface-sensitive column-averaged dry-air mole fractions of atmospheric methane (CH₄) and carbon dioxide (CO₂), i.e., XCH₄ and XCO₂, from the satellite sensors SCIAMACHY (Burrows et al., 1995; Bovensmann et al., 1999) onboard ENVISAT and TANSO-FTS onboard GOSAT (Kuze et al., 2009, 2016) significantly
10 evolved and improved (e.g., Buchwitz et al., 2015, 2016a, 2016b; Butz et al., 2011; Dils et al., 2014; Frankenberg et al., 2011; Parker et al., 2011, 2015; Schneising et al., 2011, 2012, 2014; Yoshida et al., 2013).

For this study we use the latest data sets of XCH₄ retrievals from SCIAMACHY and GOSAT as generated by different research teams of the GHG-CCI project (Buchwitz et al., 2015) of ESA's Climate Change Initiative (CCI, Hollmann et al.,
15 2013). The four satellite XCH₄ products used for this study are publicly available and have been obtained from the GHG-CCI website (<http://www.esa-ghg-cci.org>; "latest data sets" refers to data access mid 2016; new versions are in preparation and are planned to be released in March 2017), where also detailed documentation is available (e.g., Algorithm Theoretical Basis Documents (ATBDs), Comprehensive Error Characterization Reports (CECRs), Product Validation and Intercomparison Report (PVIR, Buchwitz et al., 2016a)).

20

Table 1 presents an overview about the four XCH₄ satellite data products used in this study. As can be seen, these comprise two SCIAMACHY XCH₄ data products retrieved with the WFMD (Buchwitz et al., 2000; Schneising et al., 2011, 2012, 2013) and IMAP (Frankenberg et al., 2005, 2006, 2008a, 2008b, 2011) retrieval algorithms, i.e., the GHG-CCI products CH₄_SCI_WFMD and CH₄_SCI_IMAP. In addition, we use the two GOSAT products CH₄_GOS_OCPR (Parker et al.,
25 2011, 2015) and CH₄_GOS_SRFP (Butz et al., 2011, 2012). The XCH₄ "full physics" (FP) retrieval algorithm used to generate the latter product is also known as "RemoTeC" and the algorithm to generate the CH₄_GOS_OCPR product is the University of Leicester XCH₄ "CO₂ proxy" (PR) algorithm. The two SCIAMACHY XCH₄ algorithms are also "proxy" algorithms. Here, the XCH₄ product is obtained by computing the ratio of the retrieved methane column and the simultaneously retrieved CO₂ column multiplied by a correction factor for XCO₂ variations using a CO₂ model (Frankenberg et al., 2005). The FP algorithm does not require this CO₂ correction as XCH₄ is retrieved directly, which is an advantage
30 compared to PR algorithms. However, each algorithm has different strengths and weaknesses advantages and disadvantages. An advantage of the XCH₄ PR algorithms is that atmospheric light path related errors arising from imperfect knowledge

~~of due to not perfectly considered~~ wavelength dependent scattering by aerosols and clouds largely cancel in the CH₄ to CO₂ column ratio. This source of error is consequently less of a problem for PR algorithms compared to FP algorithms, which require more complex radiative transfer modelling and stricter quality filtering compared to the PR products (see also Schepers et al., 2012, for PR and FP algorithms and corresponding data products but also Buchwitz et al., 2015, 2016a, 2016b). As a consequence, FP data products are typically much sparser compared to PR products, but are independent of the CO₂ model used.

The latest validation results for the GHG-CCI XCH₄ data products are presented and discussed in Buchwitz et al., 2016a. These were obtained by comparison of the satellite retrievals with ground-based XCH₄ observations of the Total Carbon Column Observing Network, TCCON (Wunch et al., 2011, 2015). As shown in Buchwitz et al., 2016a, the GOSAT XCH₄ products are very stable, i.e., do not show any significant trend of the difference with respect to TCCON. For SCIAMACHY the situation is more complex due to potential detector problems in later years resulting in larger noise but also bias issues, depending on retrieval algorithm. For example, as shown in Buchwitz et al., 2016a, the IMAP product suffers from a bias (a discontinuity in XCH₄) in 2010. For this reason, we decided to restrict the use of the SCIAMACHY products in this study to the period 2003 – 2009. The achieved single measurement precision (random error) for SCIAMACHY XCH₄ is in the range 30-80 ppb (2-5%) depending on time period and product and approximately 16 ppb (~1%) for GOSAT. Systematic errors (“relative accuracy” or “relative bias”) are around 10-15 ppb (~0.6%) for SCIAMACHY and approximately 6 ppb (~0.3%) for GOSAT.

Annual average composite maps of the four data products are shown in Figs. 1 and 2. Figure 1 shows year 2004 SCIAMACHY XCH₄ at 0.5° x 0.5° resolution as retrieved using the WFM-DOAS (WFMD) algorithm (Schneising et al., 2011). Also shown are zooms for the three target regions investigated in this study. Figure 2 shows year 2004 SCIAMACHY IMAP-DOAS (IMAP) XCH₄ and year 2010 XCH₄ as retrieved using the two GOSAT algorithms. As can be seen, the spatial coverage of the GOSAT products is quite sparse. A single GOSAT observation requires more time (4 seconds) compared to a SCIAMACHY observation (typically 0.25 seconds for the spectral regions relevant for this study) and, therefore, GOSAT provides less observations in a given time period than SCIAMACHY. On the other hand, the GOSAT ground pixel size is smaller (10 km diameter) compared to SCIAMACHY (approximately 30 km along track times 60 km across track), which results in a higher fraction of cloud free observations for GOSAT. Furthermore, SCIAMACHY is in nadir (downlooking) observation mode only about 50% of the time. Overall the total number of quality filtered observations as contained in the data products is larger for SCIAMACHY compared to GOSAT. Furthermore, the spatial sampling of GOSAT comprises non-contiguous ground pixels, which results in large data gaps (even in yearly averages). Consequently, GOSAT is typically (i.e., in normal observation mode) not optimal for small-scale hotspot applications but as shown in this manuscript, GOSAT provides results for the selected source regions which agree reasonably well with the results obtained using SCIAMACHY

(e.g., in terms of mean value and scatter of the resulting annual emission estimates). In the remainder of this manuscript we focus on obtaining methane emission estimates for the source areas shown in Fig. 1.

3 Analysis method

In this section we describe the analysis method used to obtain methane emission estimates for source regions such as those shown in Fig. 1, i.e., for regions showing elevated methane relative to their surrounding area in time-averaged satellite-derived XCH₄ maps.

The satellite XCH₄ input data used in this study are the GHG-CCI Level 2 (i.e., individual ground-pixel observations) data products as described in the previous section (see also Tab. 1). The first step in the analysis comprises gridding (averaging) these products using a regular latitude/longitude grid (here: 0.5°x0.5°) to obtain maps of annual averages (see Figs. 1 and 2). These mapped XCH₄ products are then used in this study for further analysis.

The second step comprises the definition of a source region and a surrounding (or background) region. The latter is an extended region surrounding the source region (specific examples are shown in Sect. 4).

The third step comprises the determination of the methane enhancement over the source region relative to its surrounding area, ΔXCH_4 . This methane enhancement is computed by subtracting the mean value of XCH₄ in the surrounding region from the mean XCH₄ value over the source region.

To reduce potential effects related to a location dependent weighting of tropospheric and stratospheric contributions on XCH₄ (~~as note that~~ mean stratospheric CH₄ mixing ratios are typically lower compared to tropospheric mixing ratios) we apply a correction called “elevation correction” (EC) similar ~~to that as also~~ described in Kort et al., 2014, and Turner et al., 2016 (and implicitly also applied in Schneising et al., 2014). The purpose is to correct for satellite XCH₄ variations due to variations of surface elevation/pressure and tropopause height. The corrected XCH₄ is obtained from the original satellite XCH₄ retrievals by adding 7 ppb per 1 km surface elevation increase relative to mean sea level. For surface elevation we use a surface elevation map (also 0.5°x0.5°) ~~calculated using based on~~ the GTOPO30 Digital Elevation Model (DEM) (obtained from <https://lta.cr.usgs.gov/GTOPO30>). The value of 7 ppb/km has been obtained by fitting a linear function to pairs of uncorrected original XCH₄ and corresponding surface elevation. We found that the exact value depends somewhat on region, time period and satellite data product but is typically within 7 +/- 2 ppb/km. We found that applying EC typically results in similar or somewhat lower emission estimates compared to inversions where this correction is not applied.

The fourth step comprises the conversion of the methane enhancement over the source region, ΔXCH_4 , to a source region emission estimate (E_e ; unit: $MtCH_4/year = TgCH_4/year$) using conversion factor CF:

$$E_e = \Delta XCH_4 \cdot CF. \quad (1)$$

5

This ~~assumes basic idea is~~ that a relatively well isolated emission source (or region of emission sources) will result in an XCH_4 enhancement, ΔXCH_4 , in an area at and around the emission hotspot relative to its surrounding, i.e., that there will be a spatial correlation between a local emission and a local XCH_4 enhancement (compare also the two maps shown in Fig. 43 top left and top right, which will be discussed in detail below).

10

The conversion factor CF in Eq. (1) is computed as follows (see also below when discussing Fig. 3, which illustrates our method Annex A for additional explanations):

$$CF = M \cdot M_{exp} \cdot L \cdot V \cdot C. \quad (2)$$

15

Here M is a constant conversion factor ($5.345 \cdot 10^{-9} MtCH_4/km^2/ppb$) needed to convert a methane mole fraction change to a methane mass change per area for standard conditions, i.e., for surface pressure $p_{surf} = 1013$ hPa. M_{exp} is a dimensionless factor used to ~~approximately~~ correct for the actual mass (mass M_i of the i-th grid cell). It is calculated using the surface elevation map also used for the determination of the elevation correction (EC) as described above:

20

$$M_{exp} = \frac{\langle M_i \rangle}{M} \approx \frac{\langle p_i \rangle}{1013.0} \approx \langle e^{-z_i/H} \rangle_i. \quad (3)$$

Here p_i is the surface pressure of the i-th grid cell (in hPa) and z_i is the surface elevation of the i-th grid cell (in km), H is the assumed scale height (8.5 km) and $\langle \cdot \rangle$ and $\langle \cdot \rangle_i$ denotes averaging over all grid cells of the source region. As shown below, the uncertainty of our method is not dominated by the approximation used to compute M_{exp} (namely the use of surface pressure or elevation rather than actual mass).

The dimension of the remaining factor ($L \cdot V \cdot C$) is $km^2/year$, i.e., area divided by time or length times velocity and can be interpreted as the effective methane emission accumulation time of air parcels travelling over the source region area or the effective velocity V of air parcels travelling an effective length L over the source region. ~~In this study Here we use the latter interpretation, i.e.,~~ L is length (in km) and V is velocity (in km/year). We compute L as the square root of the (pre-defined) source area.

30

Factor C is dimensionless and in this study we use $C = 2.0$. This choice is motivated using the simple model of an air parcel travelling with constant horizontal wind speed V over a homogeneous source region of length L accumulating methane during an accumulation time $\tau = L/V$ (see Annex A). When leaving the source area, the methane enhancement of the air parcel, i.e., the concentration difference after and before entering the source region, is twice the mean methane enhancement over the source region due to the assumed linear increase of the methane enhancement of the air parcel when travelling over the source region (see Annex A). Our method basically assumes that the emission of the source region only results in a XCH_4 enhancement over the source region.

Figure 3A1 illustrates the basic idea of the methane emission estimation method explained in Sect. 3. In particular it is illustrated how the observed methane enhancement over the source region (region A in Fig. 3A1 (a)), ΔXCH_4 , is related to the source region emission (E , in mass per time), wind speed magnitude V , and length of the source region. The source region shown here is a rectangle of area $A = L_x L_y$, where wind speed is in the x -direction. Note that length L as given in Eq. (2) corresponds to length L_y of Fig. 3A1.

The computation of the methane mole fraction enhancement over the source region relative to its surrounding, ΔXCH_4 , is computed (see also Sect. 3 and Sect. 4) by subtracting the mean value of XCH_4 in the surrounding region (region B in Fig. 3A1 (a)) by the mean value of XCH_4 over the source region (region A in Fig. 3A1 (a)). It is assumed that the surrounding region does not contain any (significant) emission sources and that neglecting atmospheric methane enhancements in the surrounding area due to outflow from the source region into the surrounding region (region C in Fig. 3b) can be neglected (region C in Fig. A1 (a)) the resulting error is small if; note that this requires that region B is much larger than region C). As a consequence, the computed mean value of XCH_4 in the surrounding is typically overestimated and, therefore ΔXCH_4 and the computed methane emission is too low, i.e., the estimated emission is (typically) underestimated a conservative estimate.

Note that the method described in Sect. 3 here and used in Sect. 4 is only applied to time averages of atmospheric XCH_4 to obtain time averaged emissions. This typically means that meteorological situations vary significantly during the selected time period (including large wind speed and wind direction variations) so that detailed structures of the atmospheric methane emission “plumes” originating from local emission sources largely average out resulting in enhanced atmospheric methane over the source region. It needs to be pointed out that Note that Figure: 3A1 (b) only illustrates a “snapshot” in time but not the average over a range of wind speeds and wind directions (assumed to be reasonably well approximated by the localized enhancement shown in Fig. 3A1 (a)).

ΔXCH_4 also depends on the (size and shape) of the surrounding region. However, also the surrounding area may contain elevated XCH_4 from sources located in the surrounding area or from methane inflow from other regions into the surrounding area (including the source region). As our method neglects this, our method tends to underestimate the emission of the

~~source region.~~ As explained below, we aim at quantifying the impact of the choice of the surrounding region by varying its size and shape.

Our method (Eqs. (1) and (2)) assumes a homogeneous distribution of emission sources (“flat source”) within the chosen source region (Fig. 3). However, one would expect that due to atmospheric transport (advection and mixing) the observed atmospheric methane (e.g., for annual averages) typically covers a larger area than the underlying emission region(s). As can be concluded from Eqs. (1) and (2) our method results in an underestimation of the emissions, when this assumption is not valid. This can be seen as follows: Let’s start with a situation, where our assumption is valid, i.e., there is a single homogeneous emission source region and its area is identical with the source region used for our analysis. In this case we obtain a certain value for ΔXCH_4 and convert it to an estimated emission E_e using conversion factor CF. Now let’s assume that the surrounding area does not contain any emission sources. If we now extend the size of the source region (region A in Fig. 3) but do not change the outer boundary of the surrounding region (region B in Fig. 3), the true emission of the extended source region would be the same as before (as no emission sources are added, when the source region is extended) but the resulting methane enhancement (ΔXCH_4) will decrease as the atmospheric methane enhancement will typically be the smaller the larger the distance from the source is. A smaller ΔXCH_4 will result in a smaller value of the estimated emission, E_e (see Eq. (1)). Conversion factor CF increases with increasing source region, i.e., the estimated emission not only depends on ΔXCH_4 but also on the size of the source region via CF. The problem is that the increase of CF is only proportional to L, i.e., to the square root of the source area, whereas the decrease of ΔXCH_4 may be proportional to the source area ($= L^2$). As a result, one would expect an underestimation of the estimated emission. This underestimation increases (gets worse) the more inhomogeneous the true emission sources are distributed within the investigated source region (an illustration is given below when discussing Figs. 9 and 10).

The value of V has been obtained by “calibrating” our method using global methane data sets obtained from the Copernicus Atmosphere Monitoring Service (CAMS, <https://atmosphere.copernicus.eu/>). Specifically, we use CAMS *a posteriori* methane emissions and corresponding atmospheric methane version v10-S1NOAA as generated via the TM5-4DVAR assimilation system assimilating National Oceanic and Atmospheric Administration (NOAA) CH₄ surface observations (an earlier version of this method and resulting data products is described in Bergamaschi et al., 2009). The CAMS data set used is based on forward modelling for the computation of atmospheric methane based on prescribed (but optimized) emissions. This is important as the calibration of our method requires atmospheric methane consistent with the underlying methane emissions. Based on this data set we computed annual emissions and corresponding annual XCH₄ at the original CAMS data set resolution of 6° longitude times 4° latitude. The corresponding maps for the year 2003 are shown in Fig. 43 (top row).

The CAMS year 2003 XCH₄ map shown in Fig. 43 top left has been used to derive methane emissions using Eq. (1) and varying parameter V (the only free parameter of our model) until the mean difference between our estimated emissions and

the “true” CAMS emissions is zero. We found that this is the case for $V = 1.1$ m/s (converted to km/year). The term “true” as used here (and below) does not imply that the CAMS emissions are perfect, i.e., free of errors. It simply means that these are the emissions which correspond to the atmospheric methane we use to calibrate our method, i.e., the atmospheric concentrations are computed using these emissions. What matters for our application is that we have a “good enough”
5 modelling of the relationship between emissions and resulting atmospheric concentrations.

~~We found that this is the case for $V = 1.1$ m/s (converted to km/year).~~ The resulting map of retrieved emissions using $V = 1.1$ m/s is shown in Fig. 43 bottom right. This map has been obtained using an automatic procedure: For all CAMS $6^\circ \times 4^\circ$ grid cells (except for the ones at the border) the XCH_4 value of this grid cell has been obtained and is interpreted as a potential source region value. The neighboring cells define the surrounding (background) of the potential source region and its XCH_4 mean value and standard deviation has been computed. A methane enhancement, ΔXCH_4 , has been computed as “source minus background value” (here “background” refers to the mean XCH_4 value in the surrounding region) as described above. If the resulting ΔXCH_4 value is larger than 0.5 times the standard deviation of the XCH_4 values in the surrounding, then the corresponding cell is flagged as a methane “hotspot cell” and its ΔXCH_4 value is converted to an emission using the
10 approach described above (Eq. (1)). The corresponding results are shown as map in Fig. 43 bottom right and can be compared with the “true” emission map shown in Fig. 43 top right. As can be seen in Fig. 43, $N = 125$ hotspot cells have been found using the described procedure.
15

Figure 43 bottom left shows x-y plots of estimated emissions versus “true” (i.e., CAMS) emissions (top) and estimated minus true emissions versus true emissions (bottom). The mean difference “estimated-true” is 0.00 MtCH₄/year (this must be the case as $V = 1.1$ m/s has been determined by minimizing this difference). The standard deviation of the difference is 0.59 MtCH₄/year, the linear correlation coefficient R is 0.81 and the red line shows the resulting line from a linear fit. As can be seen, the (red) line originating from the linear fit has a positive slope but does not perfectly agree with the (green) 1:1 line (our single parameter model does not permit to also optimize the slope of the fitted line).
20

Figure 54 is similar as Fig. 43 but shows results for the year 2012. Here the difference “estimated-true” is not exactly zero but 0.01 MtCH₄/year. In contrast to Fig. 43, V has not been fitted. Instead, the pre-defined value of $V = 1.1$ m/s has been used. Figure 54 shows very similar “estimated-true” differences compared to Fig. 43. This indicates/demonstrates that the effective wind speed V as obtained from year 2003 data is valid also for other years.
25

The results shown in Figs. 43 and 54 are combined in the single Fig. 65. As can be seen from Fig. 65 (top), the overall correlation of the retrieved and true emissions is 0.81, the mean difference (estimated minus true) is 0.00 MtCH₄/year and the standard deviation of the difference is 0.53 MtCH₄/year. As explained, these results have been obtained using constant values for wind speed fit-parameter V ($= 1.1$ m/s) and correction factor C ($= 2.0$) (Eq. (2)). Several attempts have been
30

undertaken in order to find out if the use of regionally and/or time dependent V or C values can reduce the difference of the estimated and the true methane emission, however (so far) without success. For example, it has been investigated if the emission difference is correlated with mean wind speed (using ECMWF ERA Interim data obtained from www.ecmwf.int/, Dee et al., 2011) but no significant correlation between emission error and spatially resolved annual mean wind has been found. Figure 7 illustrates this using annual mean wind speed at 900 hPa. As can be seen, there is essentially no correlation between emission error and mean wind speed (R = 0.049). Similar results have been obtained for other pressure levels (e.g., R = -0.036 for 800 hPa and R = 0.254 for the lowest ECMWF ERA Interim model level). This indicates that the use of mean wind speed (from meteorological data) does not help to improve the accuracy of our method. Future studies will show to what extent our method can be improved (or not). The year-to-year variation~~time-dependence~~ of the estimated annual emission, E_e , for a given satellite XCH₄ product is therefore ~~nearly~~ entirely driven by the satellite-derived methane enhancement, ΔXCH_4 , as parameters V and C are constant.

Finally, the (1-sigma) uncertainty of E_e has been estimated. This has been done as follows: Figure 65 also shows the emission difference (“estimated minus true”; see middle and bottom panels) as a function of the estimated emission. Figure 65 middle also shows (in red) the corresponding mean values (crosses) and standard deviations (vertical bars) for several emission bins (non-equidistant to ensure a sufficiently large number of data points within each bin). Also shown in Fig. 65 (middle and bottom) are dotted red lines computed as $f(E_e) = 0.3 + 0.5 \cdot E_e$. This function and its parameters has been chosen such that the red vertical bars (1-sigma range) are located within the range defined by $f(E_e)$, i.e., most of the emission differences are located within $\pm f(E_e)$ (Fig. 65 middle). Therefore, $f(E_e)$ is a reasonable description of the 1-sigma uncertainty of the estimated emissions. Based on this it is concluded that the 1-sigma uncertainty of the estimated emission due to uncertainty of the overall conversion factor (CF) can be well described using this formula:

$$\sigma_{CF} = 0.3 + 0.5 \cdot E_e. \quad (4)$$

Here the units of σ_{CF} and E_e are MtCH₄/year. The total uncertainty, σ_{tot} , consists of the uncertainty of the conversion factor, σ_{CF} , and the uncertainty of the obtained methane enhancement, $\sigma_{\Delta XCH_4}$, as obtained from the satellite data (see Eq. (1)). The latter is assumed to be dominated by methane variations in the surrounding area (primarily because the surrounding region may contain regions of elevated methane due to sources located outside the ~~source~~target region). This contribution to the total uncertainty is estimated by varying the size of the surrounding region (see following section). The total uncertainty is computed as follows:

$$\sigma_{tot} = \sqrt{\sigma_{\Delta XCH_4}^2 + \sigma_{CF}^2} \quad (5)$$

The method described in this section has been applied to the described SCIAMACHY and GOSAT XCH₄ data products and for each of the pre-defined source regions annual average emissions and their uncertainties have been obtained for all products. The results are presented in Sect. 4. ~~the following section~~ Before the method is applied to real data it is relevant to carry out some additional investigations using simulations as in this case the “true emissions” are known. For this purpose, a high-resolution methane data set is used to investigate how well the inversion method performs when using a different model, which simulates atmospheric methane at much higher spatial resolution than the model described and used for the results presented in this section. The high-resolution results are presented in the following sub-section 3.1.

3.1 Performance of inversion method as applied to simulations of high-resolution methane

In order to test the inversion method using a methane data set at higher resolution, simulated atmospheric methane concentrations using posterior methane emissions from Turner et al., 2015, have been used. The spatial resolution of this data set is 0.5° latitude times 0.667° longitude and it covers North America. The methane concentrations have been computed with GEOS-Chem. This data set is referred to as GCT15 in this manuscript. It covers one year (2010) and consists of methane emissions and corresponding atmospheric concentrations on the same spatial grid.

Figure 8 shows (around noon) annually averaged GCT15 XCH₄ over the USA. As can be seen, there are several regions, where methane is significantly enhanced compared to their surrounding areas. However, one would see even more “emission hotspot areas”, when zooming into this map and when using an appropriate color scale for the zoomed-in regions.

This is demonstrated in Fig. 9a focusing on central California (a region discussed in detail in Sect. 4). As can be seen, there is a region of clearly elevated methane (red color) located approximately between the two cities Modesto and Merced (not shown). This region has been selected as a source region shown as polygon (thick black line) in Fig. 9a and is referred to as “California(MM)” (CMM) in the following. The “surrounding region” as used to compute ΔXCH_4 (via “source – background” XCH₄) is shown as white rectangle. As shown in Fig. 9, ΔXCH_4 is 9.4 ppb, and the estimated emission of the CMM region, computed using Eq. (1) with the parameters described earlier, is 729 +/- 664 ktCH₄/yr. The GCT15 emissions, i.e., the “true” emissions, are shown in Fig. 9b and the emission is 727 ktCH₄/yr in the CMM source region. It needs to be pointed out that the GCT15 emissions can be large outside the selected CMM source region, in particular in the San Francisco area (the red cell corresponds to an emission of nearly 200 ktCH₄/yr) but this major source region is located outside the selected source region, which is defined based (only) on XCH₄ (Fig. 9a). The excellent agreement of the estimated emission and the true emission can, of course, be simply by chance in this case. Here it is likely that XCH₄ over the CMM region is (due to transport) significantly affected by San Francisco emissions, i.e., by emission located outside the source region (see also Bao et al., 2008, for a discussion of the meteorology in this area). Therefore, one has to be careful

when interpreting the estimated emissions as they may also be influenced by emission sources in the surroundings. On the other hand, there is also outflow from the source region into the surrounding region. All this (and other aspects) result in quite large uncertainty of the estimated emission and this is reflected in the uncertainty estimate, which is quite conservative, i.e., it is quite large. In this case, our estimated (1-sigma) uncertainty is 664 ktCH₄/yr, which is nearly 100% of the estimated emission. This uncertainty has been computed for the surrounding region shown in Fig. 9, i.e., by neglecting the additional error contribution due to variations of the surrounding region ($\sigma_{\Delta XCH_4}$ in Eq. (5)). This contribution is however small compared to error term σ_{CF} (= 664 ktCH₄/yr in this case). That the total uncertainty is typically clearly dominated by σ_{CF} is a finding that has also been confirmed when analyzing the real satellite data (see Sect. 4), where both uncertainty contributions are always considered.

Figure 10 shows similar results to those in Fig. 9 but for an extended source region, denoted CMS in the following. This region covers the region from near San Francisco in the north to Los Angeles in the south. As can be seen, ΔXCH_4 is 7.2 ppb and the estimated emission is 770 +/- 685 ktCH₄/yr, which is significantly lower than the “true” CMS region emission of 1228 ktCH₄/yr, i.e., in this case the estimated emission is wrong by -37% (computed as “(estimated – true)/true”). However, the true emission is inside the uncertainty range of the 1-sigma range of the estimated emission (but close to the upper edge of the uncertainty range, which is 1455 ktCH₄/yr). The reason for this underestimation is very likely due to the fact that the emission sources are distributed very irregularly inside the CMS region. As already explained above, a significant underestimation of the estimated emission is expected in this case.

As can also be seen from Fig. 10a, there is a region of clearly elevated XCH₄ in the southern part of the CMS source region. This region corresponds to the Los Angeles area. Figure 11a shows a zoom into this region. In this case we define the source region by a simple rectangle. The estimated Los Angeles area methane emission is 250 +/- 425 ktCH₄/yr, whereas the true emission is 367 ktCH₄/yr, i.e., the difference -32% (negative, i.e., the estimated emission is (again) underestimated).

Another interesting source region is the Four Corners, which is discussed in detail in Sect. 4. As shown in Fig. 12, the estimated emission is 795 +/- 697 ktCH₄/yr, whereas the “true emission” is 1404 ktCH₄/yr, i.e., the difference -43%.

Comparisons of estimated versus true emissions such as those presented here have also been carried out for several other of the methane emission hot spot area shown in Fig. 8. Figure 13 presents an overview of the corresponding results. As can be seen, the estimated emissions are typically underestimated by about 40%. The emission uncertainties are large (on the order of 100%) but the true emissions are within the 1-sigma uncertainty estimate of the estimated emission (with one exception: Chicago area: here the true emission is 1473 ktCH₄/yr but the upper (1-sigma) range of the estimated emission is 1322 ktCH₄/yr). Based on these results it is concluded that the estimation method as described in this manuscript provides reasonable results but with a clear tendency to underestimate the emissions (as expected from the theoretical considerations

presented earlier). To what extent the 40% value depends on the model used (in this case GEOS-Chem) and on its characteristics (such as spatio-temporal resolution) needs to be investigated (e.g., by using also other models). In any case, the results presented in this section need to be considered when interpreting results obtained from applying this method to real satellite XCH₄ retrievals as presented in the following section.-

5

4 Results and discussion

In this section we present the results from applying the methane emission inversion method described in the previous section to obtain emission estimates from satellite XCH₄ retrievals for four areas: for the Four Corners area in the south-western USA (Sect. 4.1), for ~~in~~ the southern part of the Central Valley in California (Sect. 4.2) and the for two countries Azerbaijan and Turkmenistan (Sect. 4.3). All these areas show elevated methane relative to their surrounding areas (Fig. 1). The spatial locations of these areas as well as key parameters used to convert the observed methane enhancements to annual methane emissions are listed in Tab. 2.

4.1 Four Corners area, USA

Four Corners is a region in the USA named after the quadripoint where the boundaries of the four states Utah, Colorado, Arizona and New Mexico meet. The Four Corners area is one of the largest methane hotspots in the USA (Kort et al., 2014; Wecht et al., 2014b; Frankenberg et al., 2016). The San Juan Basin, located in the Four Corners area, is a geologic structural basin and primarily a natural gas production area, mostly from coal bed methane and shale formations (e.g., Frankenberg et al., 2016, and references given therein). Figure 146 shows annually averaged XCH₄ from the four satellite XCH₄ products as used in this study at and around Four Corners. Here the XCH₄ is shown as anomaly, to be able to better compare the spatial pattern of the shown data products. As can be seen, all satellite products show that XCH₄ is enhanced in the Four Corners area relative to the surrounding area (for the OCPD product this is difficult to see because the obtained enhancement is the smallest of all products). Figure 146 shows the chosen source region as (inner) rectangle. The outer rectangle (see figures in last column and last row) shows the “default” surrounding area. As described above, the methane enhancement ΔXCH_4 is computed as the difference between the XCH₄ mean value in the source region minus the XCH₄ mean value in the surrounding region. For the inversion the size of the surrounding area is varied to determine the sensitivity of the computed ΔXCH_4 with respect to the chosen surroundingbackground region. For this purpose, the latitudes and longitudes of the rectangular box, which defines the surrounding area, are varied by adding all combinations of 0°, 1°, 2°, and 3° in the latitude and longitude directions. The standard deviation of the resulting ΔXCH_4 is used as an estimate of $\sigma_{\Delta XCH_4}$ (see Eq. (5)).

Figure 157 shows the resulting XCH₄ enhancements for all years and all satellite data products including (1-sigma) uncertainty estimates (i.e., $\sigma_{\Delta XCH_4}$) as vertical bars. As can be seen, all ΔXCH_4 values are positive. This shows that a positive Four Corners methane enhancement is present for all years in all satellite products. The methane enhancement is on average about 10 ppb but shows significant variation depending on satellite product and year.

These methane enhancements and their uncertainties are converted to Four Corners area annual methane emissions using the method described in Sect. 3. The results are shown in Fig. 168. The estimated emissions are in the range 0.42 – 0.57 MtCH₄/year (range of annual mean values of the four satellite products). Taking into account the (large) uncertainty of the estimated annual emissions, this is in good agreement with published values as shown in Fig. 168. For example, Kort et al., 2014, report 0.59 MtCH₄/year for the time period 2003-2009 (based on SCIAMACHY and ground-based Fourier-Transform (FT) spectrometer observations) and Turner et al., 2015, report the range of 0.45 -1.39 MtCH₄/year for the time period 2009-2011 (based on an analysis of GOSAT data). The good agreement with the published values indicates that the method used here appears to be capable to deliver reasonable emission estimates even if the source area is much smaller than the 6°x4° regions used for calibrating our inversion method. The agreement is surprisingly good given the large (1-sigma) uncertainty values shown in Fig. 168 (approx. 0.6 MtCH₄/year (~100%) and dominated by σ_{CF} as can be concluded from a comparison with $\sigma_{\Delta XCH_4}$ shown in Fig. 167 (~20%). Our reported uncertainty of the annual averages seems to be too conservative (at least for quantifying the Four Corners area emissions).

Figure 168 also shows the total anthropogenic emissions during 2003-2008 as obtained from the EDGAR v4.2 data base (obtained from http://edgar.jrc.ec.europa.eu/part_CH4.php) for the Four Corners source region. The mean value of the annual EDGAR emissions is 0.17 MtCH₄/year. As can be seen, the EDGAR emissions are too low by approximately a factor of three.

4.2 Central Valley, California, USA

California emits large amounts of methane, approximately 2-3 MtCH₄/year (Turner et al., 2015) and major emission sources are livestock, gas/oil and landfills/wastewater (e.g., Wecht et al., 2014b). According to the EDGAR v4.2 emission data base total anthropogenic methane emissions are largest around Los Angeles and San Francisco dominated by landfill/wastewater and gas/oil related emissions and in the area in between, in the Central Valley, emissions are dominated by livestock emissions (see Wecht et al., 2014b, their Fig. 1).

The Central Valley in California shows up as a methane hotspot in satellite data (see Fig. 179) with largest values in the southern part of the Central Valley around Bakersfield, an important oil and gas producing area (e.g., Jeong et al., 2014; Guha et al., 2015) and an area with significant methane emissions from dairy and livestock (e.g., Wecht et al., 2014b; Guha et al., 2015), extending up to the city of Fresno or even further towards Modesto / San Francisco. This southern part of the Central Valley is the San Joaquin Valley. In this study we define Central Valley as the rectangular region specified by the latitude/longitude range as listed in Tab. 2, corresponding to the region where the satellite XCH₄ is highest. This region roughly corresponds to the San Joaquin Valley. According to EDGAR this region is dominated by livestock methane

emissions with significant contributions from gas/oil and landfill/wastewater related emissions ([see also Maasackers et al., 2016, for a recent US methane emission inventory and comparison with EDGAR v4.2](#)).

Figure [179](#) shows SCIAMACHY WFMD (and IMAP) XCH₄ for year 2004 over California and also shows the Central Valley source region as defined for this study (inner rectangle of Fig. [189](#) top left) and its “default” surrounding area (outer rectangle Fig. [179](#) top right). Figure [179](#) also shows EDGAR v4.2 total anthropogenic methane emissions for the year 2004 regridded to 0.5°x0.5°. As can be seen, the spatial pattern of the EDGAR emissions significantly deviates from the spatial pattern of the satellite XCH₄. Whereas in EDGAR the highest values are around San Francisco and around Los Angeles, the satellite-derived atmospheric methane is highest in the area in between, in the Central Valley, particularly in the area around Bakersfield. Methane emissions in the Bakersfield region are supposed to be dominated by dairy and livestock operations (Guha et al., 2015, and references given therein).

For comparison with the satellite data and the EDGAR emissions also the CAMS emissions are shown (Fig. [179](#) bottom row). On the left (Fig. [179e](#)) the CAMS v10-S1NOAA product is shown, which is based on the assimilation of NOAA methane observations and on the right product v10-S1SCIA (Fig. [179f](#)) based on the additional assimilation of SCIAMACHY IMAP XCH₄. Surprisingly, the assimilation of SCIAMACHY XCH₄ reduces the derived methane emissions in this region. That the Central Valley SCIAMACHY XCH₄ enhancement is not modelled well with optimized emissions obtained from assimilating SCIAMACHY data using the global TM5-4DVAR system is also clearly visible in Bergamaschi et al., 2009 (their Fig. 2), discussing an earlier (pre-CAMS) version of this data set. As already mentioned, the emissions of California are expected to be in the range 2-3 MtCH₄/year (see Turner et al., 2015, their Fig. 6), i.e., larger than the v10-S1NOAA (Fig. [179e](#)) and v10-S1SCIA (Fig. [179f](#)) products suggests. The exact reason why the assimilation of the SCIAMACHY data does not lead to larger estimated emissions in this region is unclear but very likely this is due to the fact that the CAMS inversion system is a global system at quite low spatial resolution and therefore not necessarily optimal for providing reliable emission estimates for regions which are smaller or just on the order of the size of the 6°x4° grid cells shown in Fig. [179](#) bottom.

As can be seen from Fig. [180](#), we obtain mean annual emissions in the range 1.05-1.55 MtCH₄/year, depending on data product. The estimated uncertainty of the annual emissions is ~1 MtCH₄/year (1-sigma) and the inter-annual variations are 20-50% (1-sigma) of the mean emissions, depending on product. Our annual emission estimates are quite uncertain with mean values much higher compared to the emissions as given in the EDGAR v4.2 anthropogenic methane emission inventory. According to EDGAR the total anthropogenic methane emissions in the selected source area are around 0.197 MtCH₄/year, i.e., a factor of ~~56-89~~ lower than our annual mean estimates. This is unlikely due to the fact that our emissions are total emissions whereas EDGAR only reports anthropogenic emissions as the fraction of natural methane emissions in California is estimated to be only approximately 3% percent (Wecht et al., 2014b). Our results are broadly consistent with

recently published results from CalNex campaign (May – June 2010) aircraft observations (Wecht et al., 2014b) also showing high atmospheric methane concentrations over the southern Central Valley compared to the rest of California and concluding that EDGAR emissions in this region need to be scaled with factors up to around five (see their Fig. 2). Wecht et al., 2014a, also derived emissions in this area using SCIAMACHY IMAF retrievals. They report that their derived emissions are consistent with the ones presented in Wecht et al., 2014b, and for the Central Valley they found that the derived emissions are a factor of 2-4 higher compared to EDGAR v4.2 (~~note that~~ their definition of Central Valley is not exactly identical with our definition, which is restricted to the southern part of the Central Valley). They conclude that the livestock emissions in EDGAR are significantly underestimated.

10 Jeong et al., 2013, present an analysis of methane emissions using atmospheric observations from five sites in California's Central Valley across different seasons (September 2010 to June 2011). They obtained spatially resolved (13 sub-regions) top-down estimates of California's CH₄ emissions using in-situ tower data. They report for their region R12, which is similar to but not exactly identical with the area chosen in our assessment, emissions of 0.85 and 0.94 MtCH₄/yr (depending on *a priori* assumptions) based on inversion of in-situ tower data (see their Tab. 5 reporting methane emissions in TgCO₂eq
15 computed assuming a global warming potential of 21 gCO₂eqCH₄/gCH₄), which is a factor of 3.6 (= 17.89 / 5.01, see their Tab. 5) higher than EDGAR v4.2.

Jeong et al., 2014, also studied this region and presented a new spatially resolved bottom-up inventory of methane for 2010 focusing on methane emissions from petroleum production and natural gas systems in California. They showed that the region around Bakersfield is a major oil and gas production and transmission region in California (see their Fig. 1) and they found that their emission estimates are 3-7 times higher for the petroleum and gas production sectors compared to official California bottom-up inventories.

Our results corroborate the findings of these independent studies that inventory emissions are underestimated in this region.
25 However, we acknowledge the large uncertainty of our estimated annual emissions and cannot rule out that our emission estimates are overestimated, e.g., due to possible methane accumulation in the southern part of the Central Valley.

4.3 Azerbaijan and Turkmenistan

Azerbaijan and Turkmenistan are located next to the Caspian Sea (to the west and to the east, respectively) and both countries are important oil and gas producers. Azerbaijan and Turkmenistan are clearly visible as methane emission hotspots in satellite XCH₄ data sets (Fig. 1, Fig. 1944).

Figure 1944 shows SCIAMACHY WFMD year 2004 XCH₄ in the Azerbaijan / Turkmenistan area and emission data base results from EDGAR v4.2 (Fig. 1944, bottom left), CAMS v10-S1NOAA (Fig. 1944, bottom middle) and CAMS v10-S1SCIA (Fig. 1944, bottom right). In contrast to the results discussed in the previous section, the assimilation of SCIAMACHY data in the TM5-4DVAR assimilation system enhances the emissions around Azerbaijan / Turkmenistan (compare Fig. 1944 bottom middle with bottom right).

Figure 2042 shows Azerbaijan methane emissions as obtained with our inversion method compared to EDGAR v4.2 emissions. As can be seen, the satellite-derived emissions are consistent with EDGAR. ~~Here~~Note that the CH₄_GOS_SRFP product is not shown. Due to the sparse spatial sampling of this product the inter-annual variability is dominated by year-to-year sampling differences. Azerbaijan is surrounded by many other methane emission areas and, therefore, not a well-isolated emission hotspot, i.e., not ideal for our inversion method. The impact of this is largest for the CH₄_GOS_SRFP product, which is a sparse data set as the underlying “full physics” retrieval algorithm requires strict quality filtering.

Turkmenistan is much larger in size compared to Azerbaijan (see Fig. 1944) but also not a well-isolated emission hotspot. The results for Turkmenistan are shown in Fig. 2143. Here the mean values of all estimated emissions are positive (in contrast to Azerbaijan) indicating that the methane concentration over Turkmenistan is higher than its surrounding for all years and all four satellite products. The mean values of the derived emissions are in the range 1.85 – 2.08 MtCH₄/year, which is about 50% larger compared to EDGAR (1.33 MtCH₄/year). This may be due to an underestimation of Turkmenistan’s oil and gas related methane emissions in EDGAR but one also has to note the large uncertainty of our satellite-derived annual emissions. Furthermore, Turkmenistan is not an ideally-isolated methane hotspot, although the Azerbaijan results do not indicate that this is necessarily a significant issue. Note also that mountains are located southward and eastward of Turkmenistan and this may contribute to a local accumulation (trapping) of atmospheric methane (resulting in an overestimation of our estimated emissions) and may explain why the elevated methane over Turkmenistan as shown in Fig. 1944 is well correlated with the country boundaries. Clearly, more studies are needed to clarify this but this likely requires much more complex inversion methods than the one used in this study (e.g., similar to those presented in Wecht et al., 2014a, and Gentner et al., 2014).

5 Summary and conclusions

We have presented a simple but very fast method to estimate methane surface emissions of areas showing elevated atmospheric methane concentrations relative to their surrounding areas (“methane hotspots”) in satellite-derived XCH₄ maps, especially in those derived from SCIAMACHY/ENVISAT. The described “inversion method”, which is a simple mass balance method, is applicable to time-averaged XCH₄ data sets (as complex spatio-temporal XCH₄ variations due to varying meteorological conditions cannot be considered by our method). Hereand we focus on annual XCH₄ maps to derive annual emissions. The method is based on a direct conversion of a localized methane enhancement (relative to its surrounding area) using a conversion factor, which mainly depends on the size of the source region of interest. The method is calibrated using (global~~low-resolution~~) 2-dimensional methane emission maps and corresponding global 2-dimensional XCH₄ maps generated from Copernicus Atmospheric Monitoring Service (CAMS) 3-dimensional atmospheric methane fields. A limitation of our method is its quite large uncertainty. We estimate that the uncertainty of the method is about 80% for annual emissions around 1 MtCH₄/year but having better relative uncertainty for larger emissions (down to about 50% for very large emissions, i.e., several MtCH₄/year).

The inversion method has been tested by applying it to a high-resolution methane data set covering the USA, which has been computed with GEOS-Chem. We retrieve methane emissions for several areas where the GEOS-Chem data set shows elevated XCH₄ compared to their surrounding areas. We found that the estimated emissions are typically 40% lower compared to the emissions used in the model (which are the known, i.e., “true” emissions of this simulation experiment). The true emissions are (with one exception) located within the 1-sigma uncertainty range of our emission estimates. From theoretical considerations we expect that our method tends to underestimate emissions, i.e., that it provides rather conservative emission estimates. To what extent the 40% value depends on the model used and on its characteristics (such as spatio-temporal resolution) needs to be investigated in the future by using additional models.

We applied ourthis method to an ensemble of satellite XCH₄ data products using two products from SCIAMACHY/ENVISAT and two products from TANSO-FTS/GOSAT as made available via the GHG-CCI project website (<http://www.esa-ghg-cci.org/>) of ESA’s Climate Change Initiative (CCI). These products cover the time period 2003-2014.

The inversion method as applied to real satellite data has been applied to four source areas. Two of the source areas are located in the USA (the Four Corners area located in the southwestern USA and the southern part of the Central Valley, i.e., the region around Bakersfield and Fresno, in California) and the two other source regions are Azerbaijan and Turkmenistan,

which are both important oil and gas producing countries. All four regions clearly show elevated methane relative to their surrounding in satellite-derived XCH₄ maps.

For Four Corners we obtain annual emissions in the range 0.42 – 0.57 MtCH₄/year in agreement with published values. For Azerbaijan our estimates are on average close to the total anthropogenic methane emissions of Azerbaijan as given in the EDGARv4.2 (FT2012) emission inventory but for Turkmenistan we obtain about 50% higher emissions on average albeit with large uncertainty. Further study is needed to investigate if this is due to an underestimation of Turkmenistan's oil and gas related emissions in EDGAR.

10 For the region around Bakersfield located in the Central Valley of California, a region of significant oil and gas production and large expected methane emissions from dairy and livestock operations, we obtain mean emissions in the range 1.05-1.55 MtCH₄/year, depending on satellite data product. This is about a factor of ~~56-89~~ higher than the total methane emissions as given in the EDGAR v4.2 inventory, but of similar magnitude as reported in Jeong et al., 2013, (0.85 – 0.94 MtCH₄/year) based on inverse modelling of tower measurements. Our findings also corroborate published results from CalNex campaign
15 aircraft observations during May to June 2010 (Wecht et al., 2014b) showing high methane concentrations over the southern part of the Central Valley, in the San Joaquin Valley, compared to other parts of California and concluding that EDGAR emissions in this area need to be scaled with factors up to around five. They conclude that livestock emissions in EDGAR are significantly underestimated. Another more recent study (Joeng et al., 2014) presented a new bottom-up methane inventory for the year 2010 for California concluding that their emissions are 3-7 times higher compared to official
20 California bottom-up inventories for the petroleum and natural gas production sectors. Also the new US Environmental Protection Agency (EPA) methane emission inventory (Maasakkers et al., 2016) shows significantly larger emission in the area around Bakersfield compared to EDGAR v4.2. Nevertheless, our results need to be interpreted with care as the uncertainty of our annual emission estimates is ~~quite~~ large and we cannot entirely rule out that our estimates are somewhat overestimated, e.g., due to possible methane accumulation in the valley.

25

We recommend further studies to investigate in more detail the reported discrepancy of the satellite-derived emissions with emission inventories in particular for ~~the-Turkmenistan but possibly also for the~~ southern part of the Central Valley in California ~~and for Turkmenistan~~. We also recommend to use ensembles of satellite products as done in this study in order to determine to what extent key findings ~~dependare~~ depending on the algorithmic choices which have to be made when
30 developing a retrieval algorithm used to generate a particular XCH₄ data product and to what extent the findings depend on the particular satellite instrument used to derive the results. More detailed assessments likely require the use of much more complex approaches compared to the simple method uses in this study. Nevertheless, simple and fast approaches also have a role to play as they permit to perform quick assessments on possible discrepancies with respect to emission inventories or other data sets and can also be used for plausibility checks for more complex approaches.

It is also important to monitor the emissions of major methane source regions in the future. In this context the upcoming satellite mission Sentinel-5-Precursor (S5P) will potentially play an important role. S5P is planned to be launched ~~midend~~ of 2017~~6~~ and will deliver XCH₄ at high spatial resolution (7 km at nadir) and with good spatial coverage (2600 km swath width, i.e., daily coverage) (Veefkind et al., 2012; Butz et al., 2012) resulting in methane observations with dense spatio-temporal coverage, which is a significant advantage for methane hotspot detection and related emission quantification compared to the past and present satellites used in this study.

The longer term objective of releasing an observing system comprising instruments having the performance of CarbonSat within a CarbonSat constellation (Bovensmann et al., 2010; Velazco et al., 2011; Buchwitz et al., 2013; Pillai et al., 2016; ESA, 2015) is currently being discussed by the European Space Agency (ESA) and European Union (EU) representatives within the Copernicus program focusing on CO₂ (e.g., Ciais et al., 2015). Such a system will provide, when coupled with sparse but accurate ground-based systems, the objective evidence about the global CH₄ and CO₂ surface fluxes needed for verification and monitoring of emissions and to improve our knowledge on natural carbon fluxes.

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Annex A: Illustration of emission estimation method

Figure A1 illustrates the basic idea of the methane emission estimation method explained in Sect. 3. In particular it is illustrated how the observed methane enhancement over the source region (region A in Fig. A1 (a)), ΔXCH_4 , is related to the source region emission (E , in mass per time), wind speed magnitude V , and length of the source region. The source region shown here is a rectangle of area $A = L_x L_y$, where wind speed is in the x direction. Note that length L as given in Eq. (2) corresponds to length L_x of Fig. A1.

The computation of the methane mole fraction enhancement over the source region relative to its surrounding, ΔXCH_4 , is computed (see Sect. 3 and Sect. 4) by subtracting the mean value of XCH_4 in the surrounding region (region B in Fig. A1 (a)) by the mean value of XCH_4 over the source region (region A in Fig. A1 (a)) assuming that the surrounding region does not contain any (significant) emission sources and neglecting atmospheric methane enhancements in the surrounding due to outflow from the source region into the surrounding region (region C in Fig. A1 (a)); note that this requires that region B is much larger than region C). As a consequence, the computed mean value of XCH_4 in the surrounding is typically overestimated and, therefore ΔXCH_4 and the computed methane emission is too low, i.e., the estimated emission is (typically) a conservative estimate.

Note that the method described in Sect. 3 and used in Sect. 4 is only applied to time averages of atmospheric CH_4 to obtain time averaged emissions. This typically means that meteorological situations vary significantly during the selected time period (including large wind speed and wind direction variations) so that detailed structures of the atmospheric methane emission “plumes” originating from local emission sources largely average out resulting in enhanced atmospheric methane over the source region. Note that Fig. A1 (b) only illustrates a “snapshot” in time but not the average over a range of wind speeds and wind directions (assumed to be reasonably well approximated by Fig. A1 (a)).

References

- 25 Alexe, M., Bergamaschi, P., Segers, A., Detmers, R., Butz, A., Hasekamp, O., Guerlet, S., Parker, R., Boesch, H., Frankenberg, C., Scheepmaker, R. A., Dlugokencky, E., Sweeney, C., Wofsy, S. C., and Kort, E. A.: Inverse modeling of CH_4 emissions for 2010–2011 using different satellite retrieval products from GOSAT and SCIAMACHY, *Atmos. Chem. Phys.*, 15, 113–133, www.atmos-chem-phys.net/15/113/2015/, doi:10.5194/acp-15-113-2015, 2015.
- 30 [Bao, J.-W., Michelson, S. A., Persson, P. O. G., Djalalova, I. V., Wilczak, J. M.: Observed and WRF-Simulated Low-Level Winds in a High-Ozone Episode during the Central California Ozone Study, *Journal of applied meteorology and climatology*, Vol. 47, 2372-2394, doi:10.1175/2008JAMC1822.1, 2008.](#)

- Bergamaschi, P., Frankenberg, C., Meirink, J. F., Krol, M., Dentener, F., Wagner, T., Platt, U., Kaplan, J.O., Körner, S., Heimann, M., Dlugokencky, E., and Goede, A. P. H.: Satellite cartography of atmospheric methane from SCIAMACHY onboard ENVISAT: 2. Evaluation based on inverse model simulations, *J. Geophys. Res.*, 112, D02304, doi:10.1029/2006JD007268, 2007.
- 5 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.: Inverse modeling of global and regional CH₄ emissions using SCIAMACHY satellite retrievals, *J. Geophys. Res.*, 114, D22301, doi:10.1029/2009JD01228, 2009.
- Bergamaschi, P., Houweling, H., Segers, A., Krol, M., Frankenberg, C., Scheepmaker, R. A., Dlugokencky, E., Wofsy, S. C., Kort, E. A., Sweeney, C., Schuck, T., Brenninkmeijer, C., Chen, H., Beck, V., and Gerbig, C.: Atmospheric CH₄ in the first decade of the 21st century: Inverse modeling analysis using SCIAMACHY satellite retrievals and NOAA surface measurements, *J. Geophys. Res.*, 118, 7350-7369, doi:10.1002/jrgd.5048, 2013.
- 10 Bloom, A. A., Palmer, P. I., Fraser, A., Reay, D. S., and Frankenberg, C.: Large-scale controls of methanogenesis inferred from methane and gravity spaceborne data, *Science*, 327, 322–325, doi:10.1126/science.1175176, 2010.
- Bovensmann, H., Burrows, J. P., Buchwitz, M., Frerick, J., Noël, S., Rozanov, V. V., Chance, K. V., and Goede, A. H. P.: SCIAMACHY - Mission objectives and measurement modes, *J. Atmos. Sci.*, 56 (2), 127-150, 1999.
- 15 Bovensmann, H., Buchwitz, M., Burrows, J. P., Reuter, M., Krings, T., Gerilowski, K., Schneising, O., Heymann, J., Tretner, A., and Erzinger, J.: A remote sensing technique for global monitoring of power plant CO₂ emissions from space and related applications, *Atmos. Meas. Tech.*, 3, 781-811, 2010.
- Buchwitz, M., Rozanov, V. V., and Burrows, J. P.: A near-infrared optimized DOAS method for the fast global retrieval of atmospheric CH₄, CO, CO₂, H₂O, and N₂O total column amounts from SCIAMACHY Envisat-1 nadir radiances, *J. Geophys. Res.* 105, 15,231-15,245, 2000.
- 20 Buchwitz, M., de Beek, R., Burrows, J. P., Bovensmann, H., Warneke, T., Notholt, J., Meirink, J. F., Goede, A. P. H., Bergamaschi, P., Körner, S., Heimann, M., and Schulz, A.: Atmospheric methane and carbon dioxide from SCIAMACHY satellite data: Initial comparison with chemistry and transport models, *Atmos. Chem. Phys.*, 5, 941-962, 2005.
- 25 Buchwitz, M., Reuter, M., Bovensmann, H., Pillai, D., Heymann, J., Schneising, O., Rozanov, V., Krings, T., Burrows, J. P., Boesch, H., Gerbig, C., Meijer, Y., and Loesch, A.: Carbon Monitoring Satellite (CarbonSat): assessment of atmospheric CO₂ and CH₄ retrieval errors by error parameterization, *Atmos. Meas. Tech.*, 6, 3477-3500, 2013.
- Buchwitz, M., Reuter, M., Schneising, O., Boesch, H., Guerlet, S., Dils, B., Aben, I., Armante, R., Bergamaschi, P., Blumenstock, T., Bovensmann, H., Brunner, D., Buchmann, B., Burrows, J. P., Butz, A., Chédin, A., Chevallier, F., Crevoisier, C. D., Deutscher, N. M., Frankenberg, C., Hase, F., Hasekamp, O. P., Heymann, J., Kaminski, T.,
- 30

- Laeng, A., Lichtenberg, G., De Mazière, M., Noël, S., Notholt, J., Orphal, J., Popp, C., Parker, R., Scholze, M., Sussmann, R., Stiller, G. P., Warneke, T., Zehner, C., Bril, A., Crisp, D., Griffith, D. W. T., Kuze, A., O'Dell, C., Oshchepkov, S., Sherlock, V., Suto, H., Wennberg, P., Wunch, D., Yokota, T., and Yoshida, Y.: The Greenhouse Gas Climate Change Initiative (GHG-CCI): comparison and quality assessment of near-surface-sensitive satellite-derived CO₂ and CH₄ global data sets, *Remote Sensing of Environment*, 162, 344-362, doi:10.1016/j.rse.2013.04.024, 2015.
- Buchwitz, M., Dils, B., Boesch, H., Crevoisier, C., Detmers, D., Frankenberg, C., Hasekamp, O., Hewson, W., Laeng, A., Noël, S., Notholt, J., Parker, R., Reuter, M., and Schneising, O.: ESA Climate Change Initiative (CCI) Product Validation and Intercomparison Report (PVIR) for the Essential Climate Variable (ECV) Greenhouse Gases (GHG) for data set Climate Research Data Package No. 3 (CRDP#3), Version 4.0, 24. Feb. 2016 (link: http://www.esa-ghg-cci.org/?q=webfm_send/300), 2016a.
- Buchwitz, M., Reuter, M., Schneising, O., Hewson, W., Detmers, R. G., Boesch, H., Hasekamp, O. P., Aben, I., Bovensmann, H., Burrows, J. P., Butz, A., Chevallier, F., Dils, B., Frankenberg, C., Heymann, J., Lichtenberg, G., De Mazière, M., Notholt, J., Parker, R., Warneke, T., Zehner, C., Griffith, D. W. T., Deutscher, N. M., Kuze, A., Suto, H., and Wunch, D.: Global satellite observations of column-averaged carbon dioxide and methane: The GHG-CCI XCO₂ and XCH₄ CRDP3 data, ~~submitted to~~ *Remote Sensing of Environment* (in press), Special Issue on Essential Climate Variables, DOI: [10.1016/j.rse.2016.12.027](https://doi.org/10.1016/j.rse.2016.12.027), (link: <http://dx.doi.org/10.1016/j.rse.2016.12.027>), 2016b.
- Burrows, J. P., Hölzle, E., Goede, A. P. H., Visser, H., and Fricke, W.: SCIAMACHY—Scanning Imaging Absorption Spectrometer for Atmospheric Chartography, *Acta Astronaut.*, 35(7), 445–451, doi:10.1016/0094-5765(94)00278-t, 1995.
- Butz, A., Hasekamp, O.P., Frankenberg, C., Vidot, J., and Aben, I.: CH₄ retrievals from space-based solar backscatter measurements: Performance evaluation against simulated aerosol and cirrus loaded scenes, *J. Geophys. Res.*, VOL. 115, D24302, doi:10.1029/2010JD014514. 2010.
- Butz, A., Guerlet, S., Hasekamp, O., Schepers, D., Galli, A., Aben, I., Frankenberg, C., Hartmann, J.-M., Tran, H., Kuze, A., Keppel-Aleks, G., Toon, G., Wunch, D., Wennberg, P., Deutscher, N., Griffith, D., Macatangay, R., Messerschmidt, J., Notholt, J., and Warneke, T.: Towards accurate CO₂ and CH₄ observations from GOSAT, *Geophys. Res. Lett.*, *Geophys. Res. Lett.*, doi:10.1029/2011GL047888, 2011.
- Butz, A., Galli, A., Hasekamp, O., Landgraf, J., Tol, P., and Aben, I.: Remote Sensing of Environment, TROPOMI aboard Sentinel-5 Precursor : Prospective performance of CH₄ retrievals for aerosol and cirrus loaded atmospheres, 120, 267-276, doi:10.1016/j.rse.2011.05.030, 2012.

- Chevallier, F., Buchwitz, M., Bergamaschi, P., Houweling, S., van Leeuwen, T., and Palmer, P. I.: User Requirements Document for the GHG-CCI project of ESA's Climate Change Initiative, version 2 (URDv2), 28. August 2014, (link: http://www.esa-ghg-cci.org/?q=webfm_send/173), 2014b.
- 5 Chevallier, F., Alexe, M., Bergamaschi, P., Brunner, D., Feng, L., Houweling, S., Kaminski, T., Knorr, W., van Leeuwen, T., Marshall, J., Palmer, P. I., Scholze, M., Sundström, A.-M., and Voßbeck, M.: ESA Climate Change Initiative (CCI) Climate Assessment Report (CAR) for Climate Research Data Package No. 3 (CRDP#3) of the Essential Climate Variable (ECV) Greenhouse Gases (GHG), Version 3, pp. 94, 3 May 2016 (link: http://www.esa-ghg-cci.org/?q=webfm_send/318), 2016.
- 10 Ciaï, P., Dolman, A. J., Bombelli, A., et al.: Current systematic carbon cycle observations and needs for implementing a policy-relevant carbon observing system, *Biogeosciences*, 11, 3547-3602, www.biogeosciences.net/11/3547/2014/, doi:10.5194/bg-11-3547-2014, 2014.
- Ciaï, P., et al.: Towards a European Operational Observing System to Monitor Fossil CO₂ emissions - Final Report from the expert group, <http://www.copernicus.eu/main/towards-european-operational-observing-system-monitor-fossil-co2-emissions>, pp. 68, October 2015, 2015.
- 15 Cressot, C., Chevallier, F., Bousquet, P., Crevoisier, C., Dlugokencky, E. J., Fortems-Cheiney, A., Frankenberg, C., Parker, P., Pison, I., Scheepmaker, R. A., Montzka, S. A., Krummel, P. B., Steele, L. P., and Langenfelds, R. L.: On the consistency between global and regional methane emissions inferred from SCIAMACHY, TANSO-FTS, IASI and surface measurements, *Atmos. Chem. Phys.*, 14, 577-592, 2014.
- 20 Dee, D. P., Uppala, S. M., Simmons, A. J., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M. A., Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A. C. M., van de Berg, L., Bidlot, J., Bormann, N., Delsol, C., Dragani, R., Fuentes, M., Geer, A. J., Haimberger, L., Healy, S. B., Hersbach, H., Hólm, E. V., Isaksen, L., Kållberg, P., Köhler, M., Matricardi, M., McNally, A. P., Monge-Sanz, B. M., Morcrette, J.-J., Park, B.-K., Peubey, C., de Rosnay, P., Tavolato, C., Thépaut, J.-N. and Vitart, F.: The ERA-Interim reanalysis: configuration and performance of the data assimilation system. *Q.J.R. Meteorol. Soc.*, 137: 553–597. doi:10.1002/qj.828, 2011.
- 25 Dils, B., Buchwitz, M., Reuter, M., Schneising, O., Boesch, H., Parker, R., Guerlet, S., Aben, I., Blumenstock, T., Burrows, J. P., Butz, A., Deutscher, N. M., Frankenberg, C., Hase, F., Hasekamp, O. P., Heymann, J., De Maziere, M., Notholt, J., Sussmann, R., Warneke, T., Griffith, D., Sherlock, V., and Wunch, D.: The Greenhouse Gas Climate Change Initiative (GHG-CCI): comparative validation of GHG-CCI SCIAMACHY/ENVISAT and TANSO-FTS/GOSAT CO₂ and CH₄ retrieval algorithm products with measurements from the TCCON, *Atmos. Meas. Tech.*, 7, 1723-1744, 2014.
- 30

- ESA: Report for Mission Selection: CarbonSat, ESA SP-1330/1 (2 volume series), Noordwijk, The Netherlands. Available at: http://esamultimedia.esa.int/docs/EarthObservation/SP1330-1_CarbonSat.pdf, 2015.
- Frankenberg, C., Meirink, J. F., van Weele, M., Platt, U., and Wagner, T.: Assessing methane emissions from global spaceborne observations, *Science*, 308, 1010–1014, doi:10.1126/science.1106644, 2005.
- 5 Frankenberg, C., Meirink, J. F., Bergamaschi, P., Goede, A. P. H., Heimann, M., Körner, S., Platt, U., van Weele, M., and Wagner, T.: Satellite cartography of atmospheric methane from SCIAMACHY onboard ENVISAT: Analysis of the years 2003 and 2004, *J. Geophys. Res.*, 111, D07303, doi:10.1029/2005JD006235, 2006.
- Frankenberg, C., Warneke, T., Butz, A., Aben, I., Hase, F., Spietz, P., and Brown, L. R.: Pressure broadening in the $2\nu_3$ band of methane and its implication on atmospheric retrievals, *Atmos. Chem. Phys.*, 8, 5061 – 5075, 2008a.
- 10 Frankenberg, C., Bergamaschi, P., Butz, A., Houweling, S., Meirink, J. F., Notholt, J., Petersen, A. K., Schrijver, H., Warneke, T., Aben, I.: Tropical methane emissions: A revised view from SCIAMACHY onboard ENVISAT, *Geophys. Res. Lett.*, 35, L15811, doi:10.1029/2008GL034300, 2008b.
- Frankenberg, C., Aben, I., Bergamaschi, P., Dlugokencky, E. J., van Hees, R., Houweling, S., van der Meer, P., Snel, R., and Tol, P.: Global column-averaged methane mixing ratios from 2003 to 2009 as derived from SCIAMACHY: Trends and variability, *J. Geophys. Res.*, doi:10.1029/2010JD014849, 2011.
- 15 Frankenberg, C., Thorpe, A. K., Thompson, D. R., Hulley, G., Kort, E. A., Vanceb, N., Borchardt, J., Krings, T., Gerilowski, K., Sweeney, C., Conley, S., Bueb, B. D., Aubrey, A. D., Hook, S., and Green, R. O.: Airborne methane remote measurements reveal heavytail flux distribution in Four Corners region, *Proc. Natl. Acad. Sci USA (PNAS)*, <http://www.pnas.org/content/early/2016/08/10/1605617113>, pp. 6, 2016.
- 20 Fraser, A., Palmer, P. I., Feng, L., Boesch, H., Cogan, A., Parker, R., Dlugokencky, E. J., Fraser, P. J., Krummel, P. B., Langenfelds, R. L., O’Doherty, S., Prinn, R. G., Steele, L. P., van der Schoot, M., and Weiss, R. F.: Estimating regional methane surface fluxes: the relative importance of surface and GOSAT mole fraction measurements, *Atmos. Chem. Phys.*, 13, 5697-5713, doi:10.5194/acp-13-5697-2013. 2013.
- Gentner, D. R., Ford, T. B., Guha, A., Boulanger, K., Brioude, J., Angevine, W. M., de Gouw, J. A., Warneke, C., Gilman, J. B., Ryerson, T. B., Peischl, J., Meinardi, S., Blake, D. R., Atlas, E., Lonneman, W. A., Kleindienst, T. E., Beaver, M. R., St. Clair, J. M., Wennberg, P. O., VandenBoer, T. C., Markovic, M. Z., Murphy, J. G., Harley, R. A., and Goldstein, A. H.: Emissions of organic carbon and methane from petroleum and dairy operations in California’s San Joaquin Valley, *Atmos. Chem. Phys.*, 14, 4955–4978, www.atmos-chem-phys.net/14/4955/2014/, doi:10.5194/acp-14-4955-2014, 2014.
- 25

- Guha, A., Gentner, D. R., Weber, R. J., Provencal, R., and Goldstein, A. H.: Source apportionment of methane and nitrous oxide in California's San Joaquin Valley at CalNex 2010 via positive matrix factorization, *Atmos. Chem. Phys.*, 15, 12043–12063, www.atmos-chem-phys.net/15/12043/2015/, doi:10.5194/acp-15-12043-2015, 2015.
- Hollmann, R., Merchant, C. J., Saunders, R., Downy, C., Buchwitz, M., Cazenave, A., Chuvieco, E., Defourny, P., de Leeuw, G., Forsberg, R., Holzer-Popp, T., Paul, F., Sandven, S., Sathyendranath, S., van Roozendaal, M., and Wagner, W.: The ESA Climate Change Initiative: satellite data records for essential climate variables, *Bulletin of the American Meteorological Society (BAMS)*, 0.1175/BAMS-D-11-00254.1, 2013.
- Houweling, S., Krol, M., Bergamaschi, P., Frankenberg, C., Dlugokencky, E. J., Morino, I., Notholt, J., Sherlock, V., Wunch, D., Beck, V., Gerbig, C., Chen, H., Kort, E. A., Röckmann, T., and Aben, I.: A multi-year methane inversion using SCIAMACHY, accounting for systematic errors using TCCON measurements, *Atmos. Chem. Phys.*, 14, 3991-4012, <http://www.atmos-chem-phys.net/14/3991/2014/>, doi:10.5194/acp-14-3991-2014, 2014.
- [Houweling, S., Bergamaschi, P., Chevallier, F., Heimann, M., Kaminski, T., Krol, M., Michalak, A. M., Patra, P.: Global inverse modeling of CH₄ sources and sinks: an overview of methods, *Atmos. Chem. Phys.*, 17, 235-256, doi:10.5194/acp-17-235-2017, 2017.](#)
- 15 IPCC (2013), *Climate Change 2013: The Physical Science Basis, Working Group I Contribution to the Fifth Assessment Report of the Intergovernmental Report on Climate Change*, <http://www.ipcc.ch/report/ar5/wg1/>, 2013.
- Jacob, D. J., Turner, A. J., Maassackers, J. D., Sheng, J., Sun, K., Liu, X., Chance, K., Aben, I., McKeever, J., and Frankenberg, C.: Satellite observations of atmospheric methane and their value for quantifying methane emissions, *Atmos. Chem. Phys. Discuss.*, [16, 14371-14396](#), doi:10.5194/acp-2016-~~14371-2016~~5551-41, 2016.
- 20 Jeong, S., Hsu, Y.-K., Andrews, A. E., Bianco, L., Vaca, P., Wilczak, J. M., and Fischer, M. L.: A multitower measurement network estimate of California's methane emissions, *J. Geophys. Res. Atmos.*, 118, 11,339–11,351, doi:10.1002/jgrd.50854, 2013.
- Jeong, S., Millstein, D., and Fischer, M. L.: Spatially Explicit Methane Emissions from Petroleum Production and the Natural Gas System in California, [dx.doi.org/10.1021/es4046692](https://doi.org/10.1021/es4046692), *Environ. Sci. Technol.*, 48, 5982-5990, 2014.
- 25 Kirschke, S., Bousquet, P., Ciais, P., et al.: Three decades of global methane sources and sinks, *Nat. Geosci.*, 6, 813–823, doi:10.1038/ngeo1955, 2013.
- Kort, E. A., Frankenberg, C., Costigan, K. R., Lindenmaier, R., Dubey, M. K., and Wunch, D.: Four corners: The largest US methane anomaly viewed from space, *Geophys. Res. Lett.*, 41, doi:10.1002/2014GL061503, 2014.

- Kuze, A., Suto, H., Nakajima, M., and Hamazaki, T.: Thermal and near infrared sensor for carbon observation Fourier-transform spectrometer on the Greenhouse Gases Observing Satellite for greenhouse gases monitoring, *Appl. Opt.*, 48, 6716–6733, 2009.
- 5 Kuze, A., Suto, H., Shiomi, K., Kawakami, S., Tanaka, M., Ueda, Y., Deguchi, A., Yoshida, J., Yamamoto, Y., Kataoka, F., Taylor, T. E., and Buijs, H. L.: Update on GOSAT TANSO-FTS performance, operations, and data products after more than 6 years in space, *Atmos. Meas. Tech.*, 9, 2445–2461, doi:10.5194/amt-9-2445-2016, 2016.
- [Maasackers, J. D., Jacob, D. J., Sulprizio, M. P., Turner, A. J., Weitz, M., Wirth, T., Hight, C., DeFigueiredo, M., Desai, M., Schmeltz, R., Hockstad, L., Bloom, A. A., Bowman, K. W., Jeong, S., Fischer, M. L.: Gridded National Inventory of U.S. Methane Emissions, *Environ. Sci. Technol.*, 50, 13123–13133, doi:10.1021/acs.est.6b02878, 2016.](#)
- 10 Miller, S. M., and Michalak, A. M.: Constraining sector-specific CO₂ and CH₄ emissions in the United States, *Atmos. Chem. Phys. Discuss.*, doi:10.5194/acp-2016-643, 2016.
- Monteil, G., Houweling, S., Butz, A., Guerlet, S., Schepers, D., Hasekamp, O., Frankenberg, C., Scheepmaker, R., Aben, I., and Röckmann, T.: Comparison of CH₄ inversions based on 15 months of GOSAT and SCIAMACHY observations, *J. Geophys. Res.*, doi: 10.1002/2013JD019760, Vol 118, Issue 20, 11807–11823, 2013.
- 15 Nisbet, E., Dlugokencky, E., and Bousquet, P.: Methane on the rise – again, *Science*, 343, 493–495, doi:10.1126/science.1247828, 2014.
- Parker, R., Boesch, H., Cogan, A., Fraser, A., Feng, L., Palmer, P., Messerschmidt, J., Deutscher, N., Griffith, D., Notholt, J., Wennberg, and P. Wunch, D.: Methane Observations from the Greenhouse gases Observing SATellite: Comparison to ground-based TCCON data and Model Calculations, *Geophys. Res. Lett.*, doi:10.1029/2011GL047871, 2011.
- 20 Parker, R. J., Boesch, H., Byckling, K., Webb, A. J., Palmer, P. I., Feng, L., Bergamaschi, P., Chevallier, F., Notholt, J., Deutscher, N., Warneke, T., Hase, F., Sussmann, R., Kawakami, S., Kivi, R., Griffith, D. W. T., and Velasco, V.: Assessing 5 years of GOSAT Proxy XCH₄ data and associated uncertainties, *Atmos. Meas. Tech.*, 8, 4785–4801, doi:10.5194/amt-8-4785-2015, 2015.
- Pillai, D., Buchwitz, M., Gerbig, C., Koch, T., Reuter, M., Bovensmann, H., Marshall, J., and Burrows, J. P.: Tracking city CO₂ emissions from space using a high resolution inverse modeling approach: A case study for Berlin, Germany, *Atmos. Chem. Phys.*, 16, 9591–9610, doi:10.5194/acp-16-9591-2016, 2016.
- 25 Rigby, M., Prinn, R. G., Fraser, P. J., Simmonds, P. G., Langenfelds, R. L., Huang, J., Cunnold, D. M., Steele, L. P., Krummel, P. B., Weiss, R. F., O’Doherty, S., Salameh, P. K., Wang, H. J., Harth, C. M., Mühle, J. and Porter, L. W.: Renewed growth of atmospheric methane, *Geophys. Res. Lett.*, 35, L22805, doi:10.1029/2008GL036037, 2008.
- 30 Rodgers, C. D., *Inverse Methods for Atmospheric Sounding: Theory and Practice*, World Scientific Publishing, 2000.
- Schaefer, H., Mikaloff Fletcher, S. E., Veidt, C., Lassey, K. R., Brailsford, G. W., Bromley, T. M., Dlugokencky, E. J., Michel, S. E., Miller, J. B., Levin, I., Lowe, D. C., Martin, R. J., Vaughn, B. H., and White, J. W. C.: A 21st-century

shift from fossil-fuel to biogenic methane emissions indicated by $^{13}\text{CH}_4$, *Science*, Vol. 352, Issue 6281, pp. 80-84, doi 10.1126/science.aad2705, 2016.

- 5 Schepers, D., Guerlet, S., Butz, A., Landgraf, J., Frankenberg, C., Hasekamp, O., Blavier, J.-F., Deutscher, N. M., Griffith, D. W. T., Hase, F., Kyro, E., Morino, I., Sherlock, V., Sussmann, R., and Aben, I.: Methane retrievals from Greenhouse Gases Observing Satellite (GOSAT) shortwave infrared measurements: Performance comparison of proxy and physics retrieval algorithms, *J. Geophys. Res.*, 117, D10307, doi:10.1029/2012JD017549, 2012.
- Schneising, O., Buchwitz, M., Reuter, M., Heymann, J., Bovensmann, H., and Burrows, J. P.: Long-term analysis of carbon dioxide and methane column-averaged mole fractions retrieved from SCIAMACHY, *Atmos. Chem. Phys.*, 11, 2881-2892, 2011.
- 10 Schneising, O., Bergamaschi, P., Bovensmann, H., Buchwitz, M., Burrows, J. P., Deutscher, N. M., Griffith, D. W. T., Heymann, J., Macatangay, R., Messerschmidt, J., Notholt, J., Rettinger, M., Reuter, M., Sussmann, R., Velazco, V. A., Warneke, T., Wennberg, P. O., and Wunch, D.: Atmospheric greenhouse gases retrieved from SCIAMACHY: comparison to ground-based FTS measurements and model results, *Atmos. Chem. Phys.*, 12, 1527-1540, 2012.
- Schneising, O., Heymann, J., Buchwitz, M., Reuter, M., Bovensmann, H., and Burrows, J. P.: Anthropogenic carbon dioxide source areas observed from space: assessment of regional enhancements and trends, *Atmos. Chem. Phys.*, 13, 2445-2454, doi:10.5194/acp-13-2445-2013, 2013.
- 15 Schneising, O., Burrows, J. P., Dickerson, R. R., Buchwitz, M., Reuter, M., and Bovensmann, H.: Remote sensing of fugitive methane emissions from oil and gas production in North American tight geologic formations, *Earth's Future*, 2, DOI: 10.1002/2014EF000265, pp. 11, 2014.
- 20 Turner, A. J., Jacob, D. J., Wecht, K. J., Maasakkers, J. D., Biraud, S. C., Boesch, H., Bowman, K. W., Deutscher, N. M., Dubey, M. K., Griffith, D. W. T., Hase, F., Kuze, A., Notholt, J., Ohyama, H., Parker, R., Payne, V. H., Sussmann, R., Velazco, V. A., Warneke, T., Wennberg, P. O., and Wunch, D.: Estimating global and North American methane emissions with high spatial resolution using GOSAT satellite data, *Atmos. Chem. Phys.*, 15, 7049-7069, doi:10.5194/acp-15-7049-2015, 2015.
- 25 Turner, A. J., Jacob, D. J., Benmergui, J., Wofsy, S. C., Maasakkers, J. D., Butz, A., Hasekamp, O., and Biraud, S. C.: A large increase in U.S. methane emissions over the past decade inferred from satellite data and surface observations, *Geophys. Res. Lett.*, 43, 2218-2224, doi:10.1002/2016GL067987, 2016.
- 30 Veefkind, J. P., Aben, I., McMullan, K., Förster, H., De Vries, J., Otter, G., Claas, J., Eskes, H. J., De Haan, J. F., Kleipool, Q., Van Weele, M., Hasekamp, O., Hoogeveen, R., Landgraf, J., Snel, R., Tol, P., Ingmann, P., Voors, R., Kruizinga, B., Vink, R., Visser, H., and Levelt, P. F.: 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. *Rem. Sens. Environment*, 120:70-83, 2012.

- Velazco, V. A., Buchwitz, M., Bovensmann, H., Reuter, M., Schneising, O., Heymann, J., Krings, T., Gerilowski, K., and Burrows, J. P.: Towards space based verification of CO₂ emissions from strong localized sources: fossil fuel power plant emissions as seen by a CarbonSat constellation, *Atmos. Meas. Tech.*, 4, 2809-2822, 2011.
- 5 Wecht, K. J., Jacob, D. J., Frankenberg, C., Jiang, Z., and Blake, D. R.: Mapping of North American methane emissions with high spatial resolution by inversion of SCIAMACHY satellite data, *J. Geophys. Res. Atmos.*, 119, 7741–7756, doi:10.1002/2014JD021551, 2014a.
- Wecht, K. J., Jacob, D. J., Sulprizio, M. P., Santoni, G. W., Wofsy, S. C., Parker, R., Bösch, H., and Worden, J.: Spatially resolving methane emissions in California: constraints from the CalNex aircraft campaign and from present (GOSAT, TES) and future (TROPOMI, geostationary) satellite observations, *Atmos. Chem. Phys.*, 14, 8173-8184, 10 www.atmos-chem-phys.net/14/8173/2014/, doi:10.5194/acp-14-8173-2014, 2014b.
- Wunch, D., Toon, G. C., Blavier, J.-F. L., Washenfelder, R. A., Notholt, J., Connor, B. J., Griffith, D. W. T., Sherlock, V., and Wennberg, P. O.: The Total Carbon Column Observing Network, *Phil. Trans. R. Soc. A*, 369, 2087–2112, doi:10.1098/rsta.2010.0240, 2011.
- Wunch, D., Toon, G. C., Sherlock, V., Deutscher, N. M., Liu, X., Feist, D. G., and Wennberg, P. O.: The Total Carbon 15 Column Observing Network's GGG2014 Data Version. Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA, available at: doi:10.14291/tcon.ggg2014.documentation.R0/1221662, 2015.
- Yoshida, Y., Kikuchi, N., Morino, I., Uchino, O., Oshchepkov, S., Bril, A., Saeki, T., Schutgens, N., Toon, G. C., Wunch, D., Roehl, C. M., Wennberg, P. O., Griffith, D. W. T., Deutscher, N. M., Warneke, T., Notholt, J., Robinson, J., 20 Sherlock, V., Connor, B., Rettinger, M., Sussmann, R., Ahonen, P., Heikkinen, P., Kyrö, E., Mendonca, J., Strong, K., Hase, F., Dohe, S., and Yokota, T.: Improvement of the retrieval algorithm for GOSAT SWIR XCO₂ and XCH₄ and their validation using TCCON data, *Atmos. Meas. Tech.*, 6, 1533–1547, doi:10.5194/amt-6-1533-2013, 2013.

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Product	Sensor	Algorithm	Version	Institute	References
CH4_SCI_WFMD	SCIAMACHY on ENVISAT	WFM-DOAS (WFMD)	4.0	IUP, Univ. Bremen	Buchwitz et al., 2000; Schneising et al., 2011, 2012, 2013
CH4_SCI_IMAP	SCIAMACHY on ENVISAT	IMAP-DOAS (IMAP)	7.1	JPL/SRON	Frankenberg et al., 2005, 2006, 2008a, 2008b, 2011
CH4_GOS_OCPR	TANSO-FTS on GOSAT	UoL-Proxy	6.0	Univ. Leicester	Parker et al., 2011
CH4_GOS_SRFP	TANSO-FTS on GOSAT	RemoTeC	2.3.7	SRON/KIT	Butz et al., 2011

Table 1. Overview of the used satellite XCH₄ data products.

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Source region	Latitude range [deg]	Longitude range [deg]	Mexp (*) [-]	Length L [km]	Overall conversion factor CF (*) [MtCH ₄ /yr/ppb]
Four Corners	36.2 – 37.4	109.6W - 107.0W	0.79	176.5	0.0518
Central Valley (southern part)	35.0 – 37.0	120.0W – 118.5W	0.94	174.4	0.0605
Azerbaijan	Country shape		0.94	294.3	0.1026
Turmenistan	Country shape		0.98	698.6	0.2529

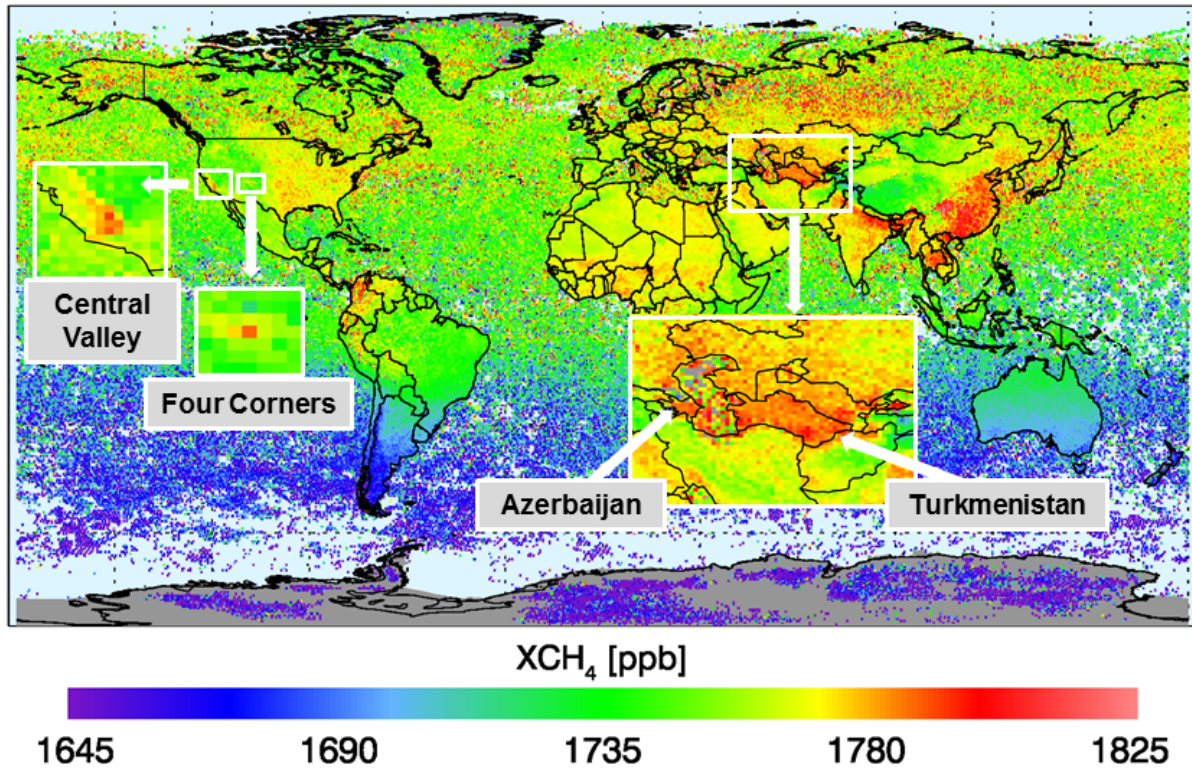
Table 2. Details related to the four source regions and their parameters as used for the emission estimation. (*) Approximate values (the exact values depend on the sampling of the satellite data in the source region, which depends on satellite product and year).

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Source region	Estimated methane emissions [MtCH ₄ /year]				Comments / Other estimates
	SCIAMACHY		GOSAT		
	WFMD	IMAP	OCPR	SRFP	
Four Corners	0.50 [0.40, 0.59]	0.57 [0.34, 0.80]	0.45 [0.14, 0.76]	0.42 [0.20, 0.64]	Kort et al., 2014 (*): 0.59 [0.54, 0.64] Turner et al., 2015: [0.45, 1.39] EDGAR v4.2: 0.17
Central Valley (southern part)	1.05 [0.53, 1.57]	1.10 [0.92, 1.28]	1.35 [0.96, 1.75]	1.55 [1.15, 1.95]	EDGAR v4.2: 0.19 7 Jeong et al., 2013: 0.85 – 0.94 (for their region R12)
Azerbaijan	0.60 [-0.01, 1.21]	0.53 [0.23, 0.83]	0.51 [-0.16, 1.18]	-	EDGAR v4.2 (FT2012): 0.74
Turkmenistan	1.89 [1.22, 2.55]	1.93 [1.66, 2.19]	2.08 [1.67, 2.49]	1.85 [1.31, 2.39]	EDGAR v4.2 (FT2012): 1.33

Table 3. Summary of estimated methane emissions in terms of annual mean value and 1-sigma range obtained from computing the standard deviation of the annual emissions. The satellite-derived annual methane emissions are covering the time period 2003-2009 for SCIAMACHY and 2009-2014 for GOSAT. (*) Kort et al., 2014, report the 2-sigma range [0.50, 0.67], not the (approximate) 1-sigma range listed here.

Methane SCIAMACHY/ENVISAT WFMD



5 **Figure 1.** Year 2004 SCIAMACHY WFMD XCH₄ at 0.5°x0.5° resolution. The ~~source~~three-target regions studied in this manuscript are indicated: Central Valley, California, USA, the Four Corners area in the southwestern USA, Azerbaijan and Turkmenistan.

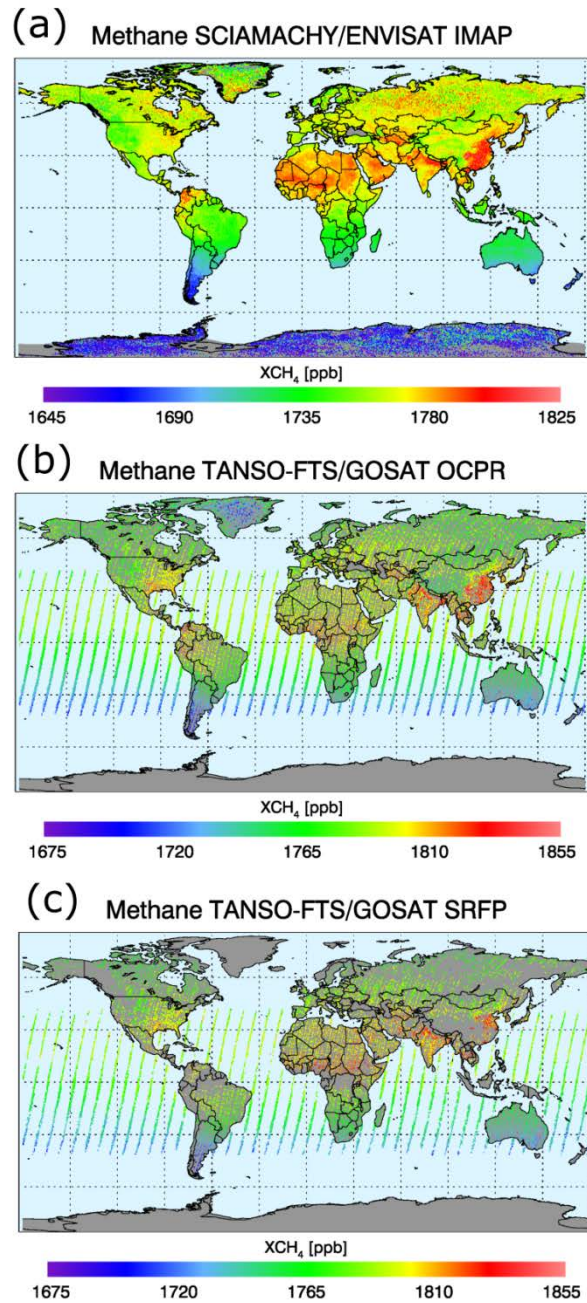


Figure 2. As Fig. 1 but for (a) SCIAMACHY IMAP XCH₄, (b) year 2010 GOSAT OCPR XCH₄ and (c) year 2010 GOSAT SRFP (“RemoTeC”) XCH₄.

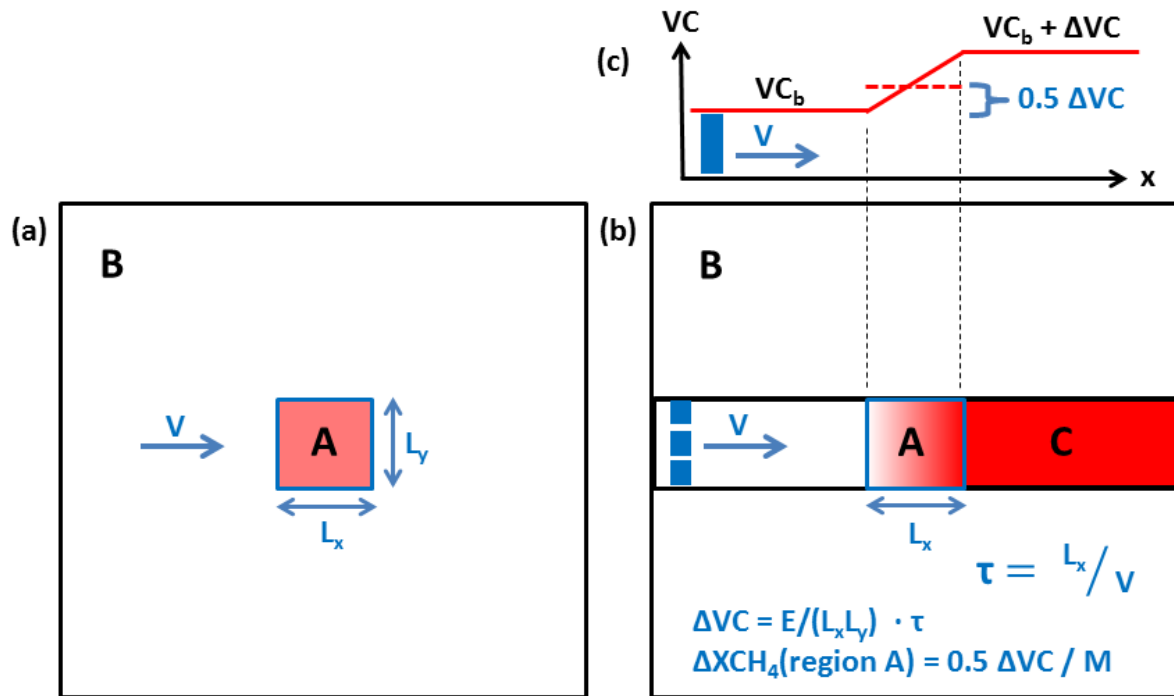


Figure 3A1. Sketch of a simple model used to explain the methane emission estimation method described in Sect. 3. (a) Source region A (of size $L_x L_y$ and with L_x in wind speed direction (wind speed magnitude V)) with elevated X_{CH_4} (light red) and surrounding (background) region B (white area). (b) Air parcels (blue squares) moving with constant speed V over a source region with emission $E/(L_x L_y)$, where E is the source area emission in CH_4 mass per time, while accumulating methane during accumulation time $\tau (= L_x/V)$. (c) Before entering the source region, the air parcels are characterized by a background methane vertical column, VC_b , in units of CH_4 mass per area. When leaving the source area their vertical column has been enhanced by $\Delta VC = E/(L_x L_y) \cdot \tau$. When passing over the source region, their vertical column increases linearly and, therefore, the average column enhancement over the source region is $0.5 \cdot \Delta VC$. VC (CH_4 mass per area) can be converted to X_{CH_4} (ppb) via a factor M (in units of mass and per area and per ppb).

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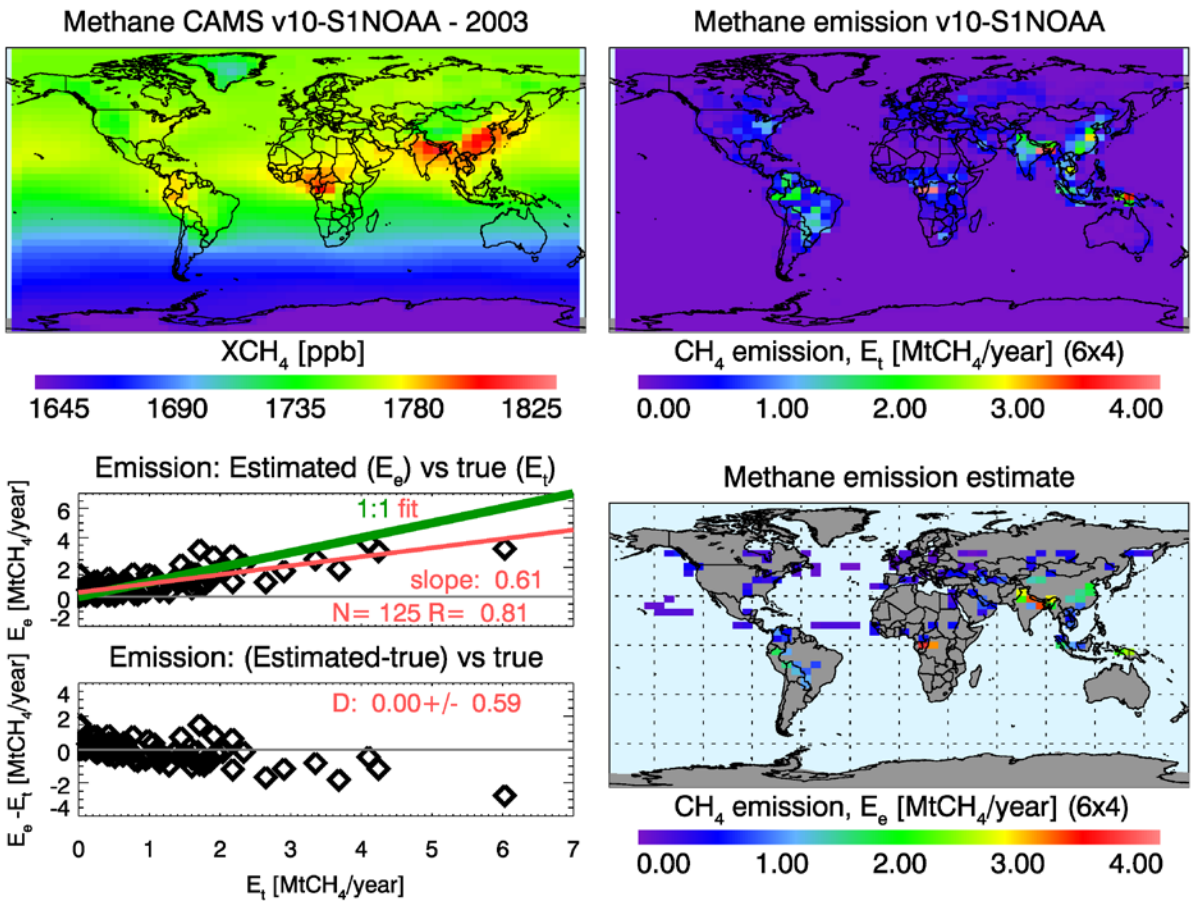


Figure 43. Methane emissions (in $\text{MtCH}_4/\text{year}$) and corresponding XCH_4 (in ppb) for the year 2003 at 6° longitude times 4° latitude resolution. Top left: XCH_4 as computed from Copernicus Atmosphere Monitoring Service (CAMS) atmospheric CH_4 fields (version v10-S1NOAA; resolution: $6^\circ \times 4^\circ$; obtained from <https://atmosphere.copernicus.eu/>). Top right: Corresponding CAMS total, i.e., anthropogenic and natural, methane emissions. Map bottom right: Methane emissions of (automatically determined potential) emission hot spots (“hotspot cells”) as derived from the top left XCH_4 map using the method described in Sect. 3. Bottom left: Comparison of retrieved emissions (map bottom right) with the “true” CAMS emissions (map top right). Here N ($= 125$) denotes the number of grid cells for which emission values have been obtained (“hotspot cells”, see main text for details), R ($= 0.81$) is the linear correlation coefficient of retrieved and true emissions, and D is the difference between the retrieved and the true emissions in terms of mean difference and standard deviation ($0.00 \pm 0.59 \text{ MtCH}_4/\text{year}$).

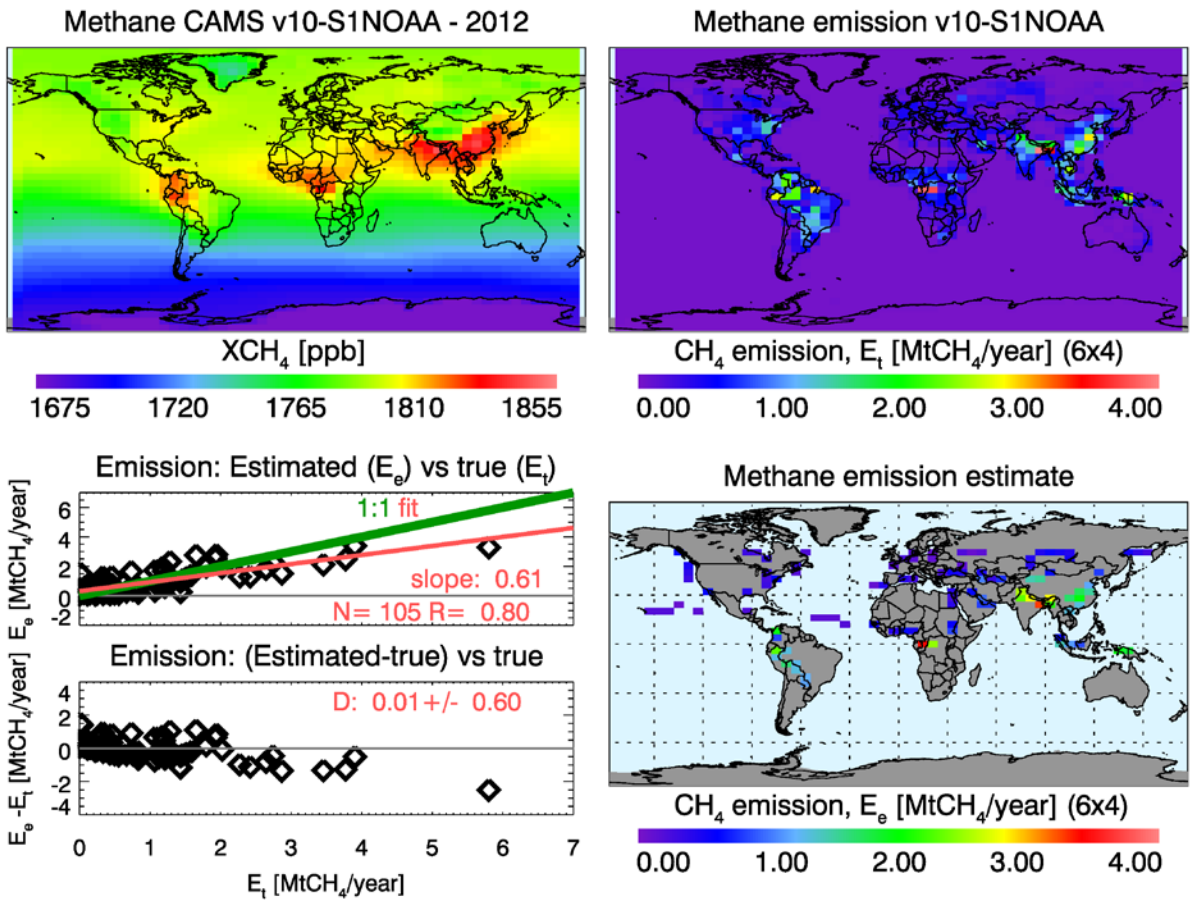


Figure 54. As Fig. 43 but for year 2012.

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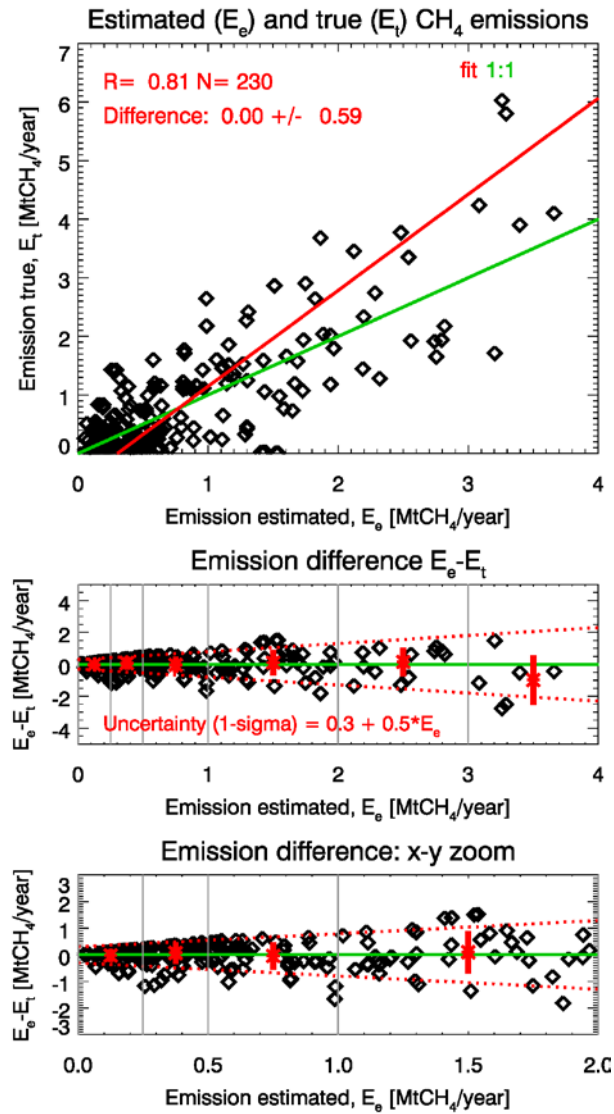


Figure 65. Top: “True” (i.e., CAMS) emission, E_t , versus estimated emissions, E_e , as obtained from the simulation-based assessment results shown in Figs. 43 and 54 (i.e., shown are all “hotspot cells” also shown in these two figures, see caption Fig. 43 and main text for details). Middle and bottom: Emission difference “estimated minus true” versus estimated emission. The grey vertical bars denote the boundaries of emission bins for which mean differences (red crosses) and standard deviations of the differences (red vertical lines) have been computed. The red dotted line shows that the relationship between the estimated emission (E_e) and its 1-sigma uncertainty (σ) can be approximately described by $\sigma(E_e) = 0.3 + 0.5 E_e$.

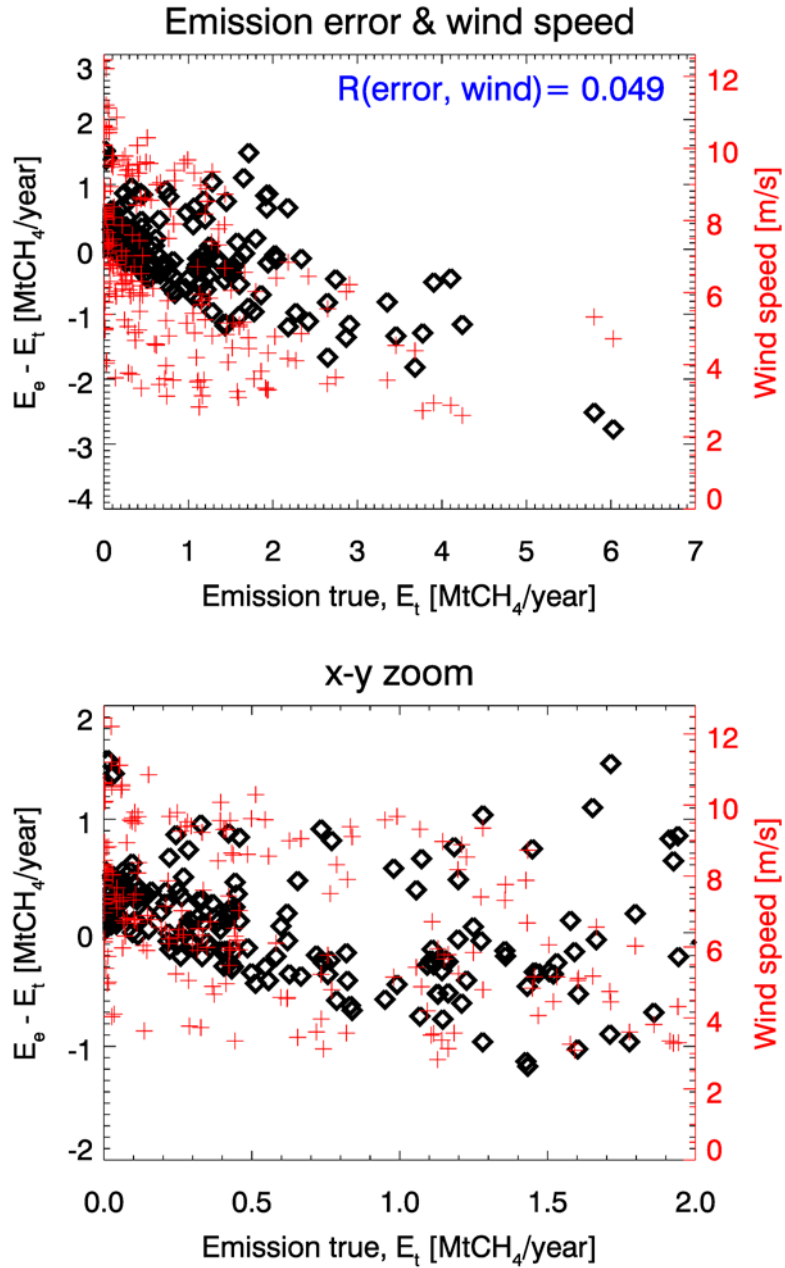


Figure 7. Error of the estimated emission (black symbols; computed as “retrieved – true”, see Fig. 6) versus annual mean wind speed (red crosses) at 900 hPa. Top: all data; bottom: same data but x-y zoom. The linear correlation coefficient between annual emission error and annual mean wind speed is 0.049.

Methane GCT15 / USA - 2010

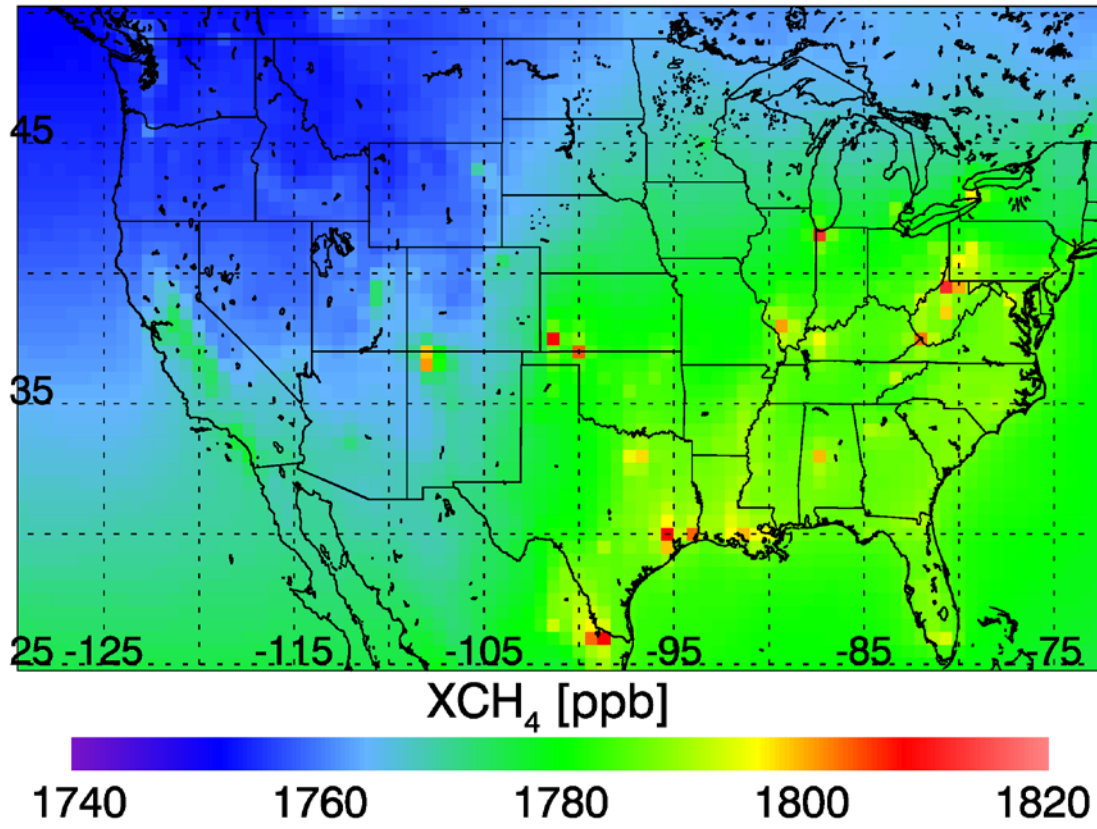


Figure 8. Annually averaged (year 2010) atmospheric column-averaged methane (XCH_4) computed with GEOS-Chem using *a posteriori* methane emissions of Turner et al., 2015 (“GCT15 data set”). The resolution of this data set is 0.5° latitude x 0.667° longitude.

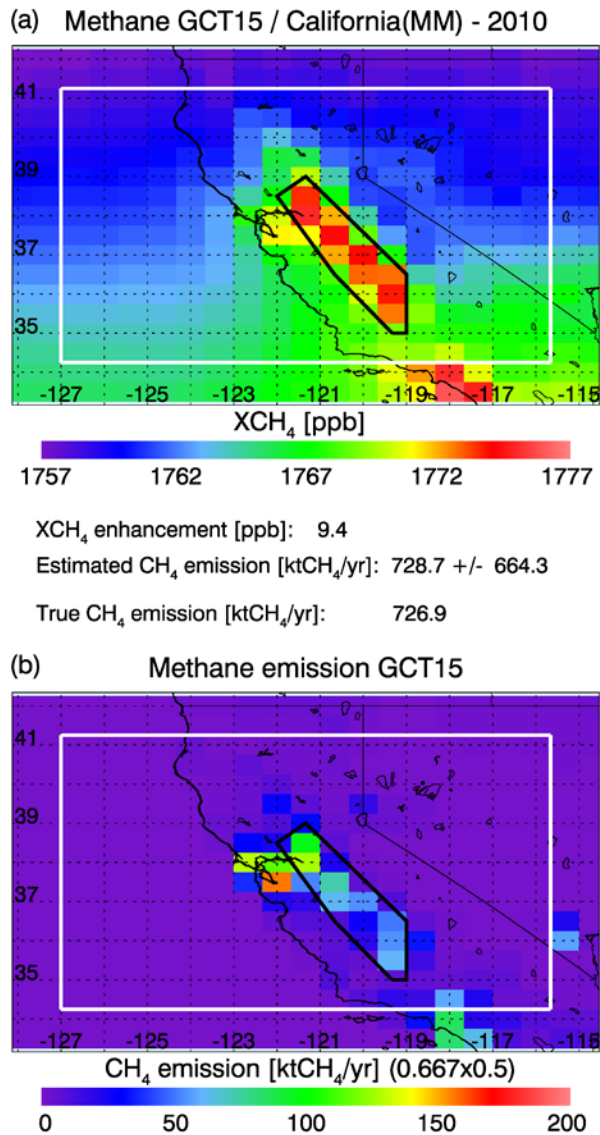
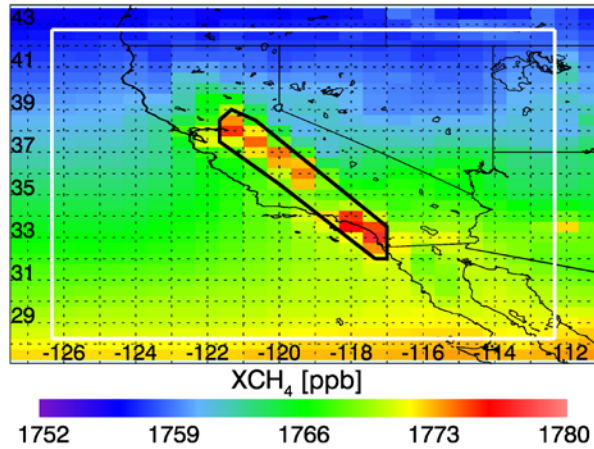


Figure 9. Top: GCT15 XCH₄ over parts of California. The white rectangle denotes the “surrounding region” of the “source region”, which is surrounded by a polygon shown as thick black line. The source region covers the area between the two cities Modesto and Merced in central California. The text below lists the XCH₄ enhancement, Δ XCH₄ (9.4 ppb) and the estimated emission (729 +/- 664 ktCH₄/yr). The “true” emission of the source region has been computed from the GCT15 emissions (bottom panel) and is 727 ktCH₄/yr.

(a) Methane GCT15 / California(MS) - 2010



XCH₄ enhancement [ppb]: 7.2

Estimated CH₄ emission [ktCH₄/yr]: 770.0 +/- 685.0

True CH₄ emission [ktCH₄/yr]: 1228.0

(b) Methane emission GCT15

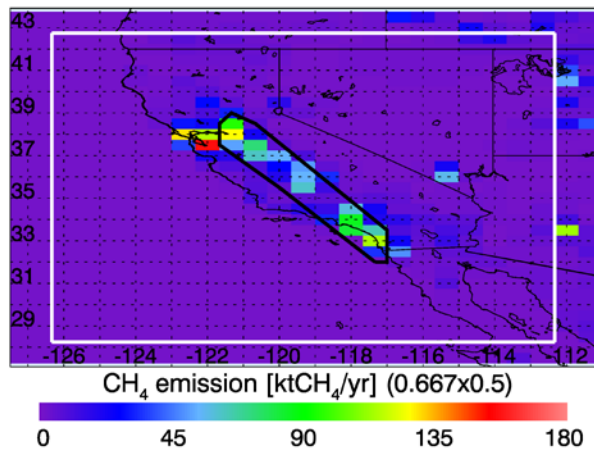


Figure 10. As Fig. 9 but for a larger part of California, referred to as “California Mid/South (MS)” in this publication.

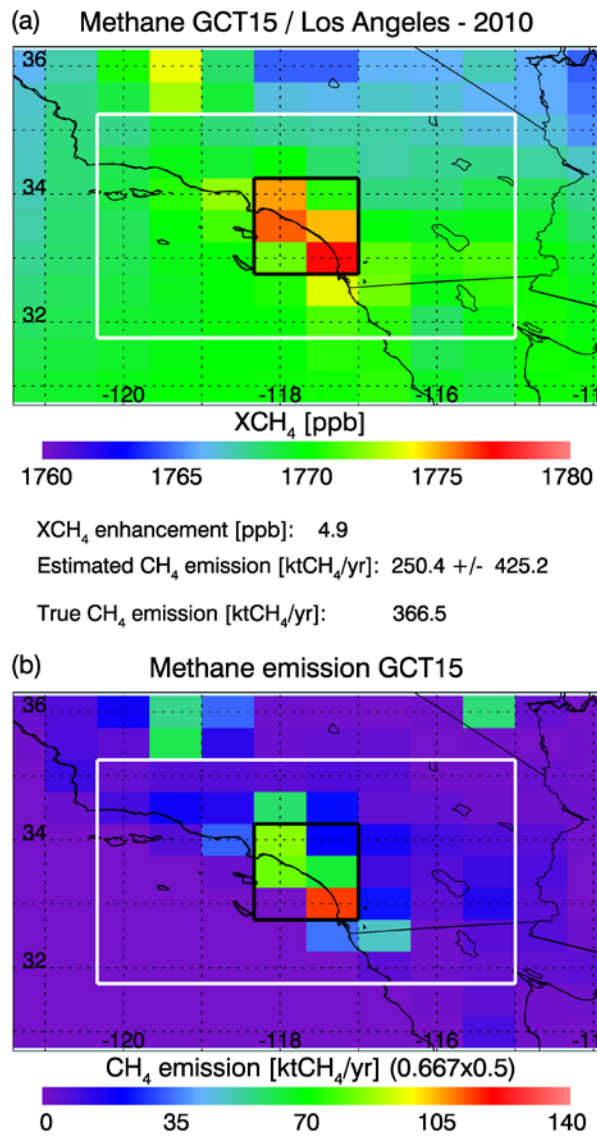
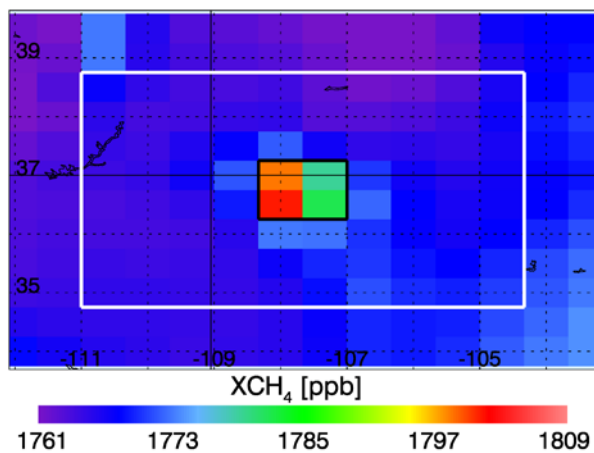


Figure 11. As Fig. 9 but for the region around Los Angeles, California (this region is located in the southern part of the source region shown in Fig. 10).

(a) Methane GCT15 / FourCorners - 2010



XCH₄ enhancement [ppb]: 23.8

Estimated CH₄ emission [ktCH₄/yr]: 794.8 +/- 697.4

True CH₄ emission [ktCH₄/yr]: 1404.2

(b) Methane emission GCT15

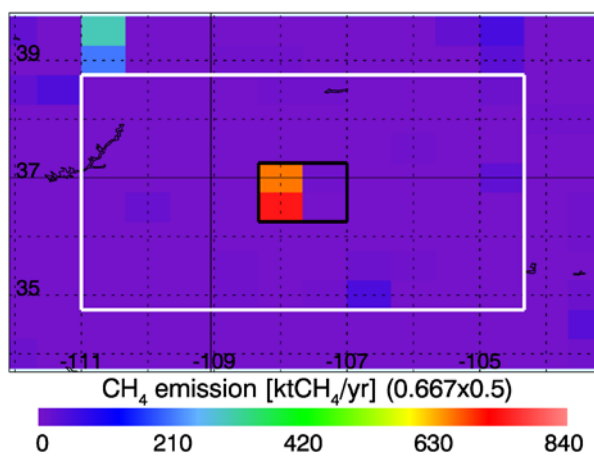


Figure 12. As Fig. 9 but for the Four Corners region (see main text for details).

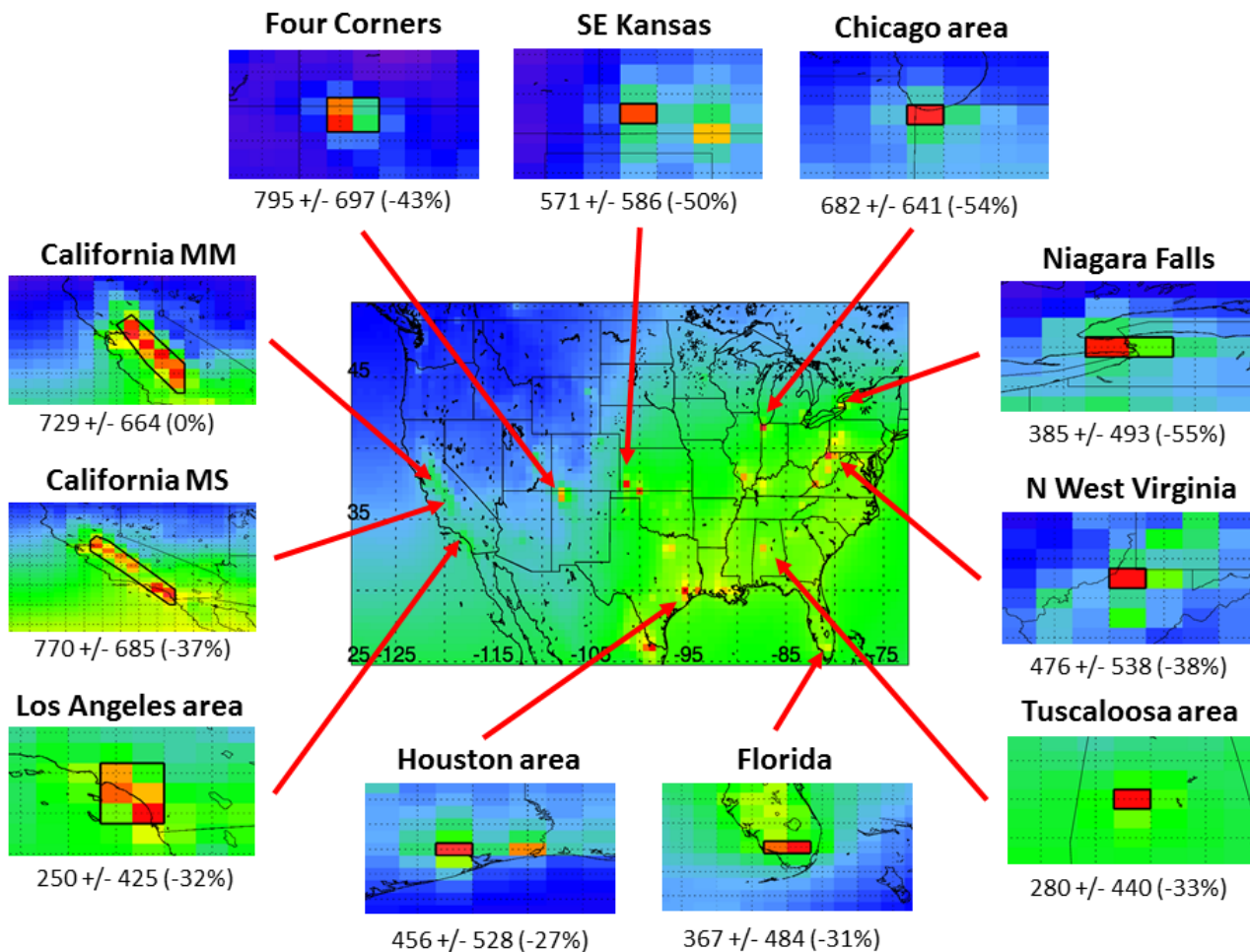


Figure 13. Methane emission estimates for several methane hot spot areas in the USA as obtained by applying our simple mass balance method to the year 2010 GCT15 data set. The figure in the center is identical with Fig. 8 and shows year 2010 XCH₄ over the USA. For each hotspot region the following three numbers are listed below each map: Estimated methane and its uncertainty in ktCH₄/yr for the shown source regions (thick black lines, mostly rectangles). The number in brackets is the percentage difference of the estimated emission and the corresponding true emission (computed as “(estimated – true)/true”), where the true emission is the source region GCT15 emission.

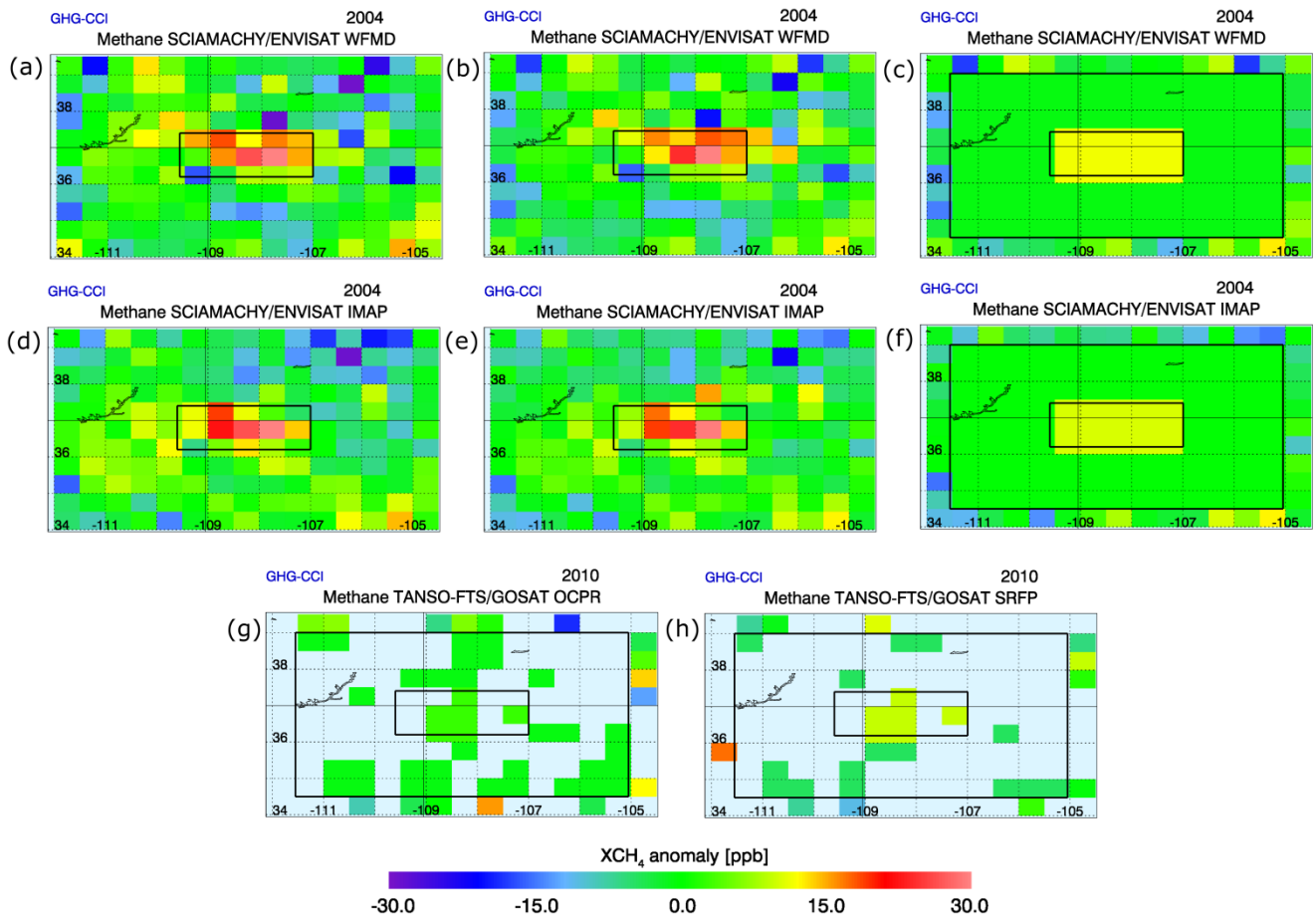
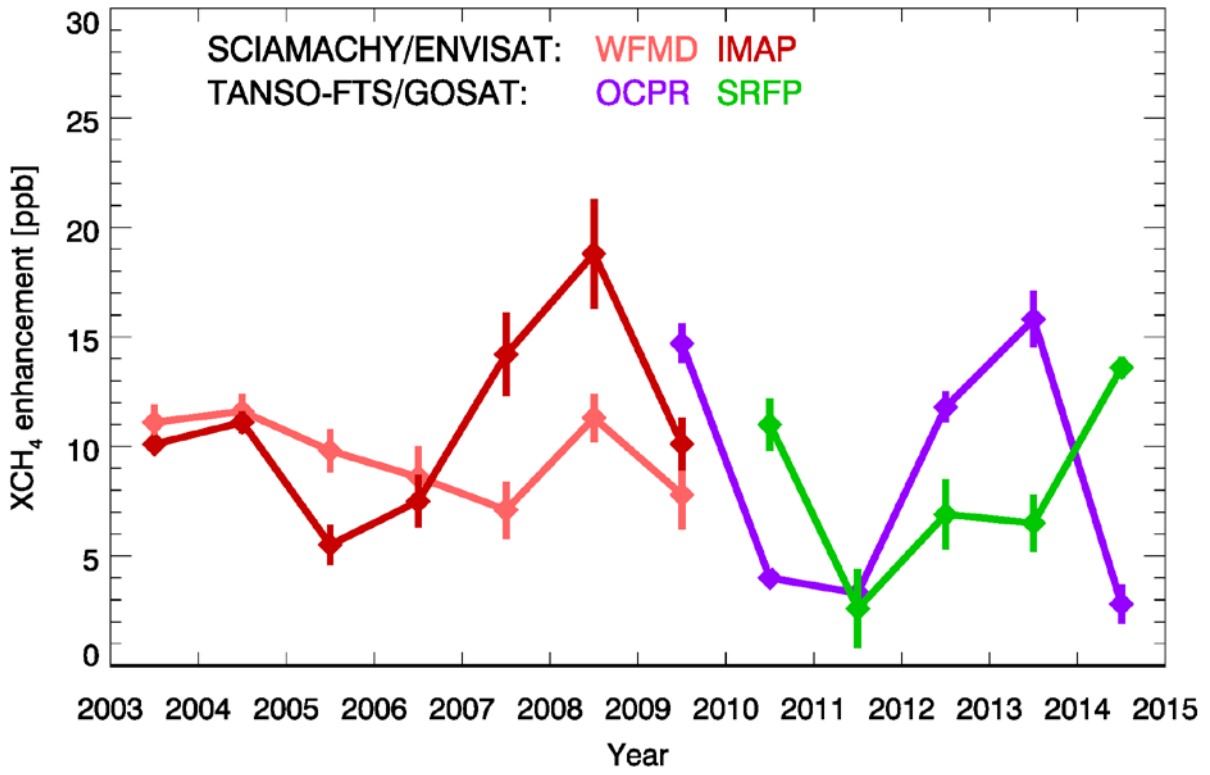


Figure 146. Satellite-derived XCH_4 anomalies (i.e., the mean value of XCH_4 has been subtracted) in and around the Four Corners region. Top row: SCIAMACHY WFMD year 2004 XCH_4 anomaly at $0.5^\circ \times 0.5^\circ$ resolution. (a) Originally gridded data. The black rectangle indicates the assumed source area (taken from Kort et al., 2014). (b) As (a) but after elevation correction (see main text for details). (c) As (b) but replacing the individual XCH_4 values by their averages in the indicated source region (inner rectangle) and its surrounding (outer rectangle). The difference between these two values defines the methane enhancement of the source region, i.e., ΔXCH_4 . Middle row: As top row but for IMAP XCH_4 . Bottom row: As last column of first two rows but for GOSAT OCPR (g) and SRFP (h) for the year 2010.

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Methane enhancement - Four Corners, USA



Methane enhancement - Four Corners, USA

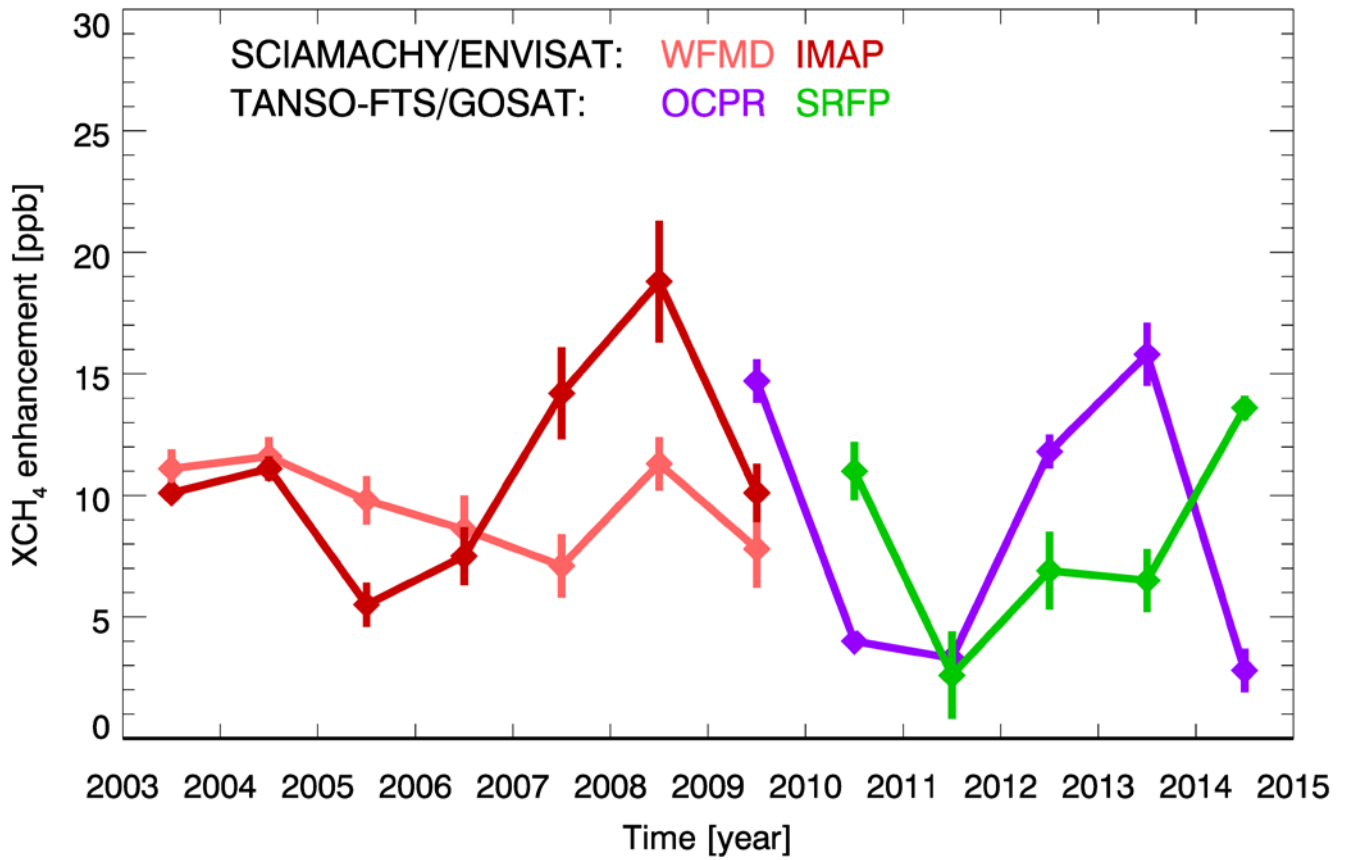
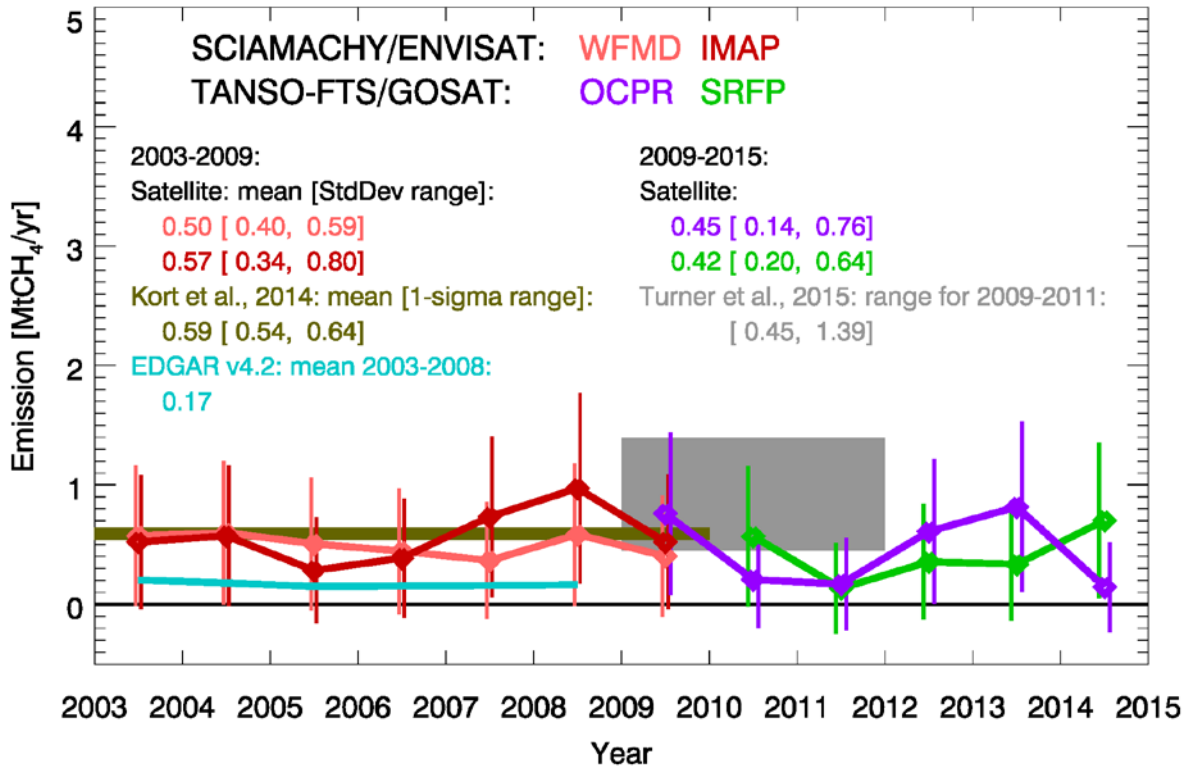


Figure 157. Methane enhancements over the Four Corners area for all years and all four satellite data products used in this study. The error bars show the standard deviation of the methane enhancements obtained by varying the size and shape of the surrounding area.

Methane emission estimates - Four Corners, USA



Methane emission estimates - Four Corners, USA

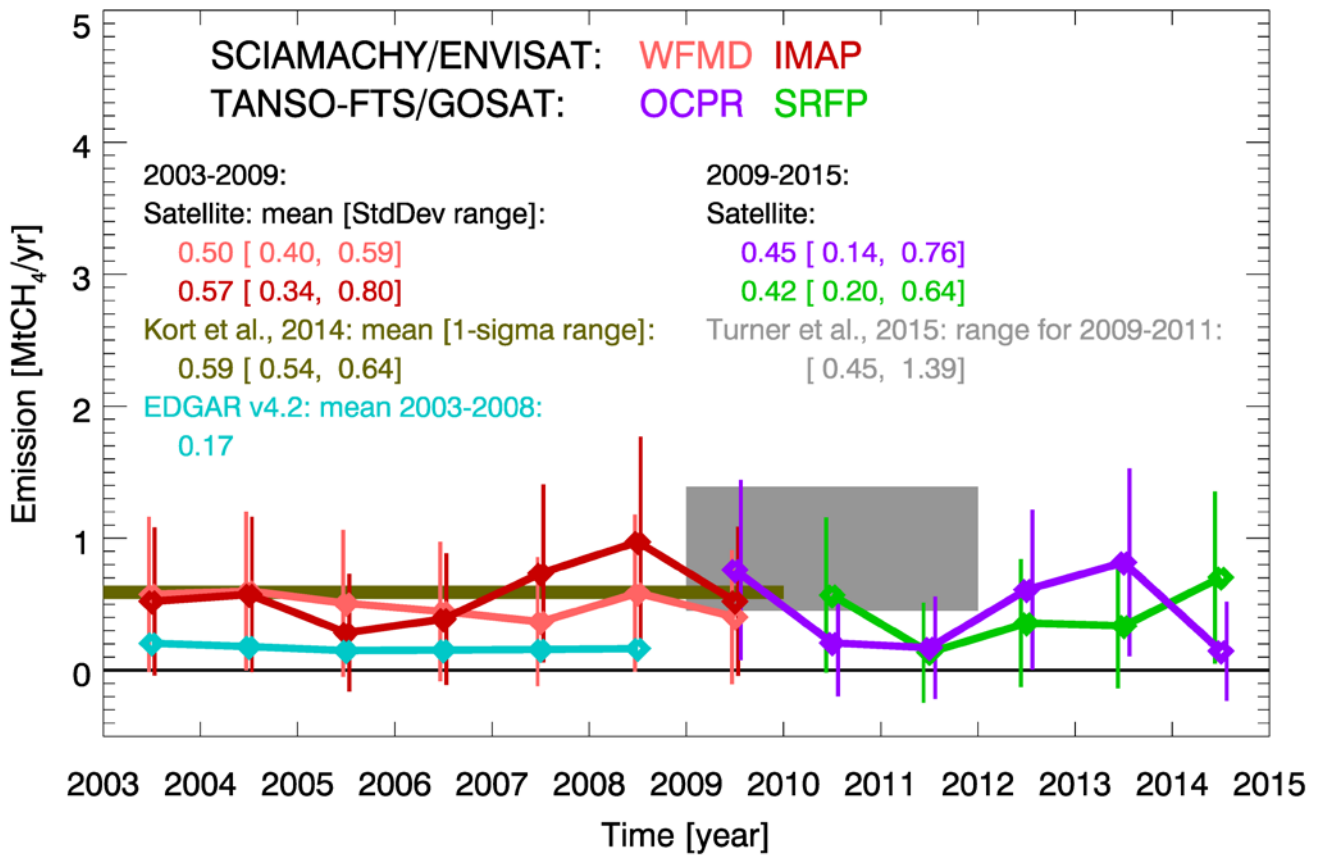


Figure 168. Methane emission estimates for Four Corners as obtained from the methane enhancements shown in Fig. 157. Shown here are the satellite-derived annual methane emissions and their 1-sigma uncertainty as derived from the four satellite data products used in this study using the method described in Sect. 3. The listed numerical values for the satellite-derived emissions are the mean value and a range defined as mean value plus/minus one times the standard deviation of the annual averages. The results are compared with published values as listed in Kort et al., 2014, (for 2003-2009; shown in dark green) and Turner et al., 2015 (for 2009-2011; shown in grey). Also shown are the EDGAR v4.2 total anthropogenic emissions during 2003-2008 (in light blue). It needs to be pointed out that the estimated emissions using satellite data are total methane emissions whereas EDGAR is (only) anthropogenic.

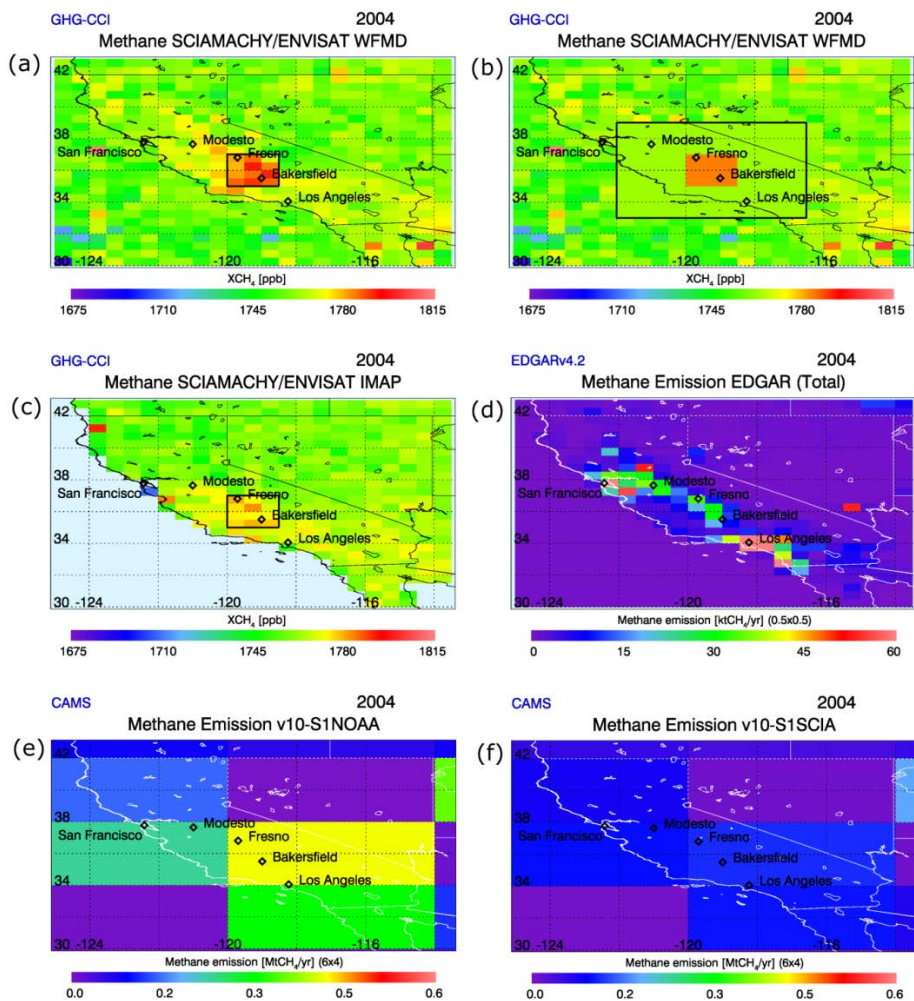
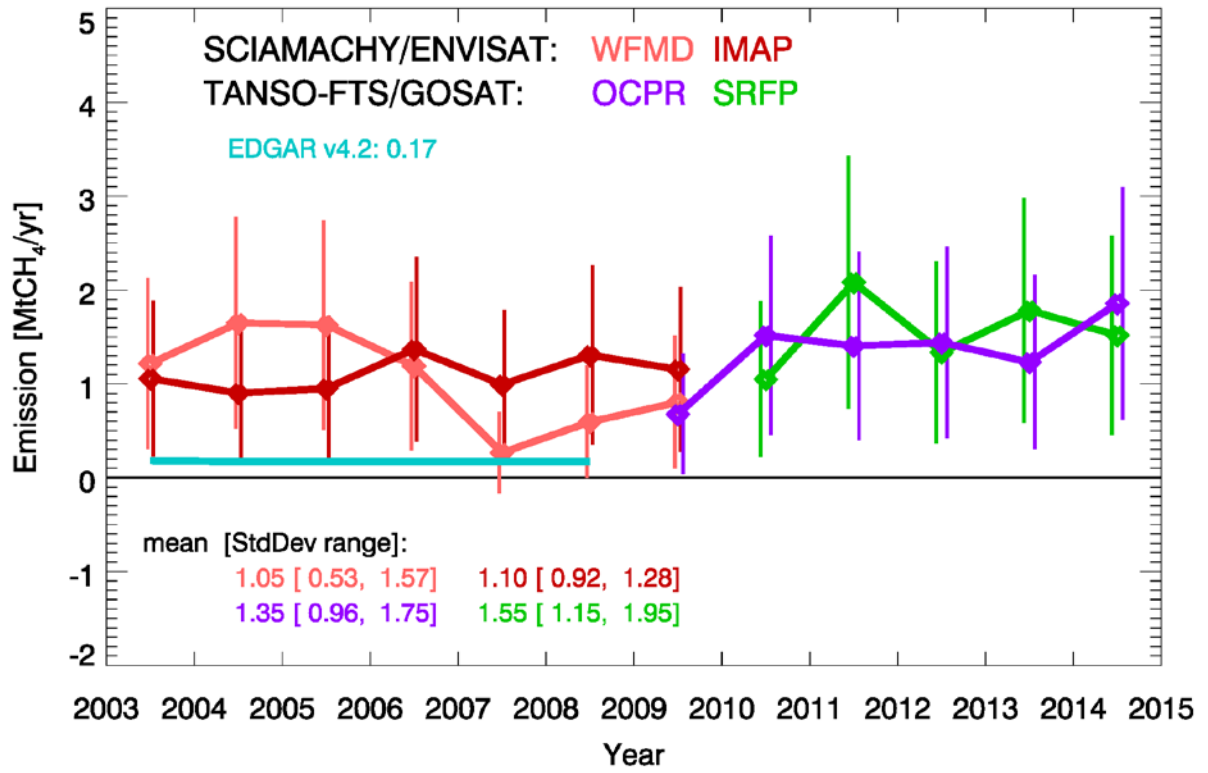


Figure 179. Methane maps for Central Valley, California. (a) SCIAMACHY year 2004 WFMD XCH₄ at 0.5°x0.5° resolution. The rectangle shows the chosen source region. (b) As (a) but showing the source region (inner rectangle) and the default background region (outer rectangle) with their corresponding XCH₄ mean values. (c) As (a) but for IMAP. (d) EDGAR v4.2 year 2004 total anthropogenic methane emissions (regridged to 0.5°x0.5° resolution). (e) CAMS v10-S1NOAA year 2004 total methane, i.e., anthropogenic and natural, emissions obtained by assimilation of NOAA methane observations (at 6°x4°). (f) As (e) but for CAMS version v10-S1SCIA, i.e., including the assimilation of SCIAMACHY IMAP retrievals in addition to the assimilation of NOAA data.

Methane emission estimates - Central Valley, CA, USA



Methane emission estimates - Central Valley, CA, USA

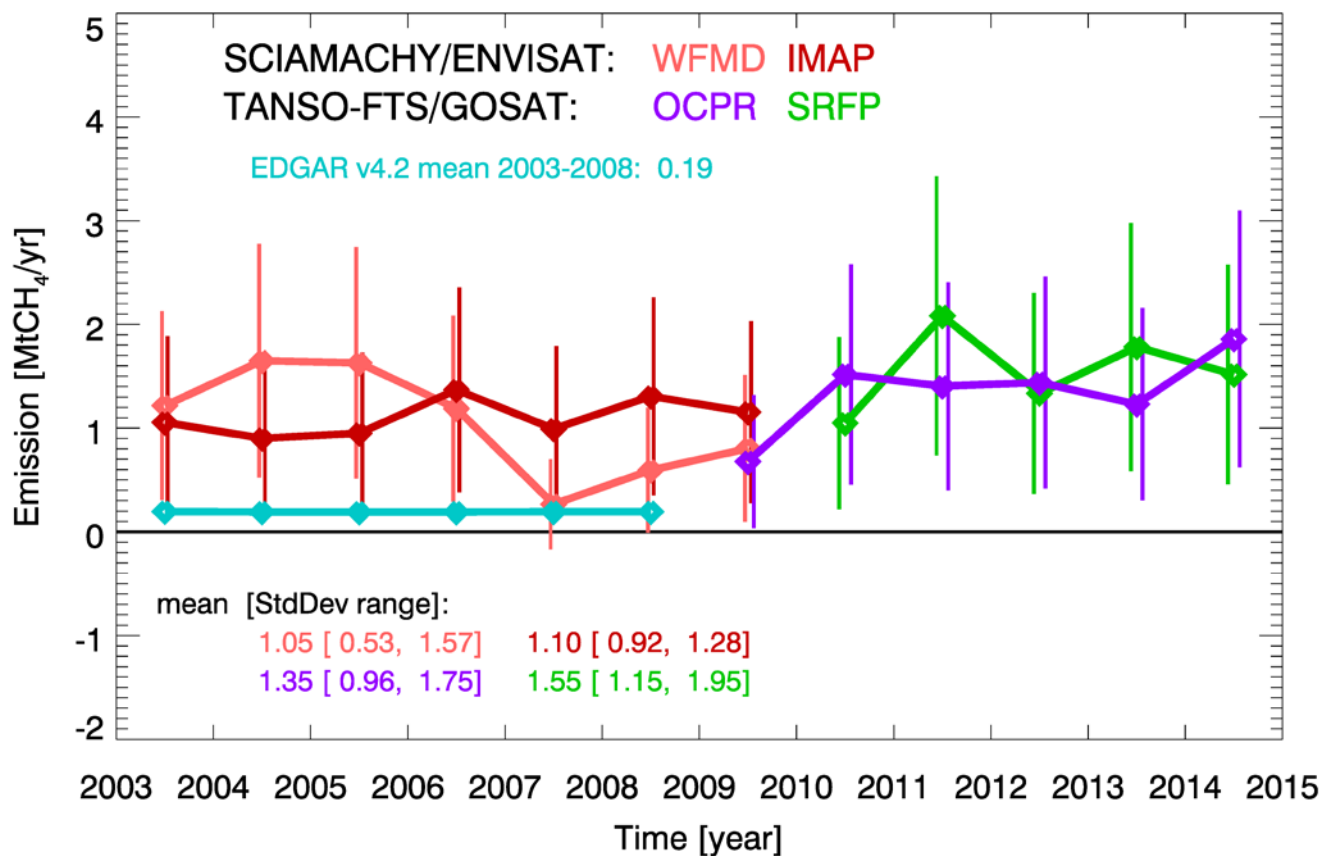


Figure 189. Methane emission estimates for Central Valley area in California, USA, as defined for this study (see Fig. 179 and Tab. 2). The blue line shows the EDGAR v4.2 (annual) anthropogenic methane emissions as computed for the Central Valley source region.

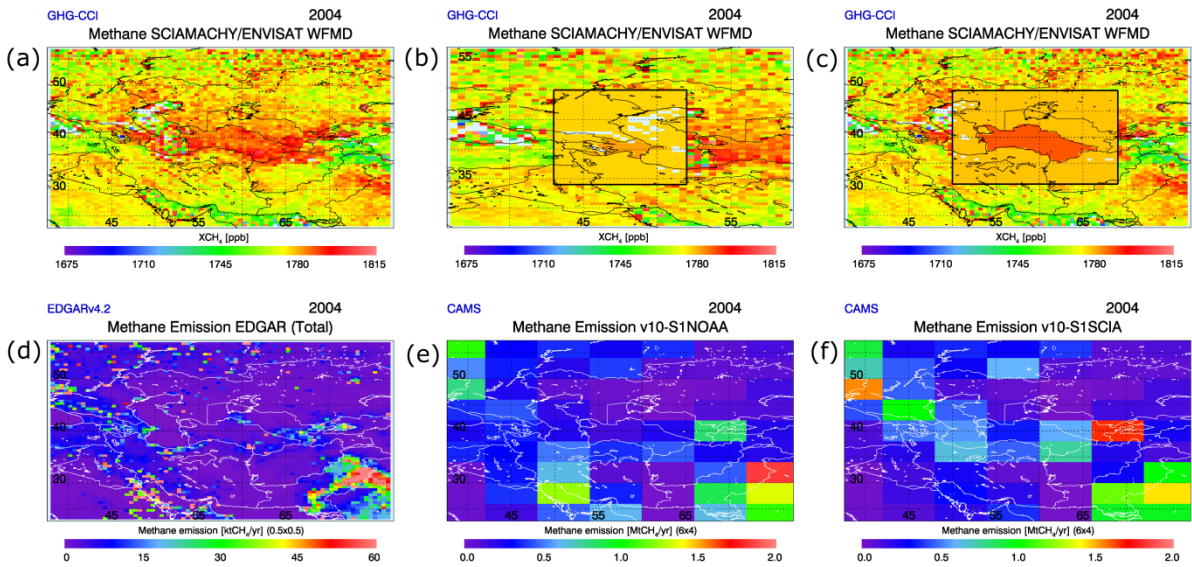
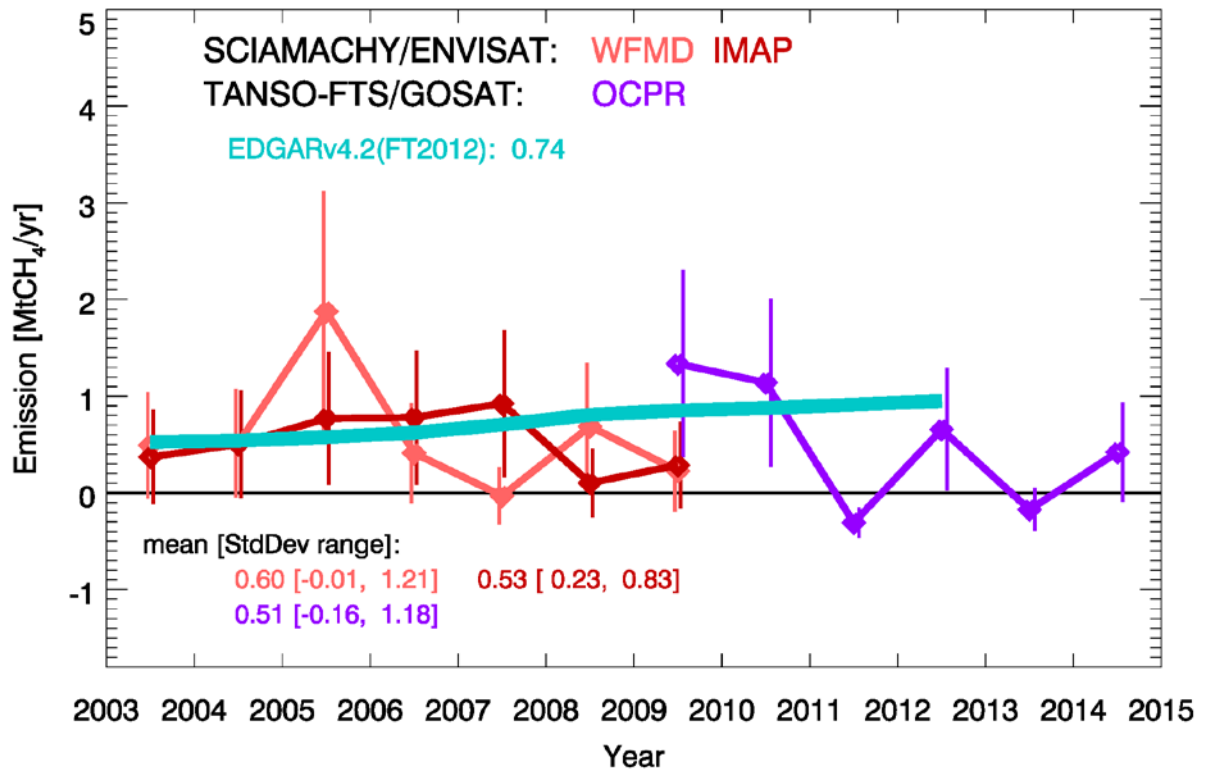


Figure 191. Top row: (a) SCIAMACHY WFMD year 2004 XCH₄ in and around Azerbaijan and Turkmenistan (resolution 5 0.5°x0.5°). (b) As (a) but showing the Azerbaijan source region (entire country of Azerbaijan) and the default background region (rectangle) (please note that this map is shifted relative to all other maps shown in this figure to place Azerbaijan in the center of the map). (c) As (a) but showing the Turkmenistan source and default background regions. Bottom row: (d): 10 EDGAR v4.2 year 2004 total anthropogenic methane emissions (at 0.5°x0.5° resolution). (e) CAMS year 2004 total anthropogenic and natural methane emissions based on assimilation of NOAA data (at 6°x4° resolution). (f) As (e) but with additional assimilation of SCIAMACHY IMAP XCH₄.

Methane emission estimates - Azerbaijan



Methane emission estimates - Azerbaijan

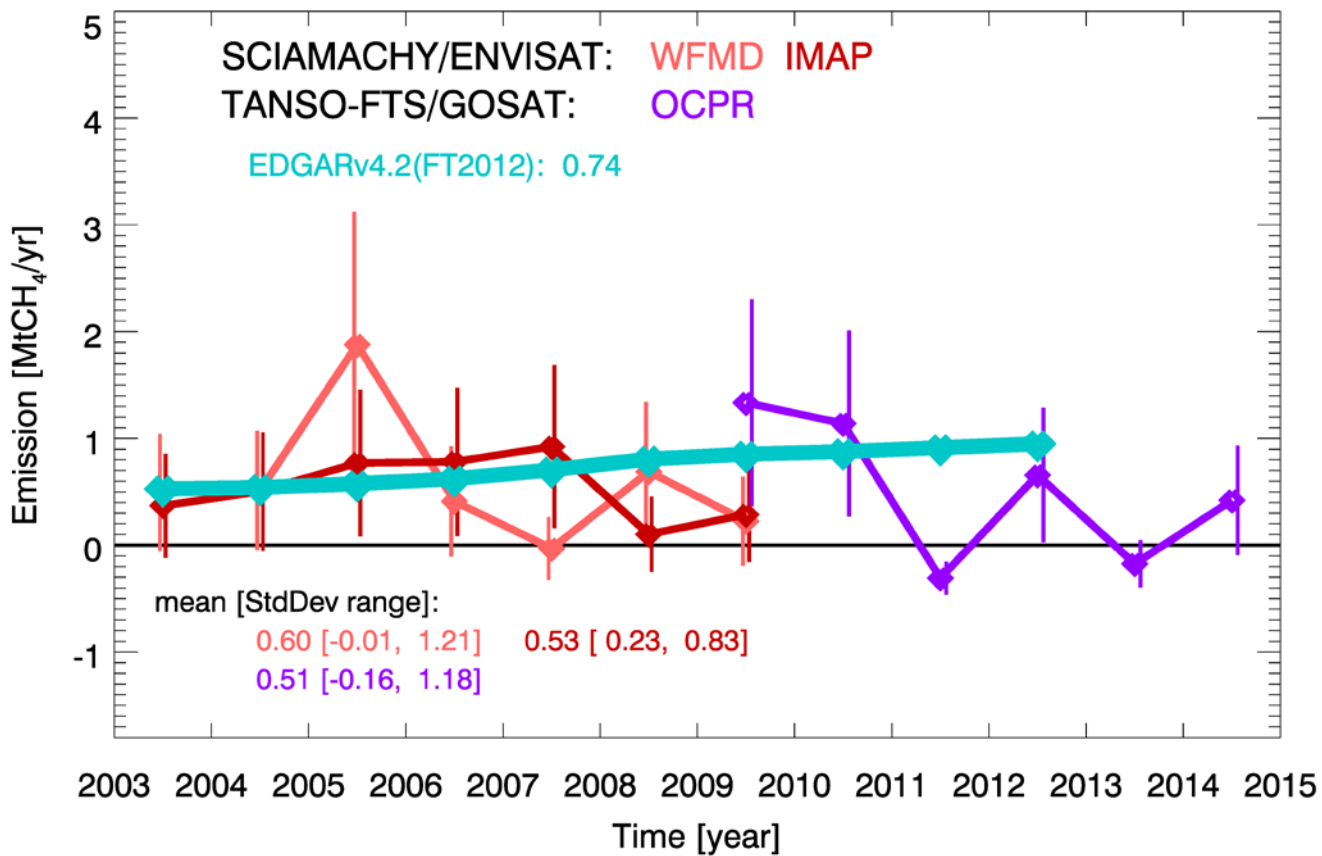
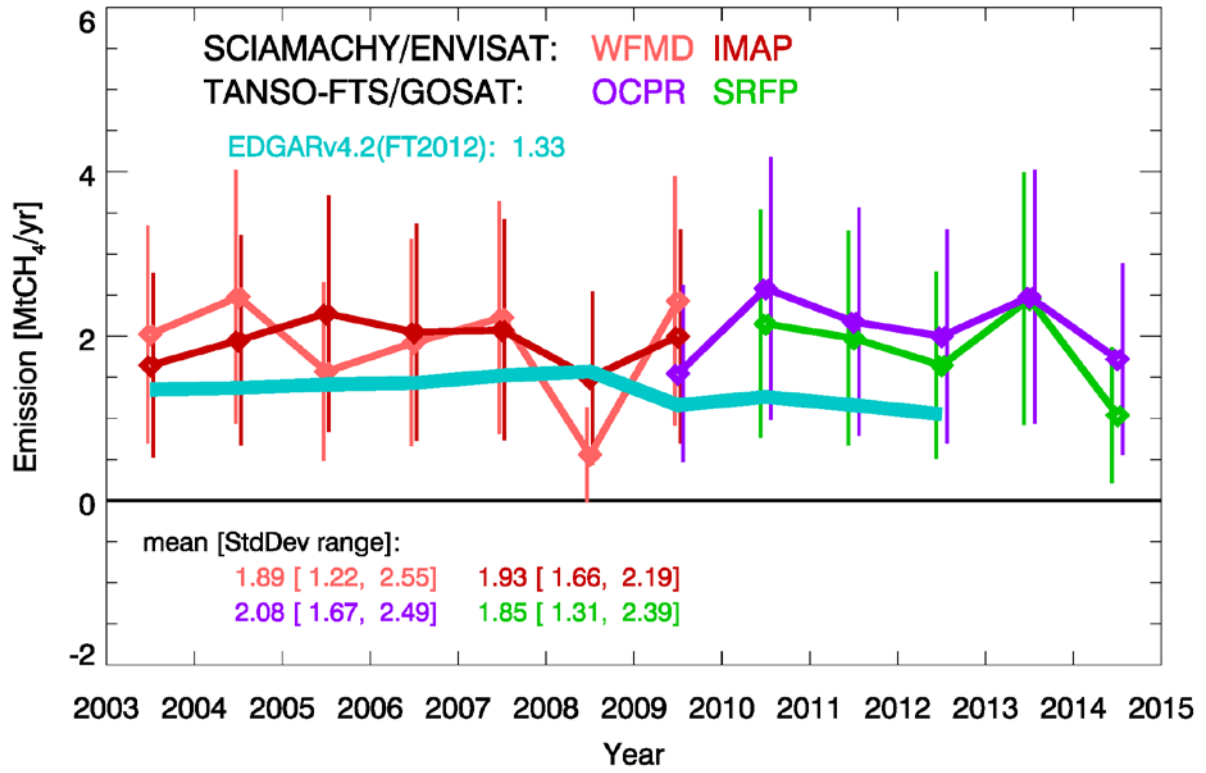


Figure 2012. Annual methane emission estimates for Azerbaijan (see also Fig. 194). The blue line shows the EDGAR v4.2 (FT2012) annual emissions for Azerbaijan.

Methane emission estimates - Turkmenistan



Methane emission estimates - Turkmenistan

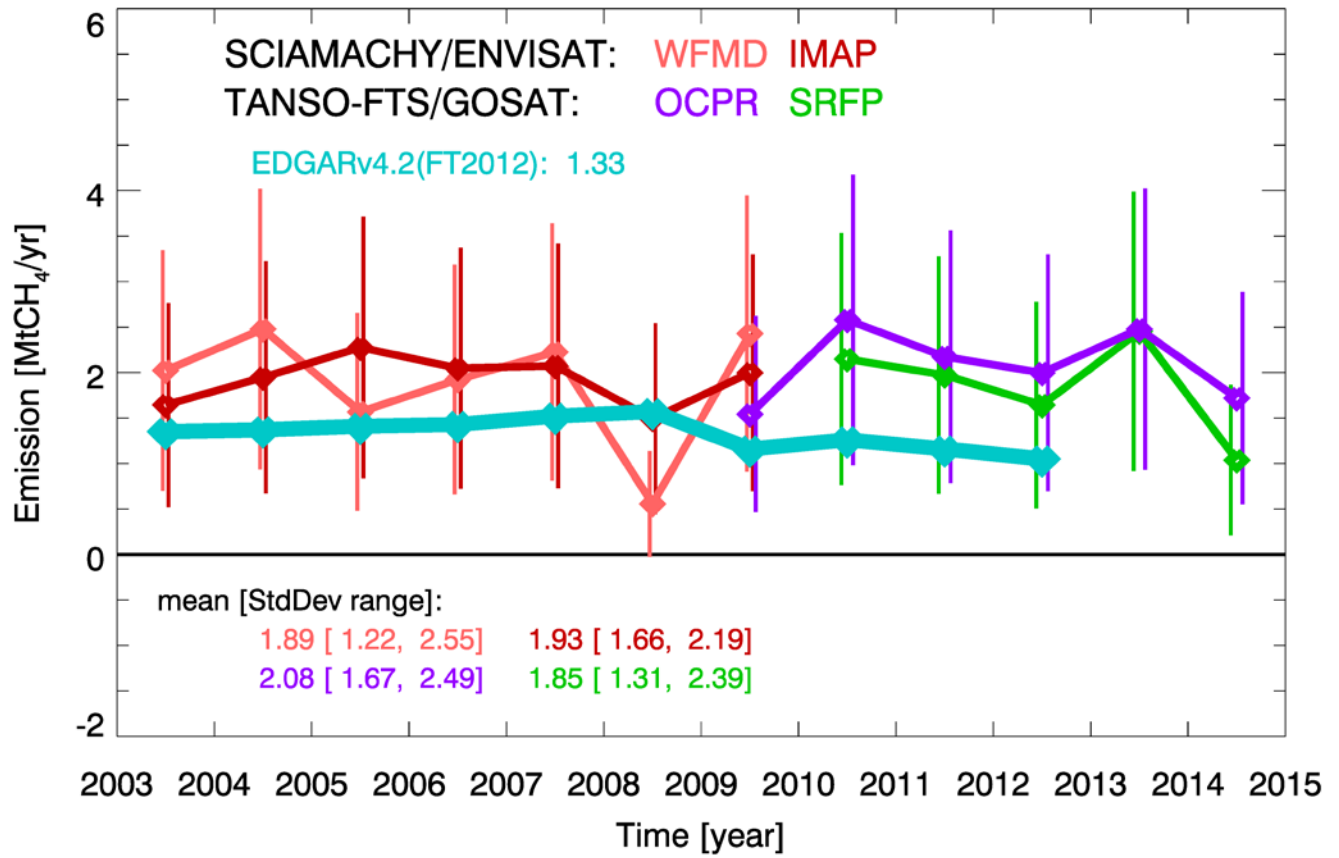


Figure 2143. Annual methane emission estimates for Turkmenistan (see also Fig. 194). The blue line shows the EDGAR v4.2 (FT2012) annual emissions for Turkmenistan.

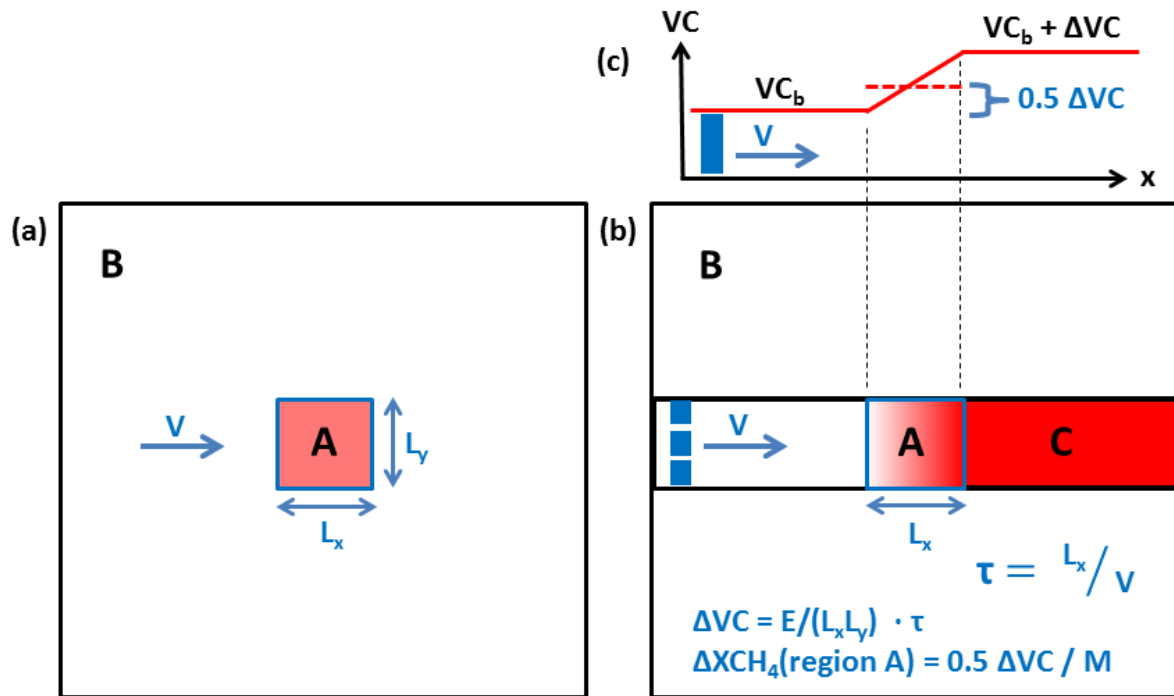


Figure A1. Sketch of a simple model used to explain the methane emission estimation method described in Sect. 3. (a) Source region A (of size L_x, L_y , and with L_x in wind speed direction (wind speed magnitude V)) with elevated X_{CH_4} (light red) and surrounding (background) region B. (b) Air parcels (blue squares) moving with constant speed V over a source region with emission $E/(L_x L_y)$, where E is the source area emission in CH_4 mass per time, while accumulating methane during accumulation time $\tau (= L_x/V)$. (c) Before entering the source region, the air parcels are characterized by a background methane vertical column, VC_b , in units of CH_4 mass per area. When leaving the source area their vertical column has been enhanced by $\Delta VC = E/(L_x L_y) \cdot \tau$. When passing over the source region it increases linearly and, therefore, the average column enhancement over the source region is $0.5 \Delta VC$. VC (CH_4 mass per area) can be converted to X_{CH_4} (ppb) via a factor M (in units of mass and per area and per ppb).