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PIONEER PROJECTS

SOURCE ATTRIBUTION OF METHANE USING SATELLITE OBSERVATIONS, ISOTOPIC MEASUREMENTS AND GEOS-CHEM SIMULATIONS

CONTRACT - BR/165/PI/SACH4

FINAL REPORT

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SUMMARY

Context

Methane (CH_4) is the second most significant anthropogenic greenhouse gas in the Earth's atmosphere. There is now a widespread scientific consensus on the profound influence of human activity on the global climatic system, particularly through increased emissions since the pre-industrial era of greenhouse gases like CO_2 and CH_4. Although CH_4 is roughly 200 times less abundant in the atmosphere than CO_2, it is a more potent greenhouse gas. The impact of CH_4 on climate change is more than 86 times greater than that of CO_2 over a 20-year period. Given its relatively short lifetime of ~9 years, it is now recognized that one of the most efficient methods to mitigate warming due to greenhouse gases on decadal time frames is to cut CH_4 emissions.

Objectives

Although its main sources and sinks have been identified, many uncertainties remain regarding the balance between those sources and sinks. In addition, CH_4 is a challenging atmospheric component to study as its non-monotonous changes in the last decades and its interannual variability remain not fully understood. The ultimate aim of this BRAIN pioneering project is to reduce these uncertainties and increase our knowledge on how the different sources and sinks influence the atmospheric abundance of CH_4, using global distribution information from IASI, and GEOS-Chem model simulations to compare with IASI. A further objective is to set up approaches for retrieving heavy methane isotopologues ($^{13}CH_4$ and CH_3D) from ground-based FTIR spectra, such as to produce time series for comparison with main methane.

Conclusions

IASI global observations have been produced for 4 years using the v4 algorithm with an additional one year (2012) using ECMWF temperature profiles (v4TECMWF). Supporting GEOS-Chem simulations were performed for 2005-2014. Comparison between v4 IASI and v11-01 GEOS-Chem showed overall, higher IASI CH_4 concentrations than GEOS-Chem over the tropics and high-latitudes and lower IASI CH_4 values at mid-latitudes. Global mean differences and standard deviations of the difference are within the estimated retrieval uncertainty of IASI of 3.73%. During several months, particularly strong biases appeared near the Polar Regions, which, given that no strong methane sources are present near the South Pole, indicates that IASI v4 data needs to be handled with care in these regions. The comparison between v4TECMWF IASI and v9-02 GEOS-Chem showed more stable results, with a significant decrease in the bias over the mid latitude regions. Only at the South Pole, a systematic bias over the entire latitude band seams to appear, however less dramatic as with the first comparison. Unfortunately, the comparison between IASI and GEOS-Chem failed thus far to reach conclusions regarding the identification of the sources/sinks at play (e.g., wetlands).

Definition of heavy methane retrieval strategies from ground-based infrared spectra has been successful for both species. Multiyear time series have been produced and analysed. They indicate an acceleration of the CH_4 rise after 2012, and if the same feature is observed for $^{13}CH_4$, it is weaker, and such a lower ratio is notably consistent with the boom in the production of shale gas (a source depleted in ^{13}C).

Keywords

Climate change and mitigation, methane, remote-sensing, greenhouse gas, atmospheric budget, heavy methane isotopologues.

SAMENVATTING

Context

Methaan (CH_4) is het op één na belangrijkste antropogeen broeikasgas in de atmosfeer. De wetenschappelijke consensus stelt duidelijk dat de mens bijdraagt aan de opwarming van de aarde door de toenemende uitstoot van broeikasgassen zoals CO_2 en CH_4 sinds het pre-industriële tijdperk. Alhoewel de concentratie aan methaan in de atmosfeer ongeveer 200 maal lager is dan deze van CO_2, wordt dit gecompenseerd door het feit dat methaan een sterker broeikasgas is. Over een periode van 20 jaar, is de impact van CH_4 op de klimaatverandering meer dan 86 sterker dan CO_2. Door zijn relatief korte levensduur (~9 jaar), is de vermindering van methaanemissies dan ook één van de meest efficiënte methodes om de klimaatverandering de komende decennia in te perken.

Doelstellingen

Het bekomen van globale informatie over methaan door het de analyse van IASI spectra en de resultaten hiervan te vergelijken met het GEOS-Chem model. Het opstellen van strategieën om zware methaan isotopologen ($^{13}CH_4$ and CH_3D) te bekomen uit spectra gemeten door grondstation FTIR instrumenten en de dus bekomen metingen doorheen de tijd te vergelijken met deze van methaan.

Besluiten

Globale IASI observaties van methaan, over een periode van 4 jaar, zijn geproduceerd met versie 4 van het algoritme, met een bijkomend jaar (2012) met behulp van ECMWF temperatuurprofielen (v4ECMWF). GEOS-Chem simulaties werden uitgevoerd voor de periode tussen 2005 en 2014. Vergelijkingen tussen v4 IASI en v11-2 GEOS-Chem tonen in het algemeen ietwat hogere IASI concentraties boven de tropen en lagere waarden boven de middenlatitudes. De gemiddelde globale verschillen en standaard deviatie van de verschillen blijven binnen de vooropgestelde IASI onzekerheid van 3.73%. Gedurende enkele maanden werden evenwel wel sterke verschillen bemerkt aan de polen die, vermits er geen uitgesproken methaanbronnen zijn in het Zuidpoolgebied, aangeven dat het gebruik van deze data in deze gebieden met de nodige omzichtigheid moet gebeuren. De vergelijking tussen v4TECMWF IASI en v9-02 GEOS-Chem toonden stabielere resultaten met een significante daling van de systematische bias over de middenlatitudes. Enkel metingen nabij de Zuidpool toonden nog een sterke maar evenwel minder uitgesproken verschil. De vergelijkingen tussen IASI en GEOS-Chem slaagden er evenwel niet in om conclusies te bekomen over identificatie van bronnen/putten (bvb moeraslanden).

Het ontwikkelen van een strategie om zware methaanisotoopconcentraties te bekomen uit grondstation FTIR spectra werd succesvol afgerond voor beide moleculen. Tijdsseries die meerdere Jaren omvatten werden geproduceerd en geanalyseerd. Zij wijzen op een versnelde toename van de CH_4 concentraties na 2012, terwijl dit veel minder opgemerkt wordt in de $^{13}CH_4$ concentraties. Een verminderende verhouding is in overeenstemming met de sterke expansie van de schaliegas productie (wiens ^{13}C inhoud lager is in vergelijking met klassieke bronnen)

Trefwoorden

Klimaatverandering en mitigatie, methaan, atmosferisch budget, afstandsbepaling van broeikasgassen.

RESUME

Contexte

Le méthane (CH_4) est le deuxième plus puissant gaz à effet de serre anthropique dans l'atmosphère terrestre. Il y a maintenant un large consensus sur l'influence majeure des activités humaines sur le système climatique global, particulièrement au travers d'émissions croissantes, depuis le début de l'ère industrielle, de gaz à effet de serre comme le dioxyde de carbone (CO_2) et le méthane. Même si CH_4 est près de 200 fois moins abondant que CO_2, il s'agit d'un gaz à effet de serre 86 fois plus puissant que ce dernier sur une période de 20 ans. Mais étant donné sa durée de vie relativement courte d'approximativement 9 ans, il est démontré que réduire ses émissions constituerait une des méthodes les plus efficaces d'atténuation du réchauffement climatique à l'horizon décennal.

Objectifs

Même si les sources et puits principaux du méthane sont aujourd'hui bien identifiés, beaucoup d'incertitudes subsistent quant à leurs intensités respectives. De plus, CH_4 est un constituant atmosphérique compliqué à étudier, ayant présenté ces dernières décennies une vitesse d'accumulation et une variabilité interannuelle très changeantes et encore peu comprises. Le but ultime de ce projet BRAIN est de réduire ces incertitudes et d'augmenter nos connaissances sur les facteurs influençant l'abondance atmosphérique de CH_4, en utilisant des distributions globales déduites de mesures satellites IASI, et de les comparer à des simulations effectuées avec le modèle GEOS-Chem. Un objectif supplémentaire est de développer des stratégies de restitution des variétés isotopiques lourdes de CH_4 ($^{13}CH_4$ et CH_3D) au départ d'observations solaires infrarouges à haute résolution enregistrées depuis le sol, de façon à comparer leurs évolutions à celle du méthane le plus abondant.

Conclusions

Des observations globales de IASI ont été produites pour quatre années avec la version 4 de l'algorithme ASIMUT. Des données supplémentaires ont aussi été générées pour 2012 en utilisant des profils de température ECMWF (v4TECMWF). Des simulations GEOS-Chem ont été réalisées pour la période 2005-2014. Les comparaisons entre IASI (v4) et GEOS-Chem (v11-01) ont généralement montré des concentrations IASI plus élevées que les simulations pour les tropiques et les hautes latitudes, et des mesures plus basses que GEOS-Chem aux latitudes moyennes. Cependant, les différences moyennes globales restent en deçà de l'incertitude de 3.7% caractérisant les produits IASI. Au cours de quelques mois, des biais plus importants ont été mis en évidence pour les régions polaires. Mais considérant l'absence de fortes sources de méthane dans ces régions, il y a lieu de considérer ces observations avec réserve. Les comparaisons impliquant IASI (v4TECMWF) et GEOS-Chem (v9-02) ont révélé des résultats plus stables, avec un biais significativement plus bas pour les latitudes moyennes. Il n'y a que pour le pôle sud qu'un biais plus important semble se manifester, sans être toutefois aussi important que pour les comparaisons déjà évoquées. A l'heure qu'il est, nous n'avons malheureusement pas encore pu tirer des enseignements significatifs de ces comparaisons, permettant d'identifier des sources ou puits à l'œuvre (p. ex. pour les zones humides).

La définition de stratégies de restitution pour les variétés isotopiques lourdes du méthane au départ de spectres enregistrés au sol a été fructueuse pour les deux espèces visées. Des séries temporelles multi-décennales ont été produites et analysées. Elles démontrent une accélération de l'accumulation de $^{13}CH_4$ après 2012. Cependant, cette dernière est moins marquée que pour le méthane principal, ce qui est compatible avec le boom dans la production de gaz de shale (ou de schiste), une source appauvrie en ^{13}C.

Mots-clés

Changement climatique et atténuation, méthane, télémesures, gaz à effet de serre, bilan atmosphérique, méthane lourd.

1. INTRODUCTION

Methane is the second most abundant anthropogenic greenhouse gas in the atmosphere after CO2. Due to its average atmospheric lifetime which is ten times shorter than that of CO2 (roughly 10 vs 100 years), reducing CH4 emissions has a stronger more immediate impact on the Earth's radiation balance compared to reductions in CO2. It thus makes sense to pay particular attention to CH4. Unfortunately its sources and sinks (and particularly the evolution of the source and sink fluxes over time) are far less understood. CO2 over the last decades has seen a steady rise with a superimposed seasonal cycle in regions with significant vegetation uptake. Methane concentration in the atmosphere on the other hand showed a steady rise until its increase leveled off in 2000, only to start rising again in 2007, even picking up the pace round about 2014. In order to effectively reduce methane concentrations, we need to figure out this conundrum and while many hypotheses have been postulated (fracking, decrease in OH levels and biomass burning emissions, increased wetland emissions due to rising temperatures…), we are still uncertain as to which (or combination thereof) holds the key answer. One important component of determining source attribution is looking at companion species (species that are simultaneously emitted by certain source types), particularly isotopes. In this work we therefore strived to come up with new strategies to retrieve heavy methane related isotopologues so as to deliver long-term data series to the scientific community. A second goal was to produce satellite observations from IASI and from the GEOS-Chem Chemistry Transport Model, and to intercompare them so as to identify the primary source regions of methane, with particular attention to the wetland emission regions.

2. METHODOLOGY AND RESULTS

The current section is organized with respect to the tasks depicted in the SACH4 contract.

Task 1.1. CH_4 satellite data

Task 1.1.1. Processing IASI CH_4 profiles

At the initial stage of the project it was agreed to compare one year of data in order to test the BIRA-IASB IASI CH_4 data quality. After initial tests with 2011 data which showed promising results, the dataset was eventually expanded to cover the 2011-2014 period. This dataset has been described in detail in De Wachter et al (2017) and has been validated with various reference data sets. Unfortunately the IASI retrieved operational L2 temperature profiles do not yield a consistent (over time) dataset, switching to different versions as the instrument matured. This effectively meant that the older IASI data relied on out-of-date temperature retrievals and further prolonging the timeseries (in the past and future) resulted in strong bias jumps which coincided with times where the L2 operational IASI Temperature algorithm versions got updated. This effectively restricted the dataset to four years. Given that the goal was to look into sources but also eventually in the evolution thereof this was not an optimal situation. Therefore a reanalysis was performed using the same CH_4 retrieval algorithm but now using ECMWF temperature profiles (example plots are shown in figure 1), starting with the year 2012. This year coincided with a –initially– planned GEOS-Chem update from 9-02 to v11 to be performed by Whitney Bader (ULiege). This could potentially overcome the operational L2 temperature retrieval issues, effectively leaving room to expand the dataset beyond the 4 year timeframe. As with the original dataset, the results look promising with the typical features when looking at a global representation of the data such as the signature Hemispherical gradient and strong signals in Africa and South-America during the local biomass burning season. Areas with missing data correspond to areas which were identified as cloudy or corresponding to pixels where the retrieval did not converge to a result.

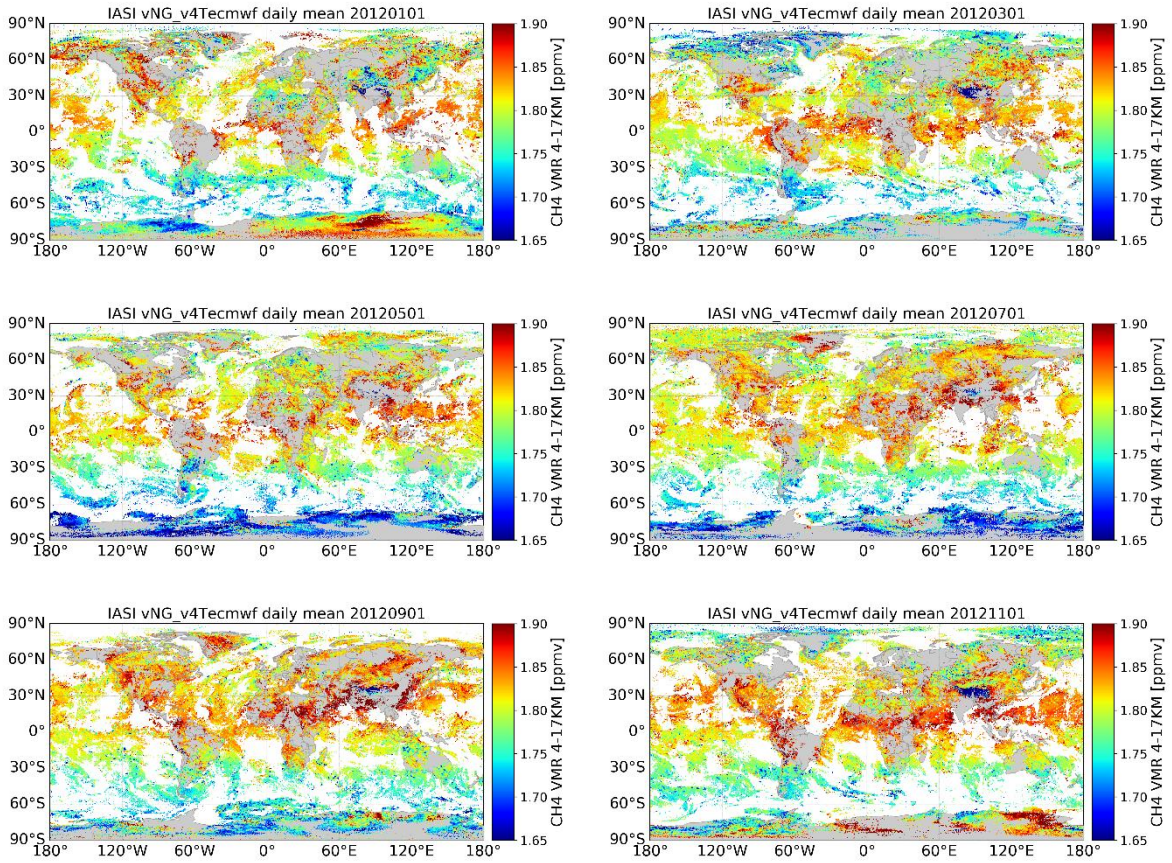

Figure 1: The v4ECMWF IASI CH4 daily mean 4-17 km partial column for the 1st day of the month (January, March, May, July, September and November 2012)

Task 1.1.2. Delivery IASI CH_4 profiles in suitable data-format

The IASI CH_4 data are kept in the original format, i.e. daytime and night time retrievals delivered in 2 separate Hierarchical Data Format (HDF) 5-files containing the retrieved CH_4 profile + error, a priori CH_4 profile, the averaging kernel, altitude and pressure grid and additional metadata. To account for the different temporal and spatial domains between the IASI observations and the GEOS-Chem model data, the model data are regridded onto the IASI data grid, partial columns and daily means are calculated (see Task 1.3.1).

Task 1.2. GEOS-Chem tagged simulations

The methane simulations used in the SACH4 project have been produced with the 3D-Chemical Transport Model (CTM) GEOS-Chem in operation at ULiège, using a 2°x2.5° horizontal resolution. CTMs require meteorological fields in order to represent vertical and horizontal transport and to prescribe temperature and pressure in each grid box. GEOS_5 meteorological fields from NASA/GMAO have been available until the end of March 2013, and a new replacement product, GEOS_FP, has been made available to perform simulations after that date. Since newer versions of GEOS-Chem allows using MERRA and MERRA 2 meteorological fields available for the last four decades, we envisaged to perform such new simulations in order to consistently conduct them over the time period addressed by the project. However, it turned out that in several successive newer versions of GEOS-Chem, the tagged methane simulations by categories was not implemented anymore, waiting for new tracers to be added and for a significant architecture update to make it

compatible with the new emission module HEMCO (Harvard EMission COmponent). A multiyear simulation using the v11-01 of GEOS-Chem and the same initial state (see Task 1.2.2.) for the total methane fields than for v9-02 was performed to support direct intercomparison between IASI and GEOS-Chem. In order to dispose of tagged information, we had to perform and validate a complete v9-02 simulations, and to achieve the GEOS_5 versus GEOS-FP consistency checks, as initially planned in the project proposal.

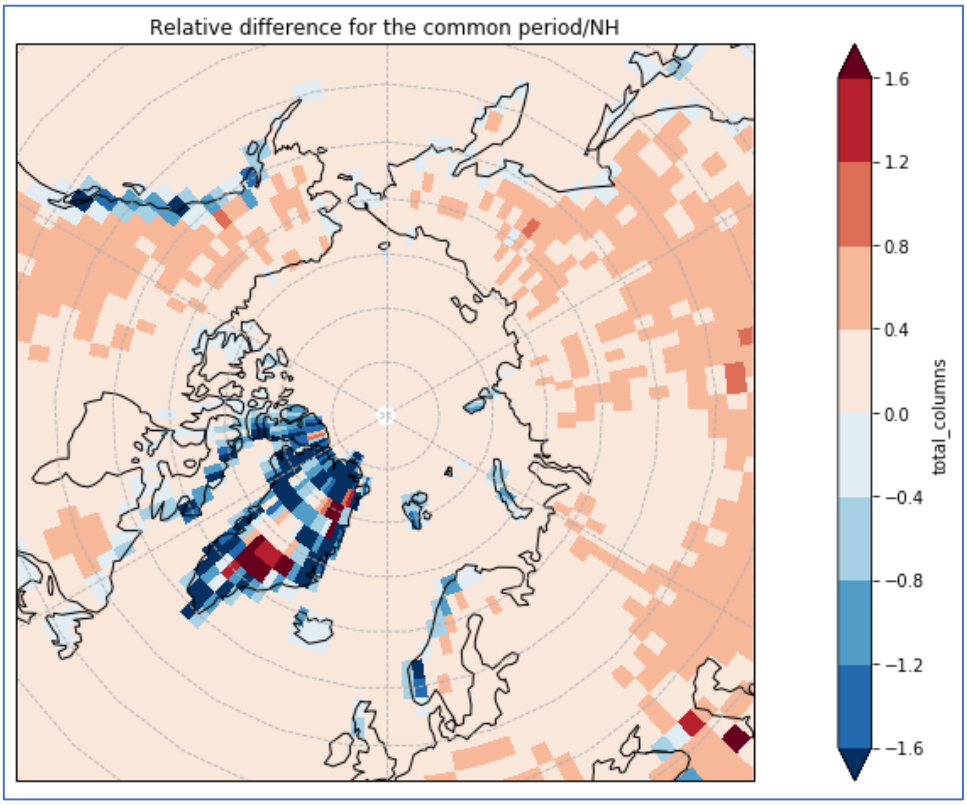

Figure 2: *Mean relative CH_4 column differences (FP/5) for Northern Hemisphere and June 2012 – May 2013.*

Thanks to an extended overlap period between these two products, we performed two parallel GEOS-Chem simulations for 2012/06 – 2013/05 and used the corresponding data to investigate possible methane column bias between the two runs. It is worth noting that these runs have been conducted with all other parameters being identical, in particular regarding the emission magnitudes and distributions, prescribed hydroxyl radical fields (main methane sink) and kinetic parameters. More information on the various parameters can be found in Bader et al. (2017).

Yearly, seasonal and monthly relative fractional differences have been computed as (FP - GEOS) / ((FP + GEOS)/2 * 100, accessing the native GEOS-Chem output files with in house Python tools. This quantitative investigation has indicated smooth patterns for the methane column differences, which are typically limited to ± 0.8% for most time periods regions and seasons. For some part of the world however, we determined larger, more systematic and inhomogeneous differences. That is true for the Arctic, Greenland and some high altitude regions. It is hard to disentangle the causes for these specific cases, but they can result from temperature differences, especially for cold regions, from contrasted wind fields having a significant impact on transport, in particular above mountainous terrains, or from a combination of these factors. Figures 2 and 3 show representative distributions of the methane column relative differences.

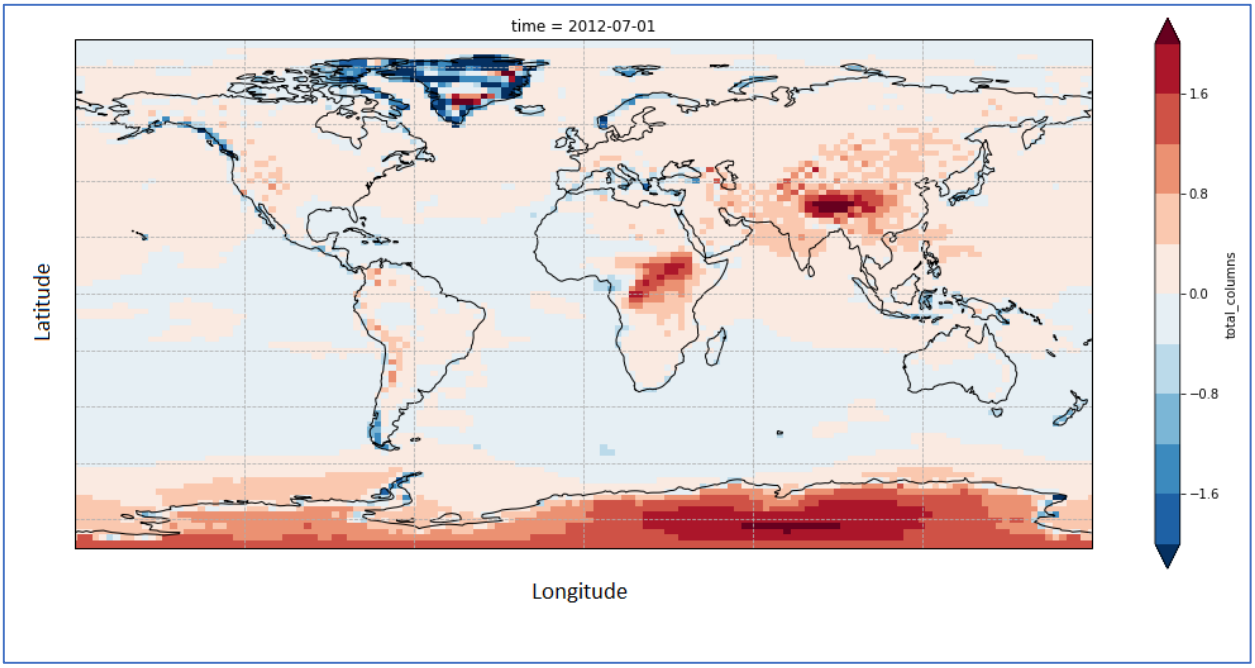

Figure 3: Monthly relative differences (FP/5) for July 2012

Task 1.2.1. GEOS-Chem model spin up

In order to generate a proper initial state to launch the GEOS-Chem simulations, it is important to perform spin-up simulations, successively looping over a given year. This way, the tracer concentrations (for the various methane tagged categories) will be progressively populated. We performed this operation using the year 2004. The total number of loops was determined by checking the consistency between the total/sum of the methane tagged tracers and total methane. Convergence and consistency were reached after 70 loops.

Task 1.2.2. GEOS-Chem model simulations with GEOS_5 and GEOS_FP

The relevant SACH4 simulations were performed with GEOS_5 meteorological fields for the 2005-2013/05 time period while GEOS-FP driven runs covered the years from 2012 onwards. The global simulations available for the common 1-year time period (from mid-2012 until mid-2013) were used in Task 1.2.1. The simulations have been made available to the coordinator using hdf archives. Another multiyear simulation using MERRA-2 and v11-01 of GEOS-Chem was also produced to support the direct comparisons planned in Task 1.3.

Task 1.3. Global comparison study between satellite and model data

GEOS-Chem v11-01 HDF-4 data files produced at a 3-h time frequency were compared to the v4 IASI retrieval product. IASI pixels were selected within 1.5 hours of the GEOS-Chem model data, within a 1°-1.25° latitude-longitude box around the GEOS-Chem latitude-longitude centre point. The GEOS-Chem CH_4 mixing ratio profiles were regridded to the IASI pressure grid. Total columns and partial columns between 4 and 17 km were calculated. The 4-17 km partial column corresponds to the vertical range of good sensitivity of the satellite (with a Degree of Freedom for Signal (DOFS)=1).

The GEOS-Chem CH4 4-17 km partial columns (see Task 1.1.2) were smoothed with the IASI averaging kernel (AK) for each corresponding IASI pixel to account for the different vertical resolution between IASI and the higher resolved model data. An example of the IASI 4-17 km partial column averaging kernel (AK) as a function of latitude for the 1st of April and the 1st of October 2011 is given in Figure 4. The AK describes the sensitivity of the measurement to the CH_4 vertical distribution. From the figures we can see that we have good sensitivity from about 600 to 80 hPa (~4-17 km) with a reduction of sensitivity to 500-200 hPa (~5.5-12 km) at high latitudes (>70°), depending on season.

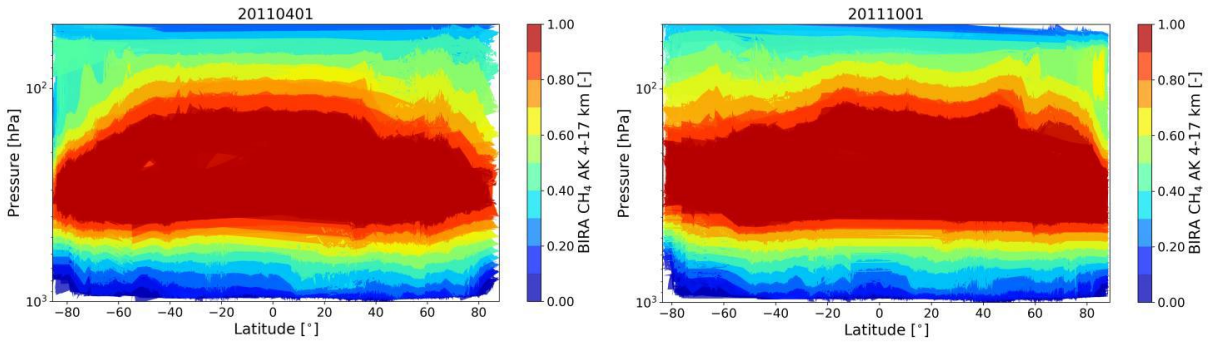

Figure 4 *: IASI 4-17 km averaging kernel as a function of latitude and pressure for the 1st of April (left) and the 1st of October (right) 2011.*

The v11-02 GEOS-Chem smoothed 4-17 km partial columns are shown in Figure 5. We find the highest CH_4 concentrations in the tropics throughout the year and the same latitudinal gradient as seen in Figure 1 and discussed in Task 1.1.1. As observed with IASI, the CH_4 concentrations are higher in July than in January in the NH.

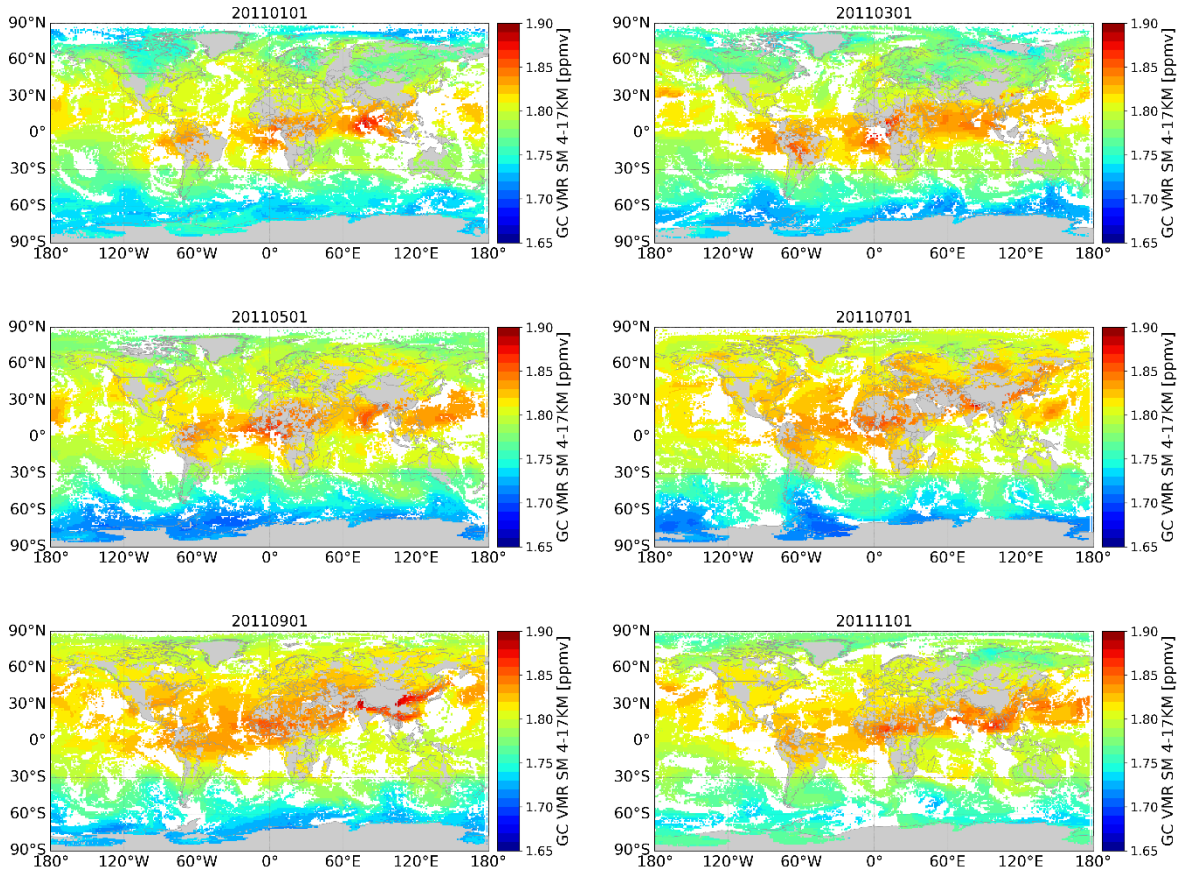

Figure 5 *: The GEOS-Chem v11-01 daily mean 4-17 km partial column smoothed with the IASI averaging kernel for the 1st day of the month for January, March, May, July, September and November 2011.*

Comparisons on a global scale allow us to identify regional discrepancies between the IASI observations and the GEOS-Chem model. These differences could suggest strong unidentified emissions at a local scale or could be attributed to an artefact in the IASI CH_4 retrievals. Relative

differences (observation-model) are given in Figure 6. Overall, higher IASI CH_4 concentrations than GEOS-Chem are found over the tropics and high-latitudes and lower IASI CH_4 values at mid-latitudes. Global mean differences and standard deviations of the difference are within the estimated retrieval uncertainty of IASI of 3.73% (De Wachter et al., 2017). We see a high discrepancy between IASI and GEOS-Chem for the Antarctic in September-November 2011. Since there are no known large sources of CH_4 in the Antarctic, this is probably a IASI CH_4 retrieval artefact and needs to be further investigated. In January 2011, a large positive difference is found between IASI and GEOS-Chem in the Norther high-latitudes; in Canada (Hudson Bay) and East-Russia. In May we see again a strong bias near the Arctic.

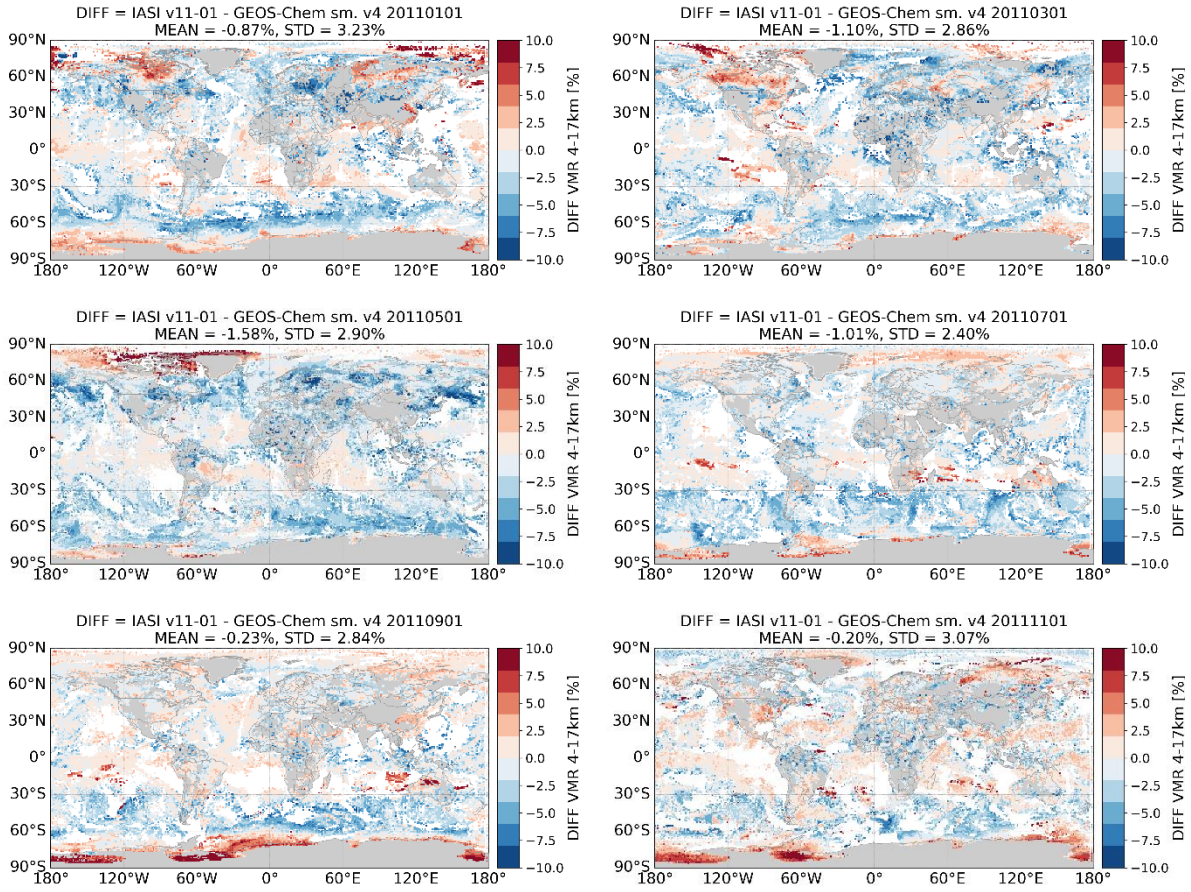

Figure 6 : Relative differences between IASI v4 and smoothed GEOS-Chem v11-02 4-17 km partial columns for the 1st day of the month for January, March, May, July, September and November 2011. Mean relative differences and standard deviations of the difference are given in the title.

Zonal mean differences between 4-17 km partial column mixing ratios for IASI and smoothed GEOS-Chem are presented in Figure 7. Largest differences between the satellite data and model are found in the SH for latitudes > 30°S. In the tropics IASI and GEOS-Chem show similar values. IASI also observes a reduction in CH_4 in the Northern mid-latitudes (around 50-60°N) which is not captured by the model.

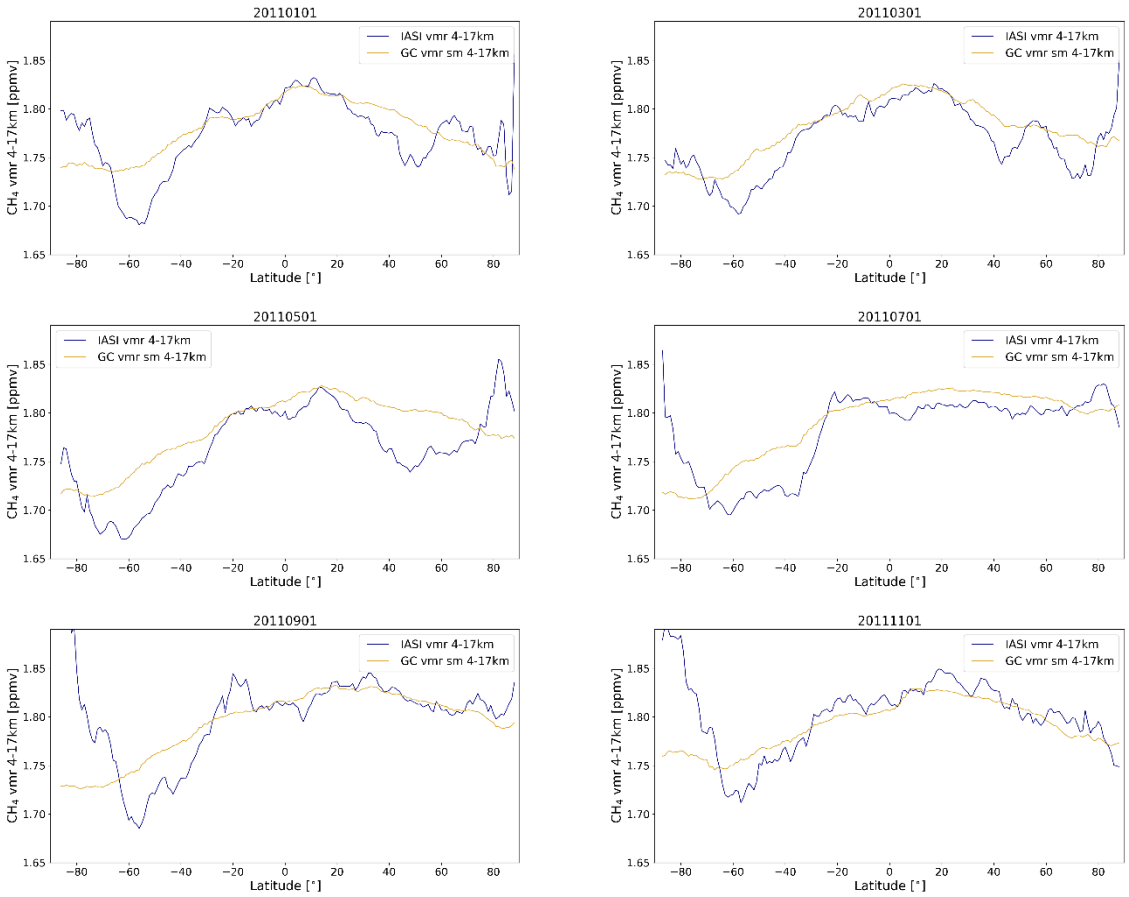

Figure 7 : Comparison zonal mean difference between 4-17km CH4 partial columns of IASI v4 and smoothed GEOS-Chem v11-01 for the 1st day of the month for March, May, July and September 2011

However as already mentioned, the v4 IASI retrieval product showed limitations in expanding its dataset while the v11-02 GEOS-Chem model data lacked tracers to effectively identify different source components in the model. Therefore, as mentioned in Task 1.1.1., a new IASI retrieval product was generated. Similarly an update of GEOS-Chem towards v12 was planned. Unfortunately due to the prolonged illness and eventual departure of the principal GEOS-Chem investigator this update never materialized and it was thus decided to compare the new IASI data with a v9-02 run which still harboured tracer information. It should be noted that while all the IASI updated data was generated by Dr Evelyn De Wachter, and all comparison software was in place and tested on the above described v4 IASI and V11-02 GEOS-Chem run, she too left right after completion of the 2012 v4TECMWF IASI dataset which still needs to be validated.

After the departure of the 2 principal investigators on this project an extension was requested and granted to effectively be able to finalize the IASI GEOS-Chem comparison. The successful completion of this aspect of the project, within a reasonable timeframe, hinged on an issue-free GEOS-Chem IASI comparison so we could focus on the tracer aspect of the study. This, unfortunately, was not the case. While we did manage to resolve these issues and finalize the comparison between the two products (discussed further below), insufficient time was left and therefore we have no other choice but to accept that the source attribution aspect of the study did not materialize the results we had hoped for.

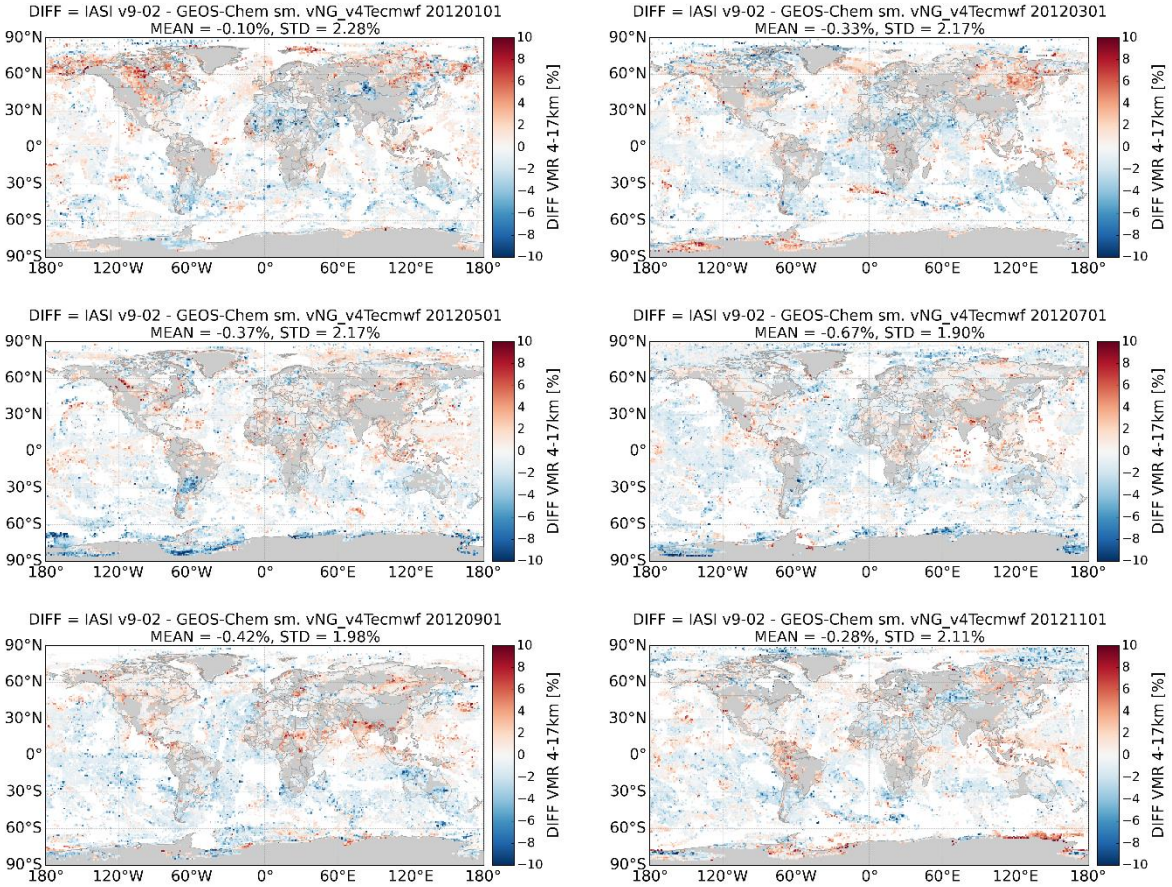

Figure 8 : Relative differences between IASI v4TECMWF and smoothed GEOS-Chem v9-02 4-17 km partial columns for the 1st day of the month for January, March, May, July, September and November 2012. Mean relative differences and standard deviations of the difference are given in the title.

Figure 8 above shows the difference between IASI v4ECMWF and GEOS-Chem 9-02 for the year 2012. Even though one has to take into account that the comparisons are between different years and model and IASI versions, if we compare the above figure to figure 6, we immediately see that the differences are overall less pronounced. The extremely high values near the South Pole (while still present as a bias) are far less pronounced. This difference is even more visible if we compare the results in figure 7 with those of figure 9 (below) which show the average latitudinal bias. While Figure 7 still shows marked patterns with mid latitudinal IASI values that are far lower than the model, any systematic latitudinal bias has almost disappeared in figure 9. Only near the South Pole do we still see, most of the time, a more pronounced bias.

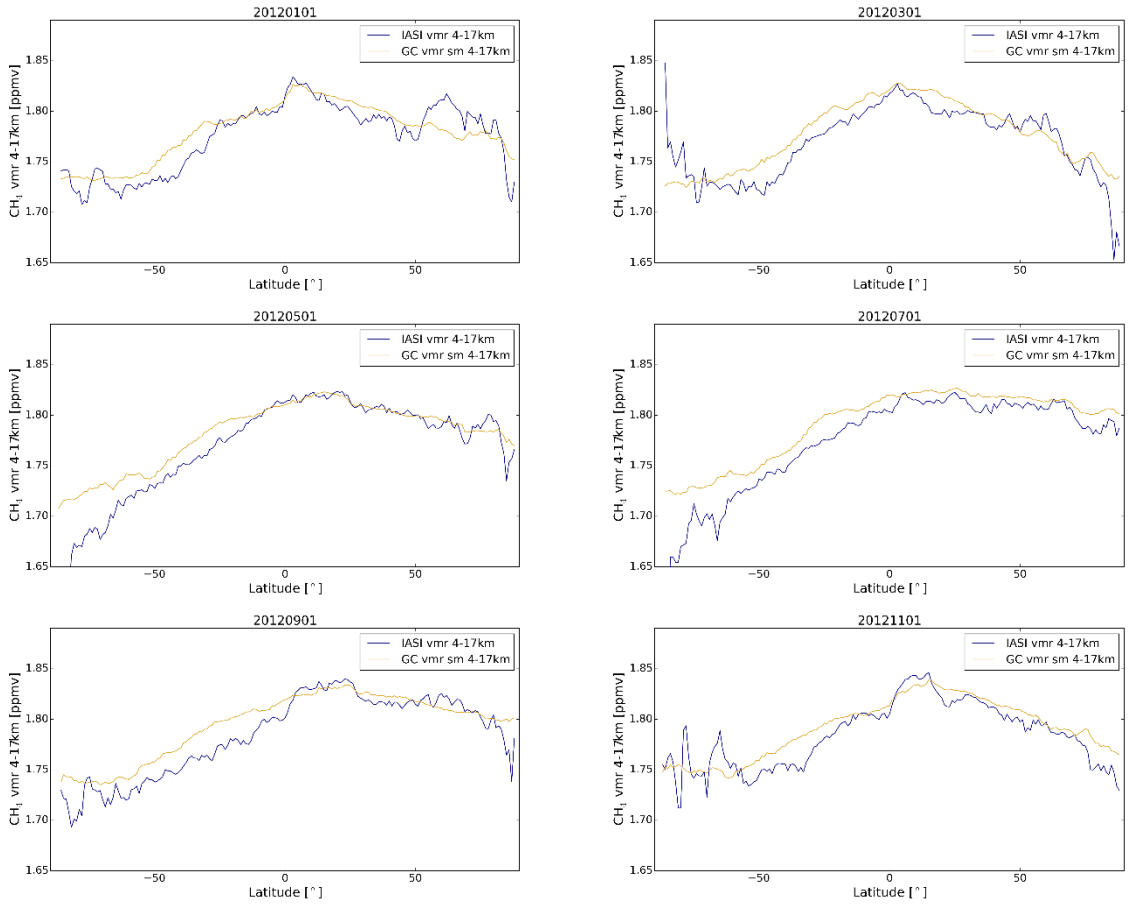

Figure 9 : Comparison zonal mean difference between 4-17km CH_4 partial columns of IASI v4TECMWF and smoothed GEOS-Chem v9-02 for the 1st day of the month for March, May, July and September 2012

This indicated that the employment of temperature profile data from ECMWF is not only a viable option to facilitate the expansion of the IASI CH_4 time series, but also that the retrieval itself seems to benefit greatly in terms of overall stability of the product. Further validation with independent sources such as ground-based FTIR measurement and in situ profile measurements need of course to confirm this, but the results in this study are more than encouraging.

Task 2.1. Retrieval of CH_3D

Task 2.1.1. Development and optimization of retrieval strategy

A retrieval strategy has been developed for CH_3D, using high resolution ground-based FTIR spectra collected at a suite of NDACC sites (Network for the Detection of Atmospheric Composition Change), including Jungfraujoch, Maido, Porto Velho and Lauder. This requires identifying the best combination of spectral windows, spectroscopic linelist, a priori mixing ratio profile, retrieval constraints, … As usual, the strategy has been developed and tuned such as to minimize the spectral residuals and maximize the vertical sensitivity range. Moreover, we took great care to define a network-wide approach, applicable to sites with very different atmospheric conditions in terms of humidity and altitude. The final settings include two windows (near 2951 and 3071 cm^{-1}) and use a Tikhonov regularisation. For Jungfraujoch, mean Degree Of Freedom for Signal (DOFS) is 1.9 (and the 10^{th}- 90^{th} percentile range is [1.5 – 2.3]), allowing deriving independent information in the troposphere and the stratosphere. Mean random and systematic relative uncertainties on the total columns derived from NDACC-compliant full error budgets typically amount to 4.4% [2.7 – 7.9%] and

6.5% [6 – 7%]. Figure 10 shows typical mixing ratio averaging kernels for Jungfraujoch, indicating a good vertical sensitivity from the ground up to about 35 km. Comparable performances are obtained for the other ground-based sites.

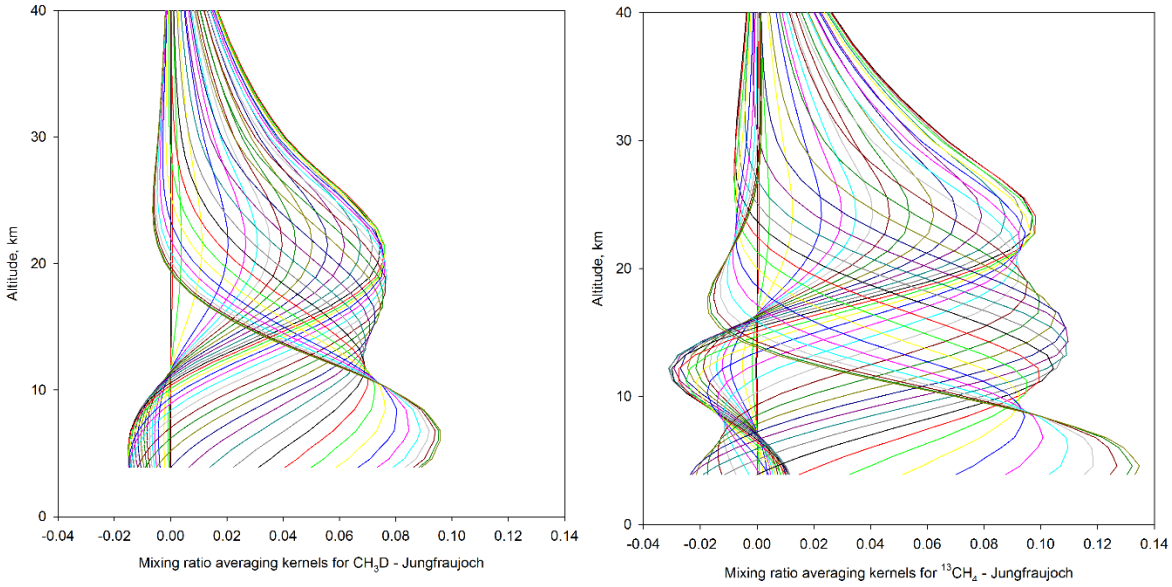

Figure 10: Typical mixing ratio averaging kernels for CH_3D (left) and $^{13}CH_4$ at Jungfraujoch.

Task 2.1.2. Quality control

Following the production of the complete time series (see next task), we set up a QA/QC control procedure in order to automatically flag poor quality measurements. This procedure makes use of a series of objective indicators for which thresholds are defined, based on statistics established considering the whole available data set for each site. The indicators include the RMS of the fits, the DOFS, the total random and systematic uncertainties. After application of this filtering procedure, 7700, 3340 and 4060 individual measurements have been kept for Jungfraujoch, Maido and Lauder, respectively.

Task 2.1.3. Time series production for CH_3D

All available FTIR spectra have been fitted consistently adopting the 2-window approach described above, leading to the production of multi-decadal FTIR time series for CH_3D. Following the approach described in section 2.4 of Barthlott et al. (2015), we further computed the total column dry-air mole fractions of CH_3D. Figure 11 shows the monthly mean time series derived for Jungfraujoch, Maido and Lauder. It is worth indicating that Jungfraujoch measurements derived from the homemade FTIR instrument in use between 1984 and 2008 are not included here because they are noisier (signal-to-noise ratio of the spectra are a bit low for such weak spectral absorptions). Also, data for Porto Velho are not shown because of a significant gap in data acquisition.

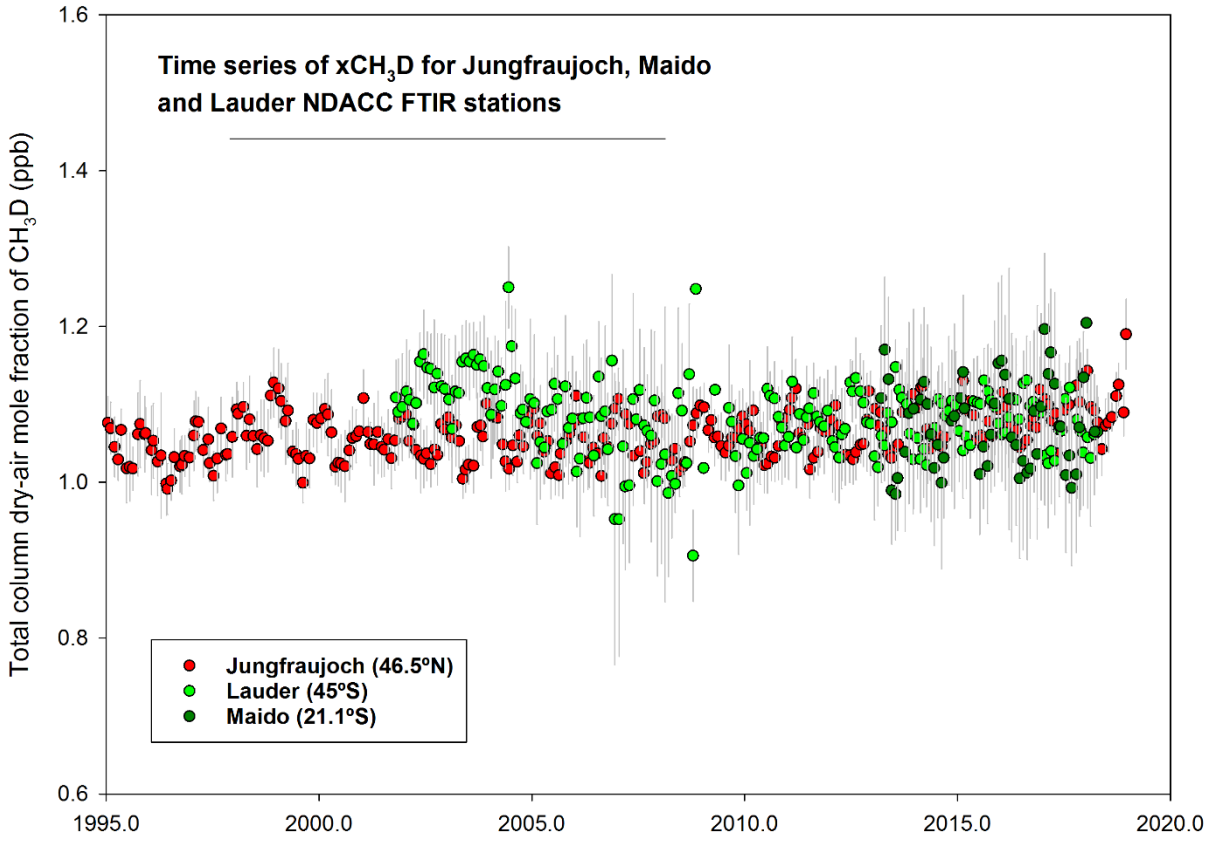

Figure 11: Time series of monthly mean total column dry-air mole fractions of CH_3D at three sites.

We note first that the mean abundances of CH_3D derived for the three sites are very close in both hemispheres, in the 1.06 – 1.08 ppb range. The second interesting characteristic of these data sets is that they indicate very little change of the CH_3D mole fraction with time, in contrast with the main methane isotopologue which, over the same time period, has been accumulating in the atmosphere (apart from the well-known plateau in 2000-2004). This means that the sources and sinks responsible for the emission and removal of CH_3D to the atmosphere should not have undergone important changes in their magnitude.

Task 2.2. Retrieval of $^{13}CH_4$

Task 2.2.1. Development and optimization of retrieval strategy

Similarly to CH_3D, systematic search for $^{13}CH_4$ lines has been performed, followed by individual evaluation of the associated information content. Several combinations of lines have then been tested using FTIR spectra for all available ground-based sites. A final 5-window approach has been validated for dry-to-humid sites, using $^{13}CH_4$ lines near 2760, 2765, 2817, 2859 and 2892 cm^{-1}. Here also, a Tikhonov regularization has been adopted. For Jungfraujoch, a mean DOFS of 2.6 is obtained [2.2 – 3.0], i.e., even more favorable than for the weaker absorptions of CH_3D. Mean random and systematic relative uncertainties on the total columns amount to 2.4 [1.8 – 3.3%] and 2.5% [2 – 3%], respectively. These figures also show a situation which is more favorable than for CH_3D. Figure 10 displays typical mixing ratio averaging kernels for Jungfraujoch, with obvious higher vertical resolution than for CH_3D.

Task 2.2.2. Quality control

Similarly to task 2.1.2., we have determined thresholds useful for filtering the $^{13}CH_4$ data sets. More than 13000 individual measurements have been kept for Jungfraujoch. In this case, data derived from the homemade solar observations were of sufficient quality to be considered in the next steps.

Task 2.2.3. Time series production for $^{13}CH_4$

As for CH_3D, a time series of total column dry-air mole fractions has been produced, considering all available observations recorded at the Jungfraujoch station with the homemade and Bruker FTIR instruments. By lack of time, the final validation of the complete time series for Maido has not been possible.

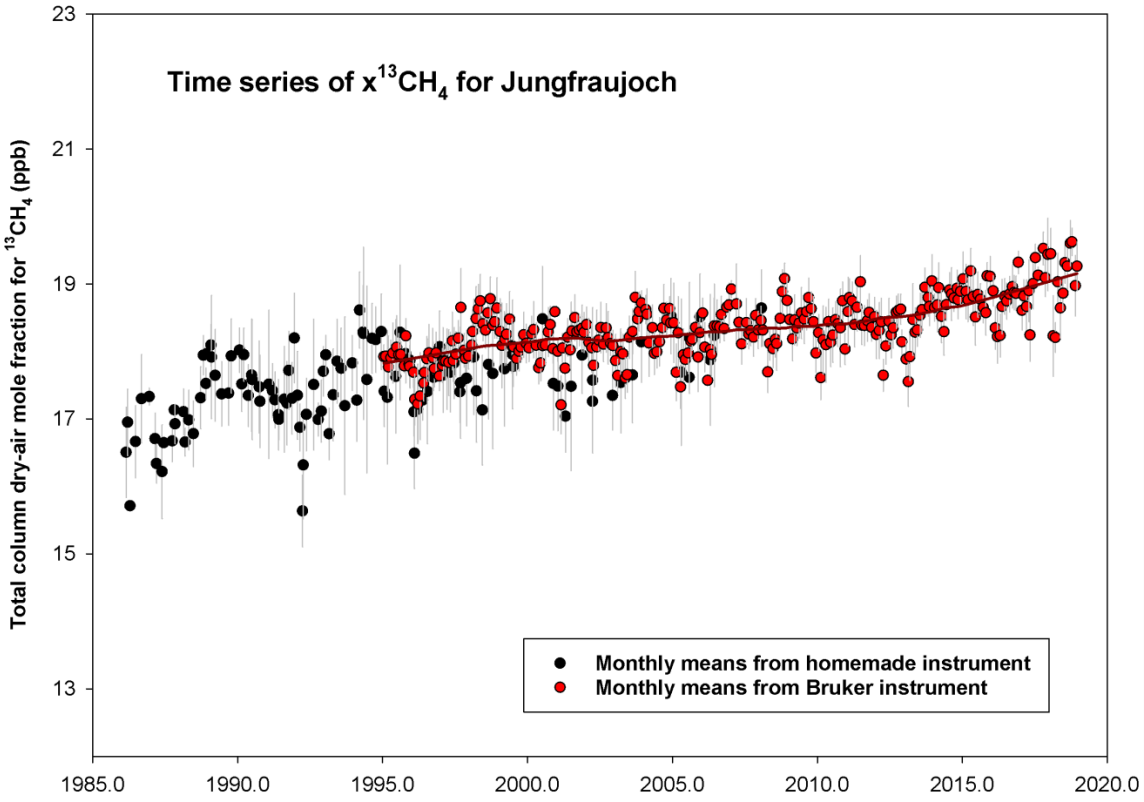

Figure 12: Time series of monthly mean total column dry-air mole fractions of $^{13}CH_4$ at Jungfraujoch.

Figure 12 shows the multi-decadal time series derived for the Jungfraujoch station. Unlike CH_3D, we observe here a buildup of $^{13}CH_4$ with time, showing only a slowing down around the early 2000s. More recently, the trend has been on the rise, exhibiting as for CH_4 (not shown) an acceleration after about 2012-2013. In order to quantify this acceleration, we computed $x^{13}CH_4$ and xCH_4 trends and associated uncertainties using a wild bootstrap resampling tool accounting for the inter correlations present in the geophysical data sets. The advantage of this method – when compared to standard bootstrap tools – is to provide more robust estimates of the uncertainties characterizing the rates of change. Table 1 provides the absolute and relative trend values for three post-plateau periods, namely 2005-2018, 2005-2011 and 2012-2018, all inclusive.

	Absolute trend (ppb/yr)	Relative trends (%/yr)
xCH_4		
2005-2018	6.4 ± 1.1	0.38 ± 0.06
2005-2011	3.6 ± 2.5	0.21 ± 0.15
2012-2018	12.2 ± 2.9	0.71 ± 0.17
$x^{13}CH_4$		
2005-2018	0.64 ± 0.10	0.35 ± 0.05
2005-2011	0.51 ± 0.23	0.28 ± 0.13
2012-2018	1.17 ± 0.27	0.64 ± 0.15
Uncertainty ranges at 2σ		

Table 1: Absolute and relative annual trends for CH_4 and $^{13}CH_4$ at Jungfraujoch.

A first important information is that all trends are positive and significant at the 2σ level of uncertainty. As anticipated when looking at Figure 12, the rates of change for $x^{13}CH_4$ have indeed been on the rise over the last years, with annual buildup going from 0.28 to 0.64%/yr. The accumulation of the main methane isotopologue shows the same behavior, with an even stronger and statistically significant jump from 0.21 to 0.71%/yr.

3. DISSEMINATION AND VALORISATION

Presentations at meetings:

Participation to the NDACC-Infrared Working Group meeting in May 2017. The oral presentation can be found at https://orbi.uliege.be/handle/2268/213114

De Wachter, E., W. Bader, E. Mahieu, B. Langerock, N. Kumps, A.-C. Vandaele, and M. De Mazière, SACH4, Source attribution of CH_4 using satellite observations, isotopic measurements and GEOS-Chem simulations, poster presentation at 14th International Workshop on Greenhouse Gas Measurements from Space (IWGGMS-14), University of Toronto, 8-10 May 2018, Toronto, ON.

Mahieu, E. et al., Update on the FTIR monitoring program at the Jungfraujoch station, oral presentation at the “GAW-CH Landesausschuss" Fall Meeting, 31/10/2018, Zuerich, Switzerland.

Close contacts have been established with PIs from the Infrared working group of the NDACC network to inform them about the investigations conducted, aiming at retrieving useful information from ground-based FTIR spectra for two heavy stable isotopologues of methane. This resulted in early participation of D. Smale from NIWA. A first tangible result is the availability of a complete and validated time series for CH_3D for the Lauder site.

Further contacts have been established with colleagues from Empa who recently started in situ (flasks) measurements of $^{13}CH_4$ and CH_3D. This opens the way for future intercomparison of in situ and remote-sensing data.

4. PERSPECTIVES

While the previous iteration of the IASI product featured some marked systematic differences with the GEOS-Chem model data, its latest iteration using ECMWF temperature profile data yielded far more stable results when compared to the model. While we could not embark on the source region analysis within the, already extended, timeframe of this study due to the departure of the two principal investigators in this project, these results certainly merit further investigation. Likewise a validation of these new IASI data with independent ground-based remote sensing and profile in situ measurements is certainly warranted. When these studies confirm the quality of the product, an expansion of the 2012 data will certainly be desirable. This study however already suggests that the global monitoring of methane using IASI has reached maturity. In the future, such a multiyear expanded data set produced with the ASIMUT retrieval software developed at BIRA-IASB certainly deserves to be exploited in inverse modeling studies, as was recently the case for GOSAT data.

Another important outcome of this project is the successful definition of a retrieval strategy for $^{13}CH_4$ and CH_3D, valid for dry-to-humid sites. This approach provides measurements characterized by good information content, with independent tropospheric and stratospheric pieces of information. This product also reaches a good level of precision, especially for $^{13}CH_4$, with typical random errors of 2.5%.

At a time where important questions remain regarding the respective contributions from various sources to the renewed rises of CH_4 and $^{13}CH_4$, either enriched or depleted in ^{13}C, from agriculture, from the oil and gas sector as well as the possible changes in the OH sink, the application of this approach at the NDACC network level will lead to the production of a useful near global data set. This ensemble will be very valuable to advance our understanding of the methane budget, to help in the definition of relevant measures and emission cuts for successful mitigation of climate change.

5. PUBLICATIONS

De Wachter, E., Kumps, N., Vandaele, A. C., Langerock, B., and De Mazière, M.: Retrieval and validation of MetOp/IASI methane, Atmos. Meas. Tech., 10, 4623–4638, https://doi.org/10.5194/amt-10-4623-2017, 2017

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De Wachter, E., Kumps, N., Vandaele, A. C., Langerock, B., and De Mazière, M.: Retrieval and validation of MetOp/IASI methane, Atmos. Meas. Tech., 10, 4623–4638, https://doi.org/10.5194/amt-10-4623-2017, 2017.