*R*eply to Dr. Gambacorta's TCD review

Our discussion paper (https://www.the-cryosphere-discuss.net/tc-2018-237/) analyses AIRS v.6 Arctic Barents and Kara seas (BKS) data. We are happy to respond to public comments by Dr. Antonia Gambacorta as this provides us with our first chance of a public discussion, which we believe will be helpful to the larger community. Below, we provide validation of retrieved CH_4 data for both AIRS and IASI for the study area, the BKS. Most of these data and figures are from several publications and from unpublished manuscripts, as well as in numerous oral and poster reports. For data prepared specifically for this Reply, no references are given.

Firstly, concerning the geophysical problem and the importance of Thermal Infrared (TIR) data for the Arctic.

The Barents and Kara Sea region is a shallow continental shelf with water depths between 0 and 400 m. Its floor has numerous proven deposits of oil and natural gas (which is ~90% methane). Also, there are many spots with proven emission from methane hydrates that are stable below 250 m depth that can be impacted by warming seawater leading to seabed methane emissions. The Kara Sea seabed and that of the southern region of the Barents sea are affected by submarine permafrost. Emissions from methane hydrates and permafrost are temperature-dependent. Methane bubbles ascend from the sea floor and dissolve into the water depending on size with larger bubbles rising higher. Due to the great depth direct bubble mediated transport of methane to the sea surface is not expected to be dominant. However, indirect transport by turbulent diffusion and/or thermal convection can drive the methane to the sea surface where it evades and can be detected by satellite. Note, such emissions will not be in a focused plume, but be as an area average enhancement. That said, given the size of satellite pixels, whether the molecules are in a focused plume or a diffuse haze makes no difference.

A number of recent papers have argued that diffusion (mixing) is negligible allowing efficient microbial oxidation of almost all methane. Our satellite data contradict this viewpoint. Underlying it are two incorrect assumptions – in fact, transport timescales are short because the BKS are very stormy and suffer from thermal convection, and because oxidation timescales outside of plumes are long compared to the time between storms. We have demonstrated a correlation between the oceanic mixed layer depth and anomaly of atmospheric methane [7,8]. Methane emission from BKS occurs mostly after the summer-time stable stratification breaks down [7], when thermal convection due to seasonal cooling is more effective.

On the other hand, the BKS is a gateway of warm Atlantic water for the Arctic Ocean. It is this impact by warm Atlantic currents that makes BKS climatically important and very interesting for year-round methane TIR satellite measurements. Measurements over the ice-free Barents Sea are possible even during the polar night. Passive (based on solar radiation) shortwave (SWIR) is ineffective, and active SWIR instruments are unavailable (a first lidar instrument MERLIN is still under construction and/or lab testing). Five TIR spectrometers are in orbit now and operational, but they have a reduced sensitivity for the lower troposphere. We maintain that "reduced sensitivity" does not necessarily mean negligible sensitivity. Analysis of AIRS v.6 methane variation for the lower troposphere (LT) 0-4 km altitude is a focus of our discussion paper. Other papers [1-8] consider similar data from another TIR spectrometer, IASI. As a matter of fact, wind speed in the lower 4 km altitude and, especially in the planetary boundary layer (PBL), is lower than above 4 km. Therefore, investigation of LT methane is more promising for locating sources than upper troposphere methane or total column methane.

There are two ways to proceed. We may accept a position of infeasibility of TIR satellite methane measurements in the Arctic and wait until MERLIN is launched and provides sufficient data - i.e., ~10 years from now, presuming it achieves orbit – not an insignificant risk given what happened to OCO-1. Such future possible data necessarily miss any historical data. Alternatively, we can retrieve as much information from current and previous (to 2003) TIR measurements as possible.

The most important points concerned by Dr. Gambacorta are as follows:

1. Filtering technique is wrong. Dr. Gambacorta believes that the degree of freedom (DOF, archived by NUCAPS and AIRS v6), as well as Averaging Kernels (AK, archived by AIRS v6, but not archived by NUCAPS/IASI) should be used to estimate the sensitivity of TIR to LT. Our position is that lapse rate (in our terms, Thermal Contrast or ThC) is a better filtering parameter for the Arctic, discussed below:

2. Representativeness of data for the BKS is questionable. The number of single retrievals per grid cell is sufficiently high and does not depend significantly on the season (see below).

3. Coastal effects are artifacts. IASI clearly demonstrates enhanced methane along some coasts in BKS (west of Novaya Zemlya and Greenland, around Spitsbergen, northern coast of Norway, around Franz-Josef Land). The effective spatial resolution of IASI is ~ 24 km in nadir and most of the fields of view with enhanced methane cover both land and sea. The retrieval technique is less accurate in these situations. See below for details and discussion. That said, our analysis is not focused on coastal regions, and our interpretations are drawn largely from offshore pixels.

4. Sensitivity to LT is negligible. See discussion below.

Summarizing these concerns, the discussion question may be formulated as: "**Are the anomaly patterns and the statistically significant trends in these anomaly patterns observed by AIRS and IASI over BKS real or are they instrumental/retrieval artifacts**?"

Before going into details on our approach, we note that the presence of statistically significant trends in methane argues against the retrieved methane concentrations being pure noise and artefacts. In addition, when we started the study, we noted that the anomalies were consistent with geology and oceanography, which we acknowledge does not demonstrate a causal relationship, yet is contrary to expectations for noise. These two observations were sufficient for us to put together a successful proposal to investigate in detail the potential for a causal relationship.

1. Filtering technique is wrong

We are confident that our filtering technique is fundamentally necessary and applicable. Plots in ***slide 1*** are from [1, 4]. All methane Nov.-Dec. lower troposphere (LT) data over the Arctic ocean to the N from 60° N were plotted versus the thermal contrast, ThC, defined as the difference between Sea Surface Temperature and Air Temperature at 600 hPa, measured by IASI (Fig. 1A). For ThC<10°C, the ensemble of data and the average curve drop to unrealistically low CH_4 (below the range of *in situ* NOAA data (mean±STD) for 5 Arctic sites and for the same time period is shown by a rectangle). Values lower than ~1800 ppb (see also maps on other slides) correspond to cold, thick sea ice, which has low ThC. Given the importance of thermal contrast to TIR retrievals, we propose that low lapse rates over cold surfaces reduces sensitivity to the LT, leaving the retrieval techniques to emphasize radiation from upper atmospheric levels, where concentrations are lower and thermal contrast is greater. The filtering technique used by us is a fundamental condition, we require that the data with ThC <10°C are excluded from consideration. The bottom graph (Fig. 1C) is for a different time period, land and sea; the same pattern is observed.

The reviewer suggested DOF as a filtering parameter. Standard QC recommends DOF>0.3 (X. Xiong, personal communication, 2016). Similar to graph 1A, LT CH_4 was plotted as a function of DOF [1]. Fig. 1B. Applying this criterion does not filter out unrealistically low LT CH_4 (below 1750-1800 ppb) characterized by a wide range of DOF with a maximum of 1.3. After averaging (circles with error bars), CH_4 for DOF 0.4-0.5 is just 10 ppb less than for DOF 1.1-1.3. We conclude that DOF is not the best parameter for filtering Arctic CH_4, while acknowledging that it may be useful for other, lower latitude, areas. It is noteworthy that a ThC filtering has been successfully used and validated for such a LT species such as NH_3 [9], the only difference is that [9] uses 1.5 km as the upper level for ThC calculation.

Slide 2 [4] presents two consecutive ascending granules of L2 AIRS v6 LT data with standard filtering (left) and after additional ThC filtering (right) in March, 2013. Missing points on the left map are filtered for DOF<0.3 and/or overcast cloudiness. Filtering due to insufficient thermal contrast removes more pixels from the map, e.g., all land points. Two retrievals over the Barents and Laptev Seas are highlighted by red ovals. The averaging kernels for these pixels are plotted in the middle row of figures; temperature profiles for them are on the bottom-left figure.

The effect of filtering in high Northern latitudes on a monthly basis in winter is shown in ***slide 3*** (3A and 3B) [1]. Note that much of the Barents Sea is ice-free in winter (3C). The Kara Sea is ice-covered in January, but has many cases of ThC >10°C, allowing reliable retrievals. Relatively warm open water surface in winter favors high ThC and, consequently, high sensitivity. See also maps below.

2. Representativeness of data

Detection of Ssmall variations of atmospheric CH_4 superimposed over high background concentrations (lower than 4-5%) requires either a high accuracy of individual measurement, or averaging of a suitably large number of measurements. Typical data scatter is ± 20-30 ppb, whereas typical Arctic CH_4 anomalies are in the range 20-50 ppb with 80-100 ppb as a maximum. Normally there are ~30 points per 0.5x 0.5° grid cell per month (***slide 7***). One-month temporal resolution is quite reliable even for 0.5x0.5° maps. A 5-day time span requires at least 1x1° gridding. Daily maps are hardly feasible with this instrument and this retrieval algorithm, which was why we chose to improve our SNR by monthly averaging.

3. Coastal areas

We have to recognize that this is the weakest point in our study. Retrieval NUCAPS problems for partial land/sea pixels makes coastal anomalies suspicious. The plots in ***slides*** **8-9** present single pixels and binned dependencies of retrieved CH_4 for the coastal atmosphere. We have no convincing explanations for CH_4 increases there. Most striking is that these spikes only appear in certain areas (Norway, West of Novaya Zemlya, West of Greenland) and do not appear in other areas (East of Novaya Zemlya, East of Greenland; also no effect in Kara sea, etc.), suggesting this is not a simple artifact (it may be a complicated artifact depending on unique atmospheric characteristics). Therefore, the coastal results admit a dual explanation. Very small spots of high methane emissions at sub-pixel scales along the coasts cannot be ruled out. A further study of these effects may include one-pixel version of NUCAPS, comparisons with *in-situ* data, etc., and is a topic for future research.

4. Does the DOF and/or ThC correlate with CH_4 in the BKS and in North Atlantic?

No, they do not. See ***slides*** **4, 5, 6**.

5. Are the CH_4 anomalies induced by clouds?

AIRS and IASI techniques have similar cloud filtering procedures based on combined microwave and TIR fields of view and matrices 3x3 (AIRS) and 2x2 (IASI) for TIR pixels. Level 3 AIRS v6 used in the TCD paper are cloud-cleared already. A standard clearing parameter set for IASI was supplied by X. Xiong, namely: ispare(:,1) = 1; rspare(:,4) < 1.8; rspare(:,57)<=3. This technique allows using data for partially-clouded fields of view and removes overcast conditions. As stated in the manuscript, all pixels are cloud cleared.

We further investigated the accuracy of this approach by additional filtering with MODIS data (***slide 10***). MODIS cloud fractions (CF) were gridded to 0.5x 0.5°. The IASI subset data for coinciding grid cells and CF = (0-0.2) and CF = (0.8-1.0) were calculated for 7 boxes stretching from the North Atlantic to the northern Kara Sea (slides 9-10). The thick line (***slide 10***) is for the whole IASI data set, three thin lines are for different subsets with different ranges of CF. We found just a 2 ppb maximum difference between the data. The conclusion is that excluding all data with CF > 0.2 would increase the observed CH_4 only by 2 ppb. Thus, the hypothesis that unfiltered clouds result in increases of retrieved CH_4 by 20-30 ppb is incorrect.

***Slide* 11** shows changes in CH_4 from year to year: mean November -December distribution (A); change of IASI LT methane along the corridor for different years (B); and monthly means for land, sea, and all high Northern latitudes between January 2010 and January 2018 (C).

6. Are the LT IASI and AIRS CH4 concentrations representative for the lower troposphere?

They may be compared with mean 0-4 km CH_4 measured *in-situ* using an aircraft. That was done for three sites in the USA (***slide 12***) [6]. For both instruments, mid-upper tropospheric (MUT) CH_4 has a slope close to 1. LT slopes are 0.4-0.5. We propose these slopes as empirical sensitivities of LT and MUT TIR data to CH_4. These slopes, obtained experimentally, are analogs for the Averaging Kernel (**AK)** matrix:

AK=delta **ret CH_4** / delta **real CH_4**

delta **ret CH_4** is the difference between retrieved profile and a priori profile
delta **real CH_4** is the difference between real profile and a priori profile

Empirical sensitivity, *S* (number), is simply a slope of the regression line:

S = delta retrieved CH_4 / delta real CH_4
"delta retrieved CH_4" and "delta real CH_4" are numbers: profiles averaged over altitude ranges.

Here, *S* is the effective sensitivity for an altitude range; sensitivities to sub-layers within the column may be different. Measured CH_4 anomaly may be interpreted as a weighted average with still unknown weighting function. This function could hardly be derived from experimental data. It may be derived from AK calculated using Radiative Transfer model with a better resolution as is usually done. We assume that the required resolution should be finer than the PBL depth. Layer's depth may be 100-200 m; a two-km resolution would be too coarse.

Conclusion.

We maintain that we have shown that TIR spectrometers can supply important information on lower troposphere methane trends and distributions in the Arctic, both land and sea. Several points require special consideration (vertical sensitivity, including ability to measure PBL CH_4, coastal effects, ice and snow surfaces). Those uncertainties may be studied numerically in conjunction with the observed meteorology for specific Arctic conditions. Ongoing versions of NUCAPS, CLIMCAPS, three IASIs, two CrISs represent a considerable hardware/software basis for that. Standard global NUCAPS algorithm evaluated here is just a first approach, its improved versions may become a critical tool for resolving the problem of Arctic methane.

References

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2. Yurganov L.N., & Leifer I. (2016a). Otsenki emissii metana ot nekotorikh arkticheskikh i priarkticheskikh rayonov po dannym orbital'nogo interferometra IASI (Estimates of methane emission rates from some Arctic and sub-Arctic areas, based on orbital interferometer IASI data). Sovremennye problemy distantsionnogo zondirovaniya zemli iz kosmosa 13(3), 173-183. *DOI: 10.21046/2070-7401-2016-13-3-173-183*
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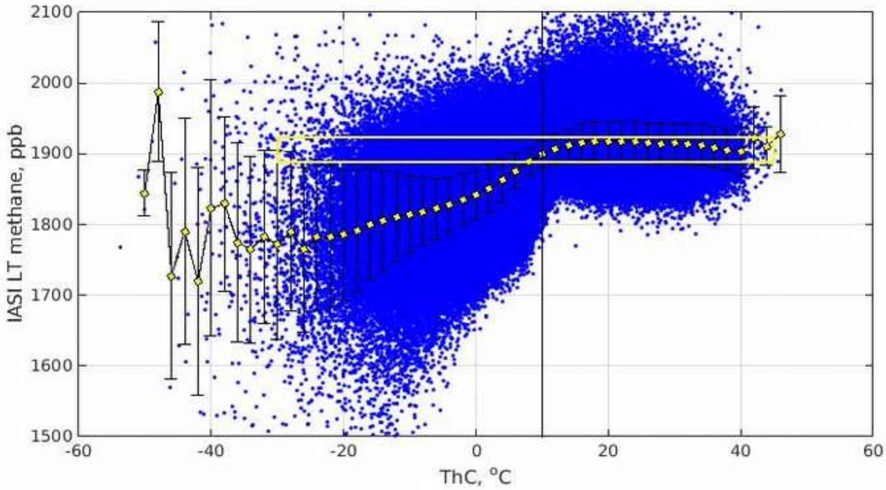
8. Yurganov, L., F. Muller-Karger, and I. Leifer, 2018. Methane Variation Over Terrestrial And Marine Arctic Areas (2010 – 2017): IASI Satellite Data. Rejected by GBC and RSE.
9. vanDamme M. et al. 2014. Global distributions, time series and error characterization of atmospheric ammonia (NH 3) from IASI satellite observations. Atmos. Chem. Phys., 14, 2905–2922, 2014, www.atmos-chem-phys.net/14/2905/2014/, doi:10.5194/acp-14-2905-2014

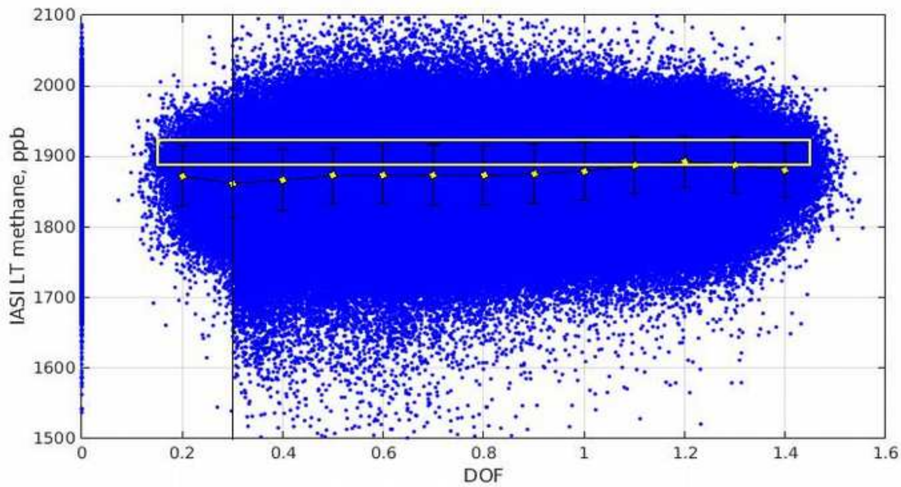
Justification of filtering criteria to exclude unreliable data. Standard flag is DOF >0.3. We use a flag ThC>10 C (temperature difference between the surface and altitude 4 km asl).

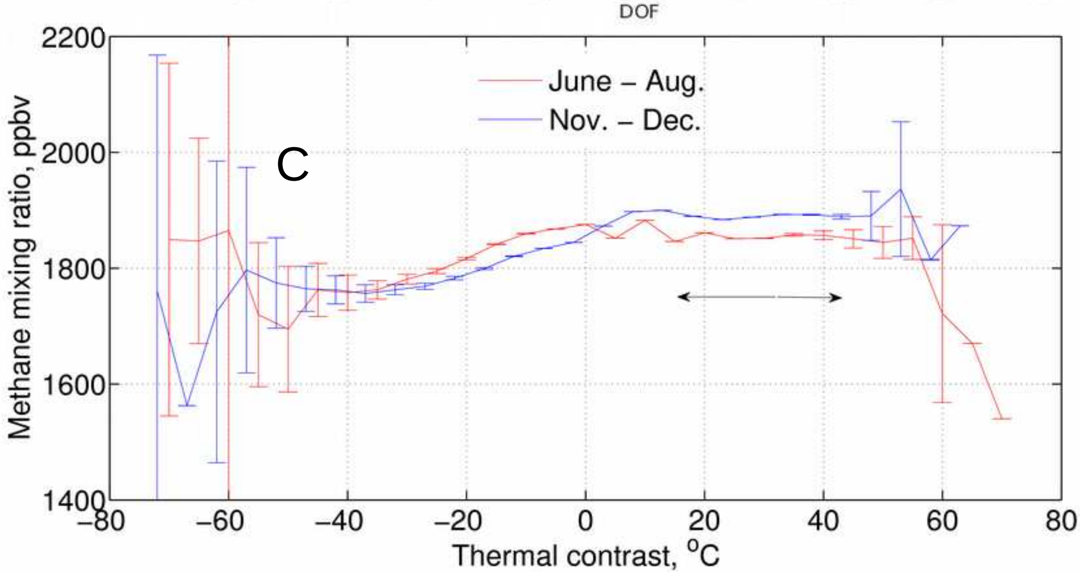
Slide 1

A



B



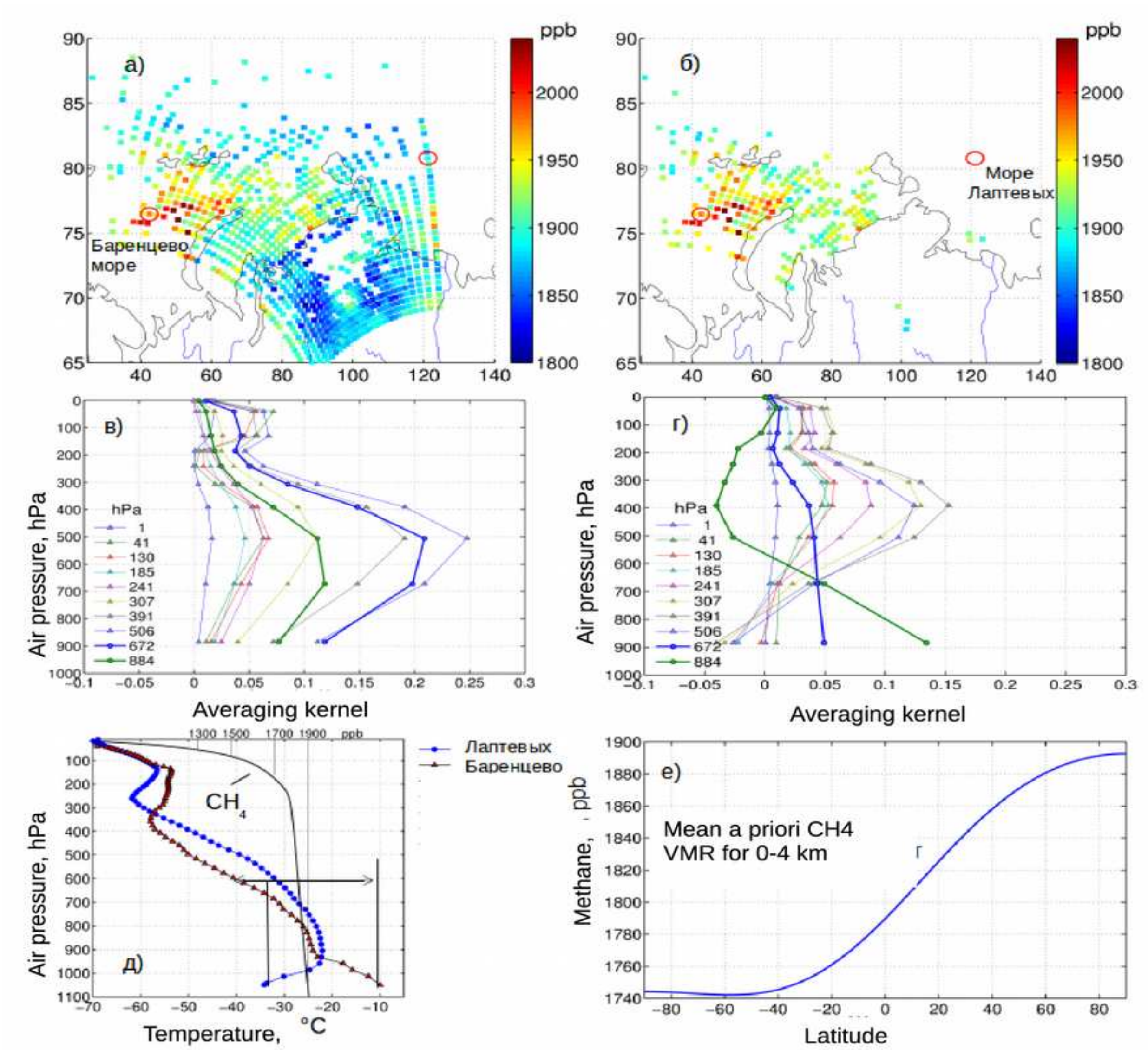


A) A dependence of retrieved LT methane VMR on the Thermal Contrast (ThC). Nov.-Dec. 2009-2012, sea only. For ThC >10° C this plot is flat, i.e., retrieved methane VMR does not depend on ThC [1, 4]. Yellow rectangular corresponds to the range of surface flask measurements at 5 coastal Arctic NOAA sites for the same period (mean ± STD) https://www.esrl.noaa.gov/gmd/dv/data/.

B) A dependence of retrieved LT methane VMR on Degree of Freedom(DOF). 2009-2012, sea only, to the North from 60 N. Error bars for std. [6]

C) IASI lower troposphere CH_4 VMR for for 2010-2016 versus ThC binned with a step 5° Error bars for 4*std /sqrt(N), Land and sea, to the North from 60 N. [8]

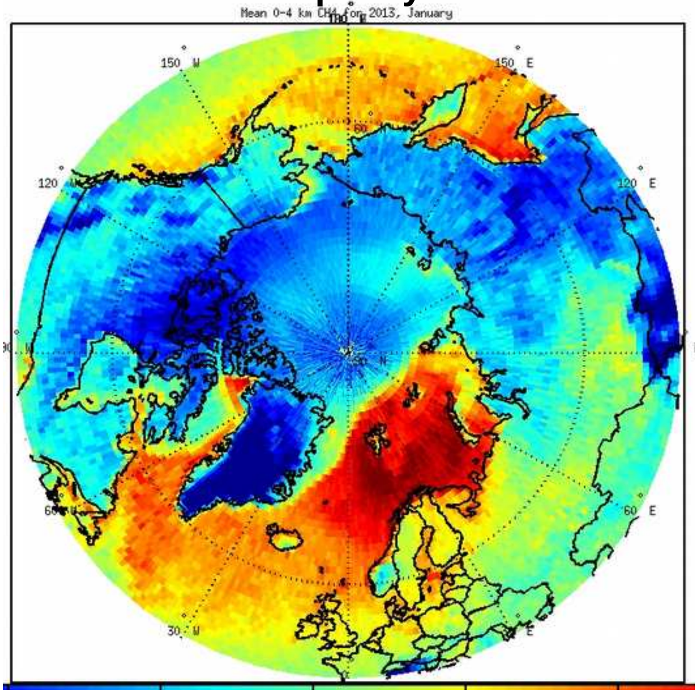
Case study



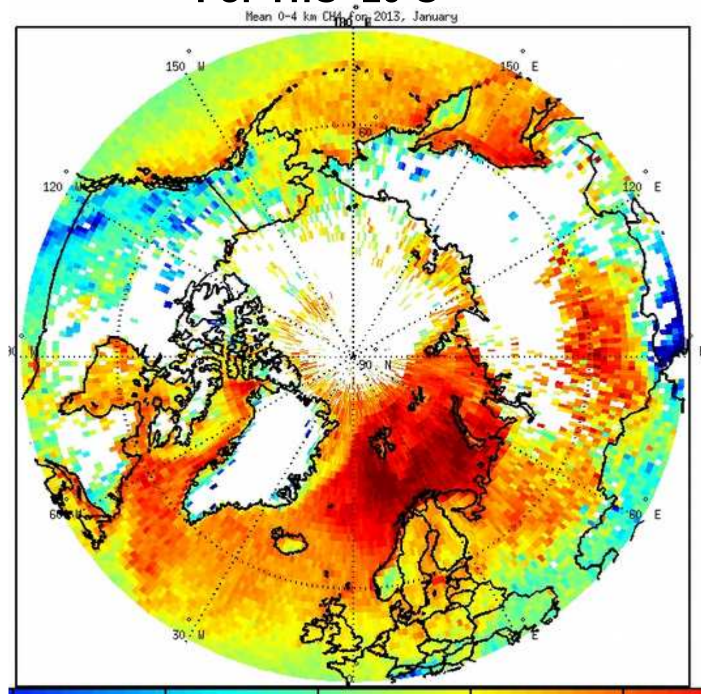
Top. Left: AIRS v6 retrievals for March 6, 2013 for the Arctic selected using DOF and cloudiness criteria only. Red circles for Barents and Laptev seas point out special cases for AK and temperature profiles. Right: Additional points selected for ThC>10 C were removed.
Middle. Averaging kernels for Barents (left) and Laptev (right) cases
Bottom. Left: Temperature profiles retrieved by AIRS: Barents (red triangles) and Laptev (blue circles). Right: A priori CH4 mean 0-4 km concentration for different latitudes, the same for all seasons.
Adopted from [4].
Temperature inversion over Laptev Sea resulted in low ThC disturbs the retrieval algorithm. For the Barents Sea the retrieval is successful, but sensitivity below 600 hPa (~ 4 km) is reduced.

January 2013 monthly IASI L3 CH4 (0-4 km, 1x1 gridded)) before and after ThC filtering

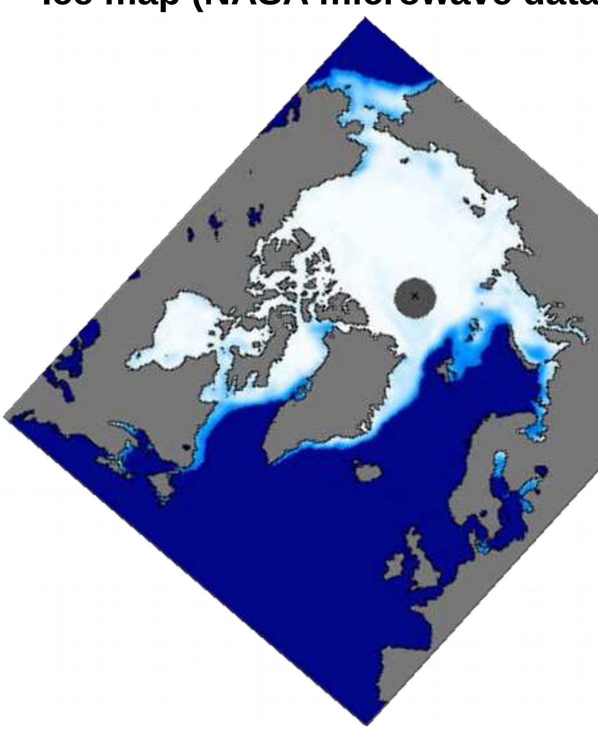
Standard quality control



For ThC>10 C

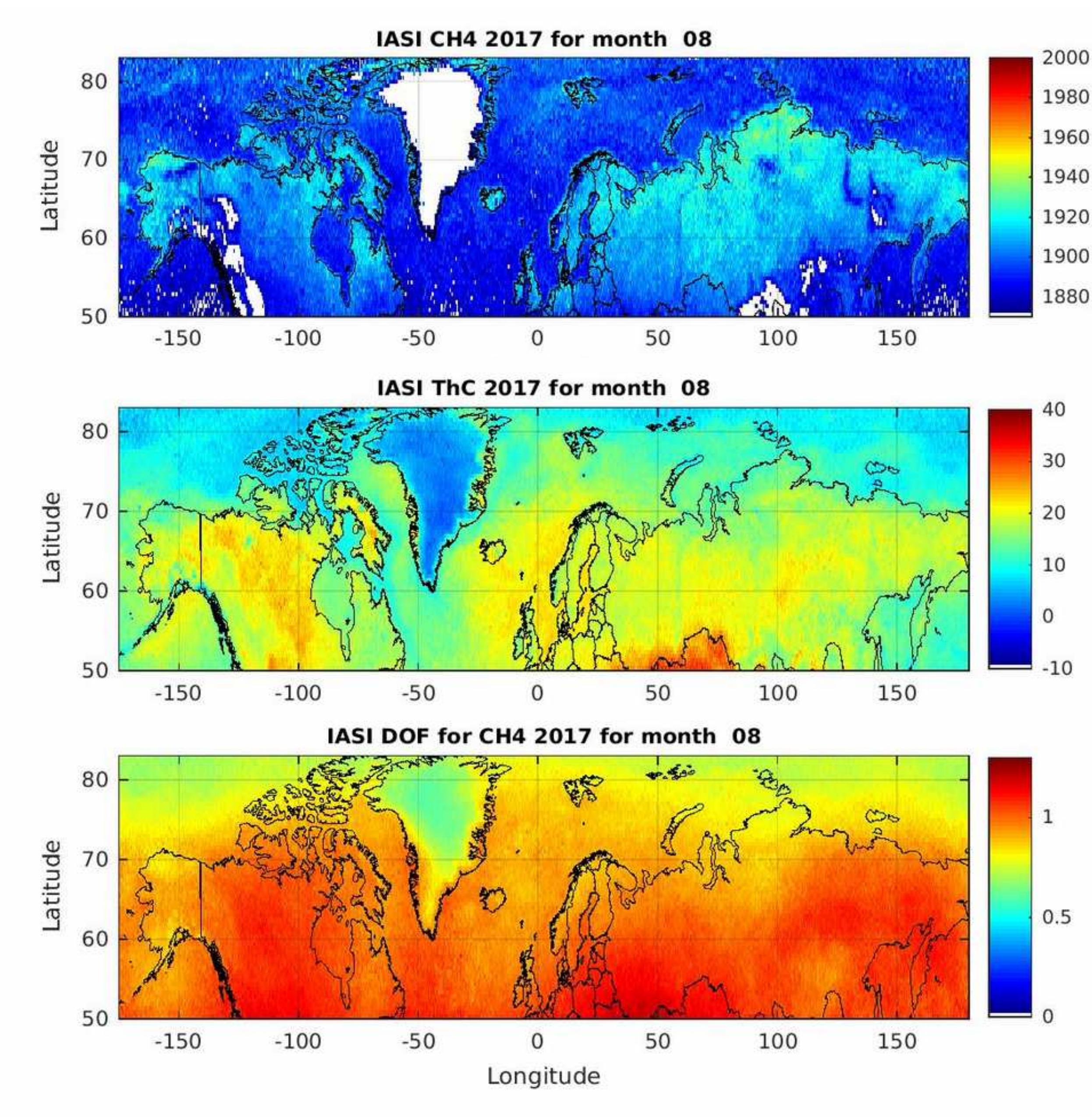


Ice map (NASA microwave data)



The original CH4 data (top left map) is a mixture of individual retrievals with different ThC, from lowest to highest. After filtering CH4 points with low ThC BKS data do not change: that means practically all points are “good”. Over Northern Siberia and ice-covered ocean all retrievals in January are “bad” (blank areas). In South-West Siberia and Europe CH4 levels increase after filtering. South Canadian CH4 were low before and after filtering: CH4 was really low for conditions with high ThC. [1]

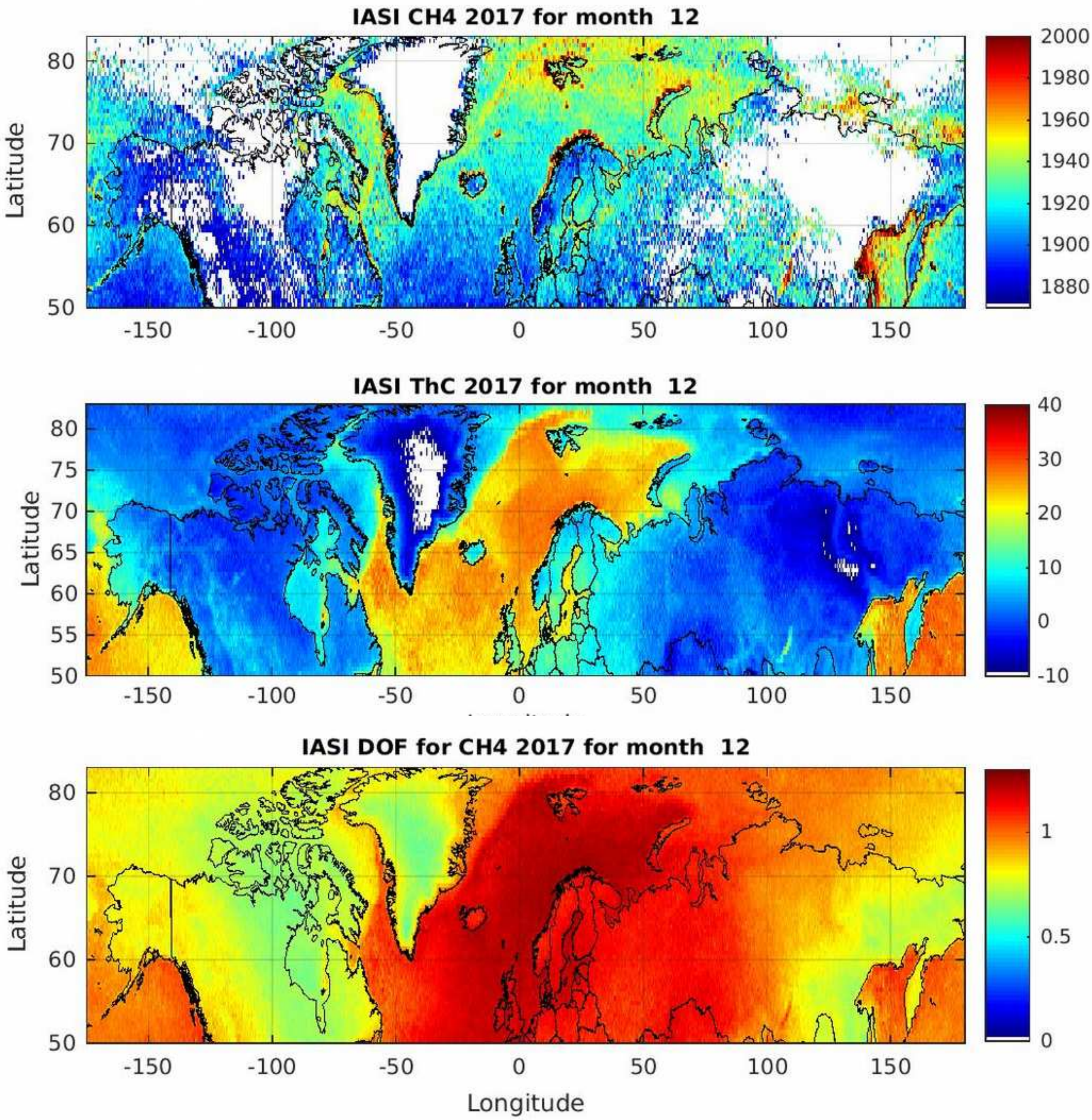
Does CH4 correlate with ThC or DOF? Summer.



Monthly mean IASI LT CH4, ThC and DOF for August 2017

In August CH4 is well mixed horizontally, ThC is between 15 and 20 C, DOF =~0.9

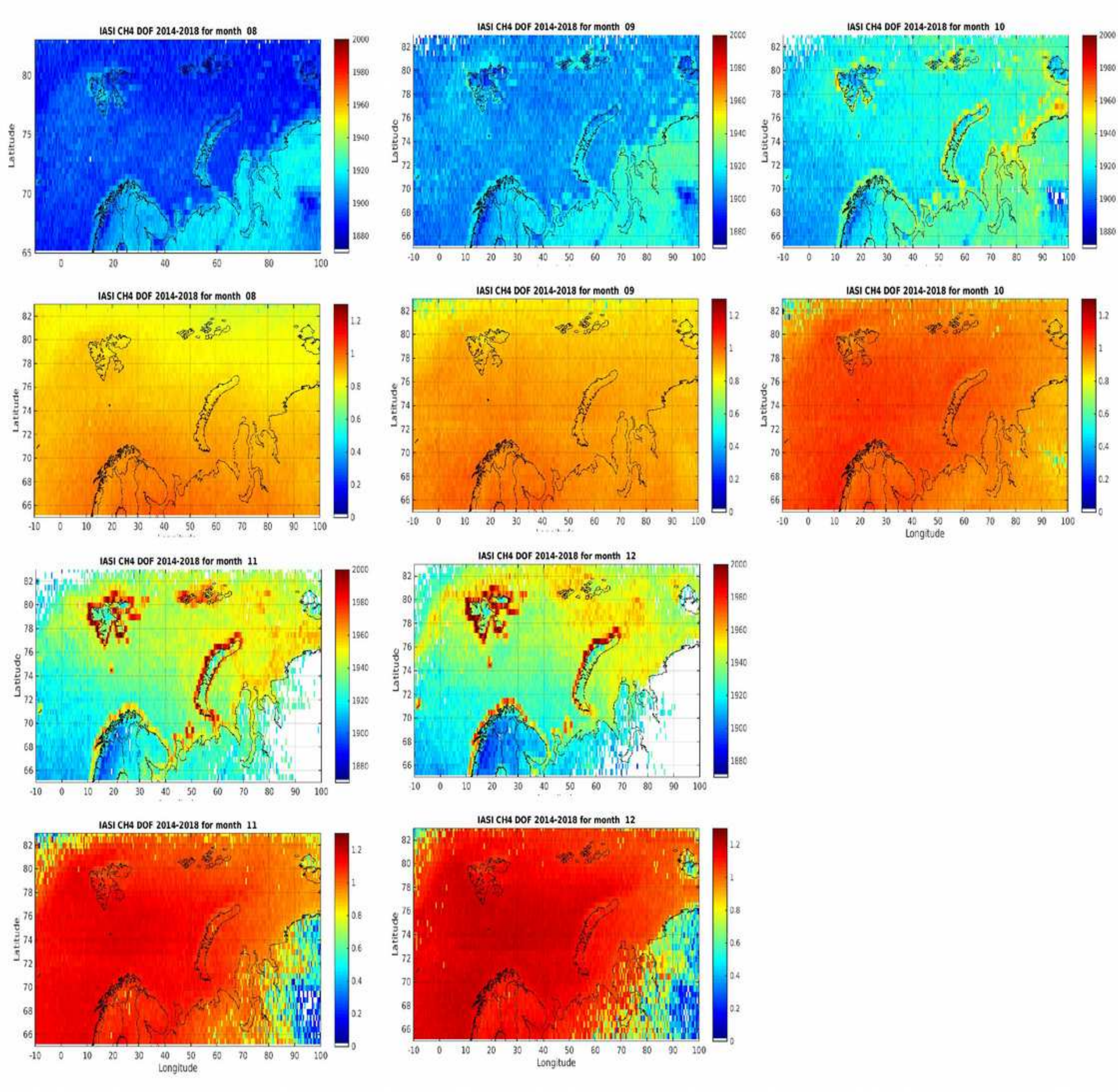
Does CH4 correlate with ThC or DOF? Winter.



Monthly mean IASI LT CH4, ThC and DOF for December 2017.
Top. Blank areas correspond to completely lacking data with ThC<10 C, e.g., East Siberia.

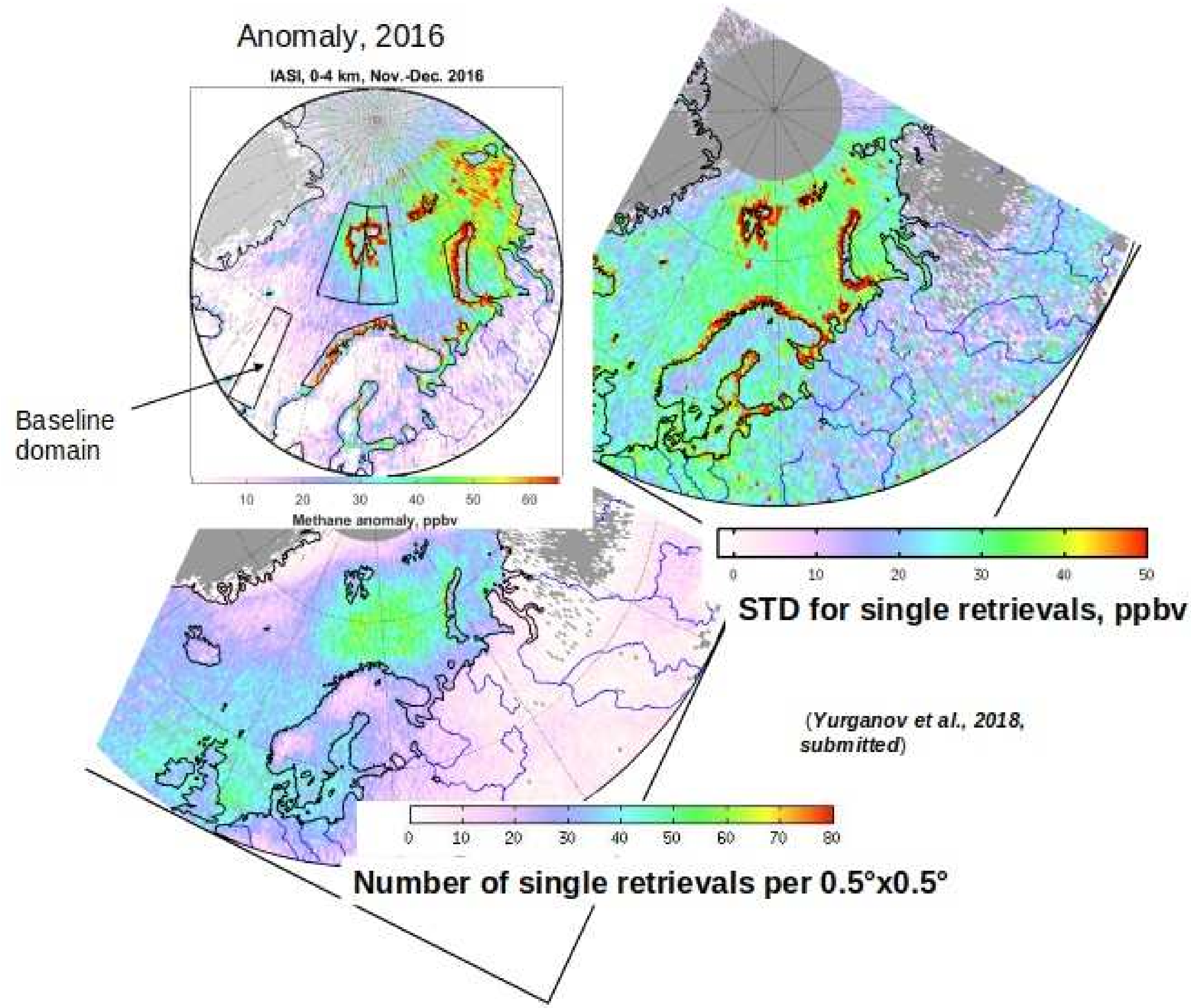
In December LT CH4 in BKS is higher than that between Iceland and Norway by 40-50 ppb. ThC for both areas ~25 C. DOF = (1.2 – 1.3).

CH4 and DOF from August to December



Changes of CH4 retrieved from IASI from August to December (averaging over 2014-2018) and corresponding changes in DOF.

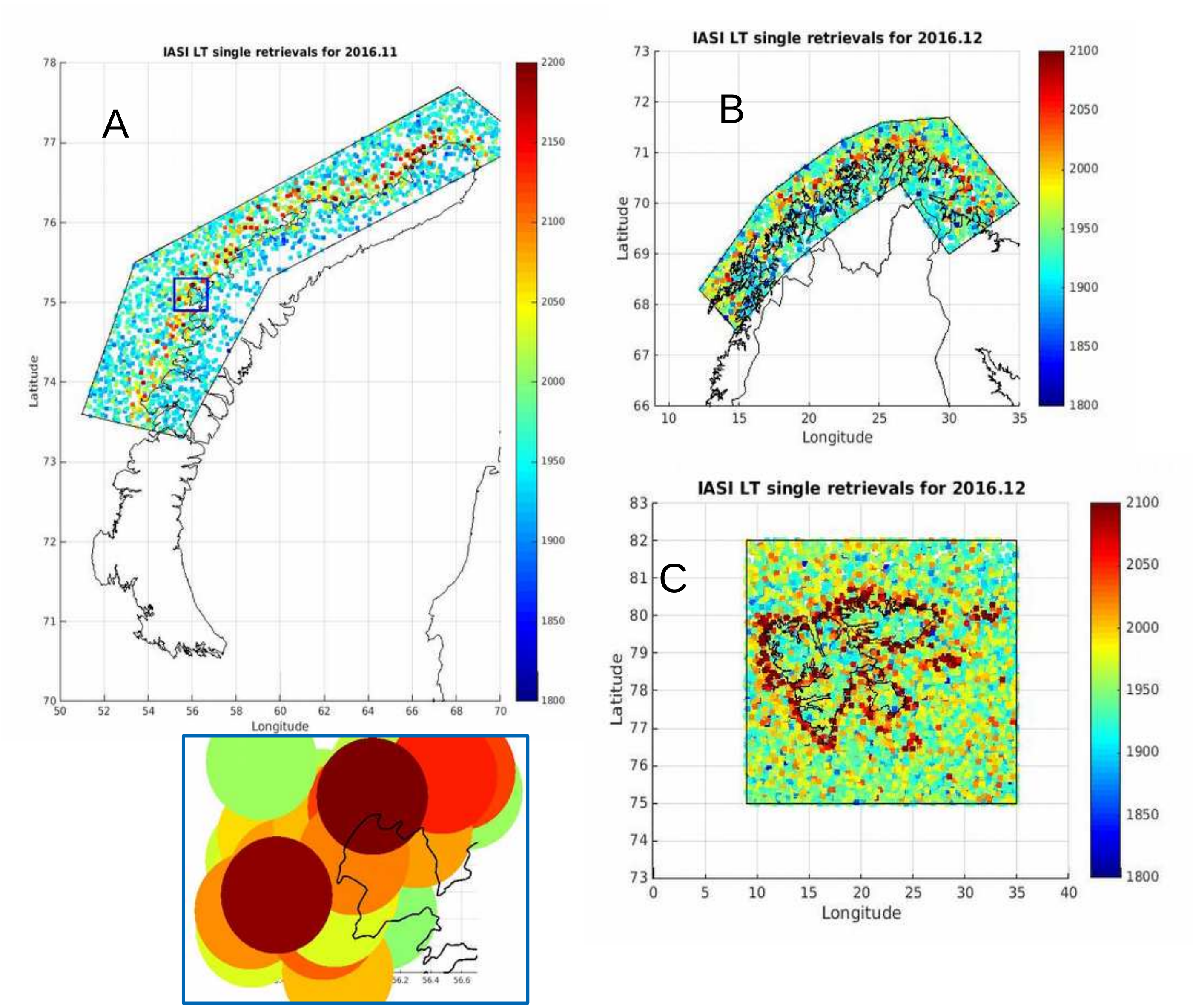
IASI LT CH4 winter anomaly, number of points, variability



Two-month average LT CH4 anomalies, standard deviation and number of points per the grid cell. [8]. Anomalies are calculated as a surplus of CH4 over the baseline domain (shown).

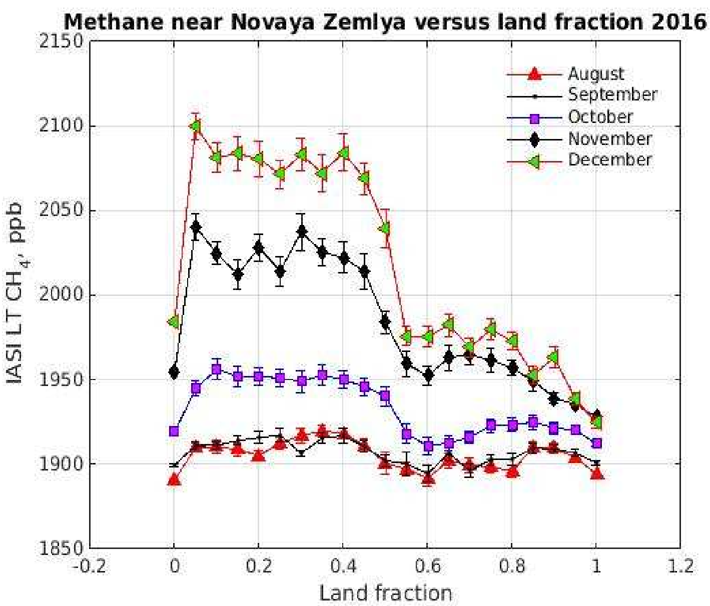
2016 was the year of the maximum CH4 anomaly at the BKS. Number of single retrievals after two types of quality filtering (standard and ThC) in Barents sea is ~ 55 points per 0.5x0.5 cell and two months interval. Typical standard deviation is 30 ppb. Near the shore anomalies and STD increase.

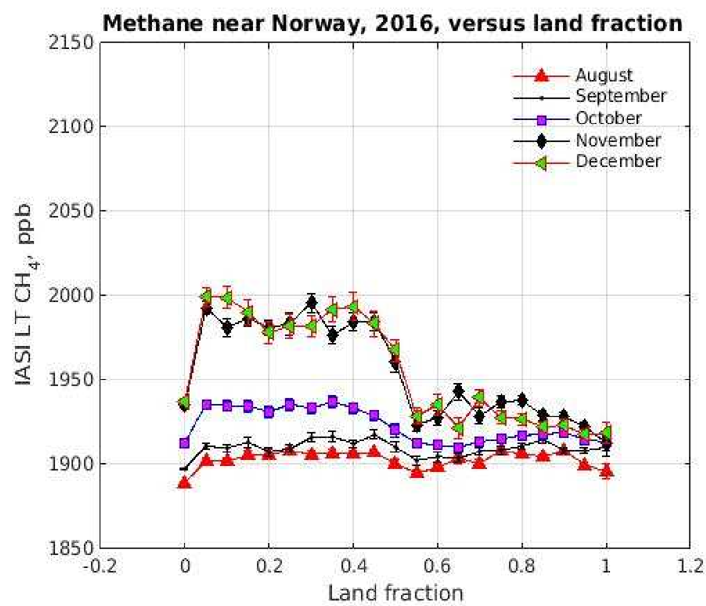
Coastal spikes

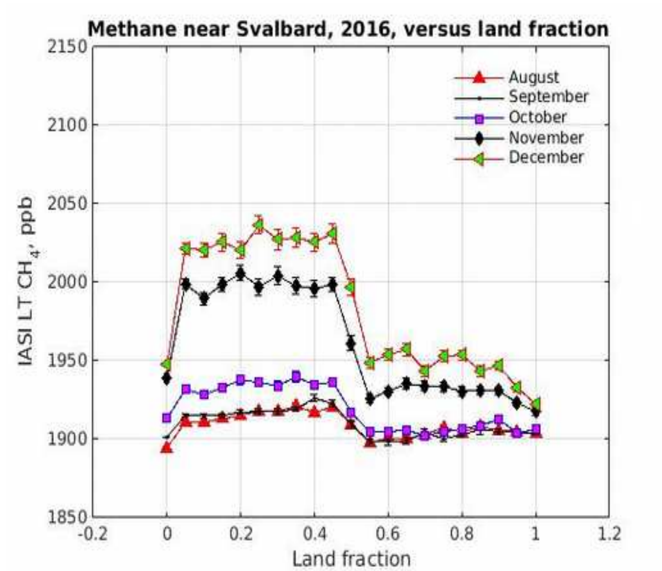


Individual CH4 retrievals for A) Novaya Zemlya, B) Arctic coast of Norway, and C) Spitsbergen for December, 2016. Point size is less than actual to show all points. For A a blue square was zoomed in and the actual size of pixels is shown.

Methane versus fraction of land in a pixel



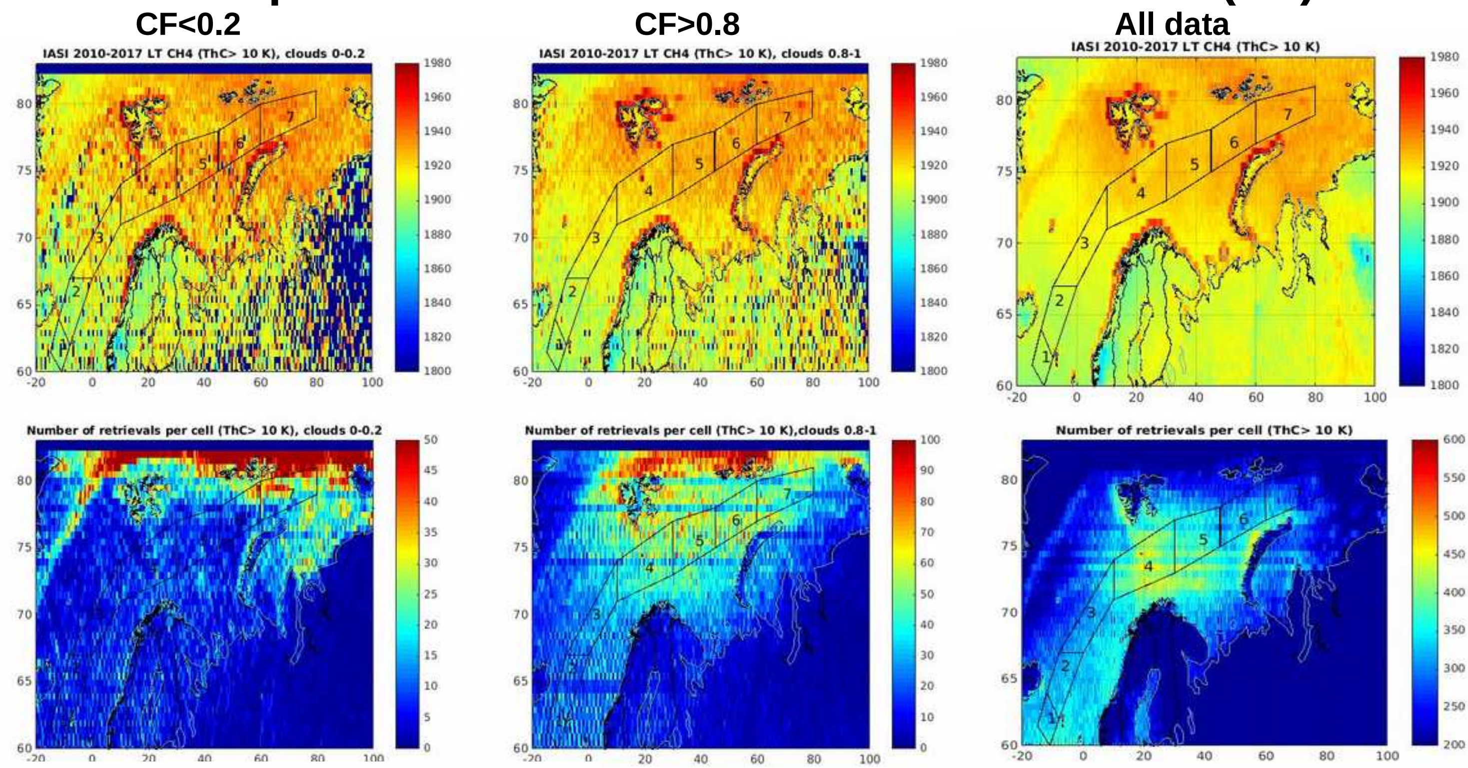


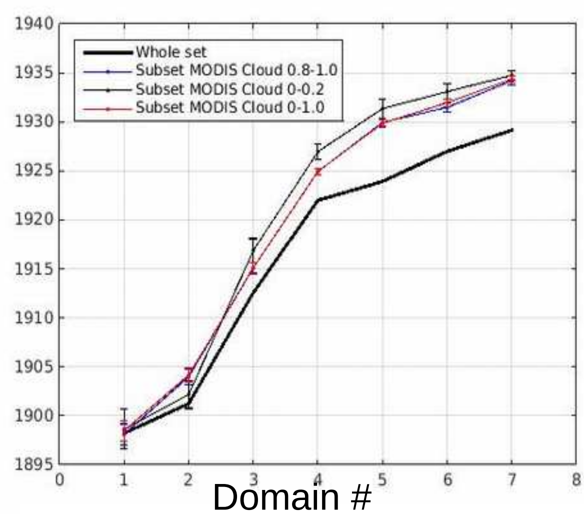


CH4 vs land fraction for three coastal areas in 2016 from August to December. Error bars are 2*STD/sqrt(N).

LT IASI CH4 were plotted versus land fraction for each pixel and binned. Coastal areas for averaging are shown on the previous slide.

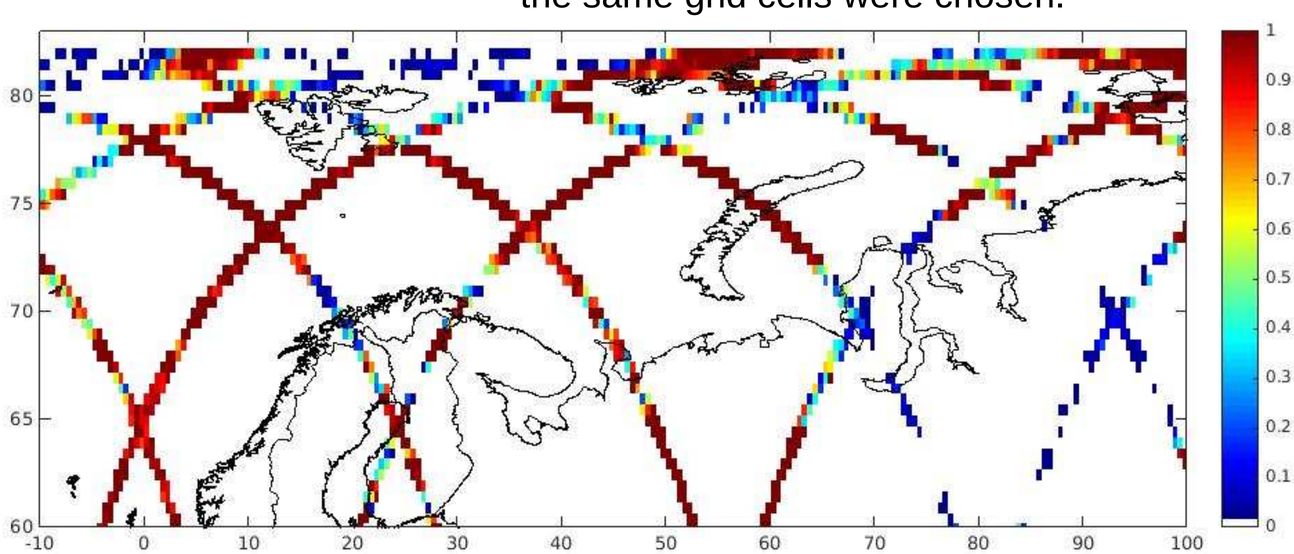
Impact of cloudiness: MODIS cloud fraction (CF)



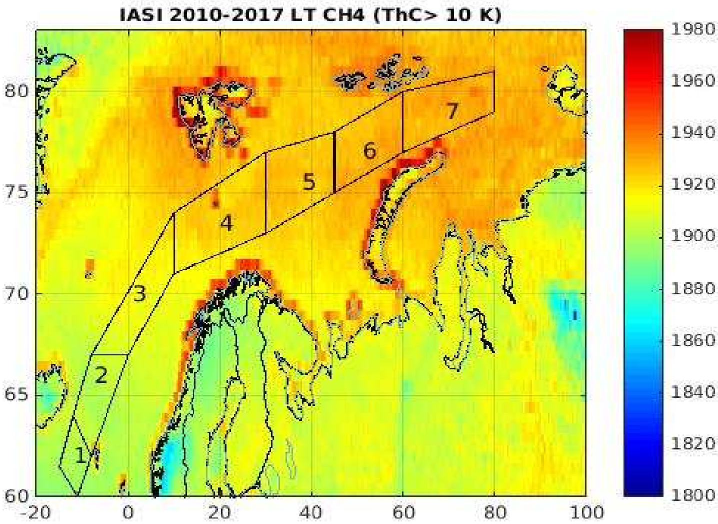


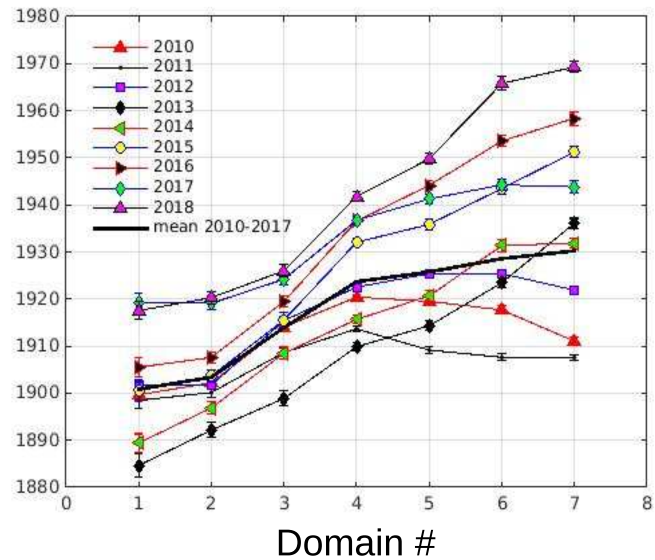
A study of cloud filtering influence on concentrations of methane along the shown corridor for November-December, 2010-2017 time interval. Numbers on X-axis correspond to the domain number.

MODIS cloud fraction data (example for one day, Nov. 26, 2010, is below) were used for additional filtering IASI results. MODIS daily data were gridded for 0.5x0.5 grid and a subset of CH_4 for the same grid cells were chosen.

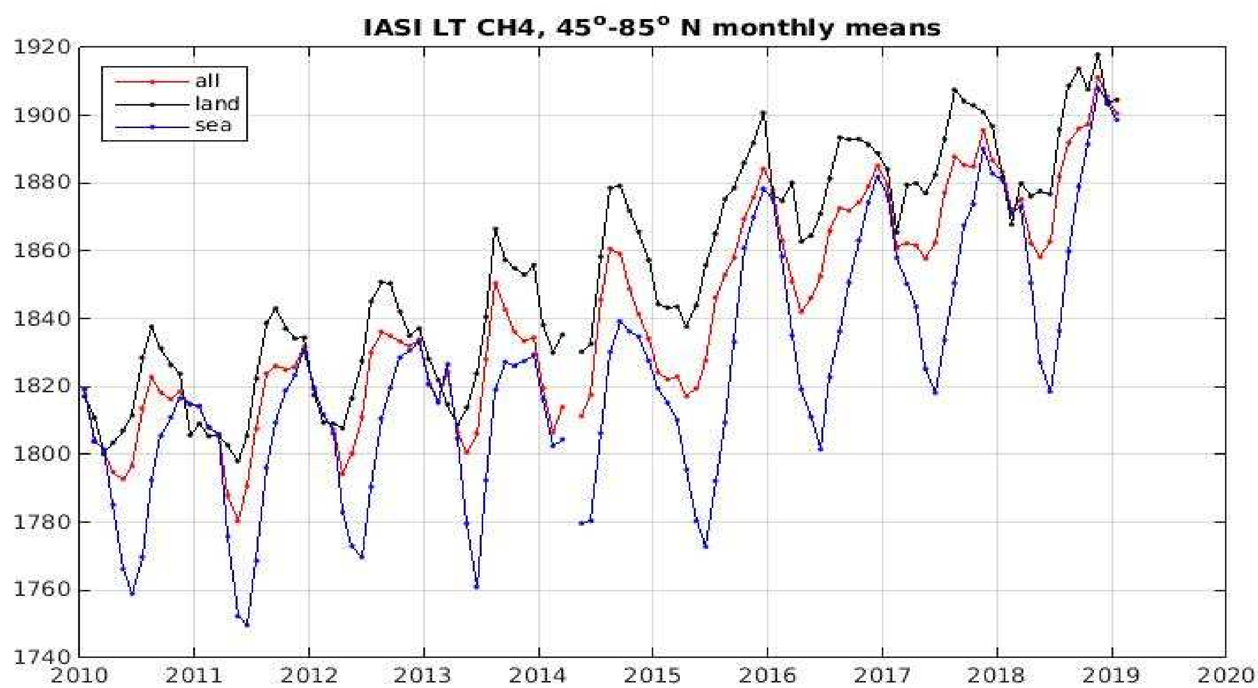


Nov.-Dec. Cross section and monthly means for land and sea



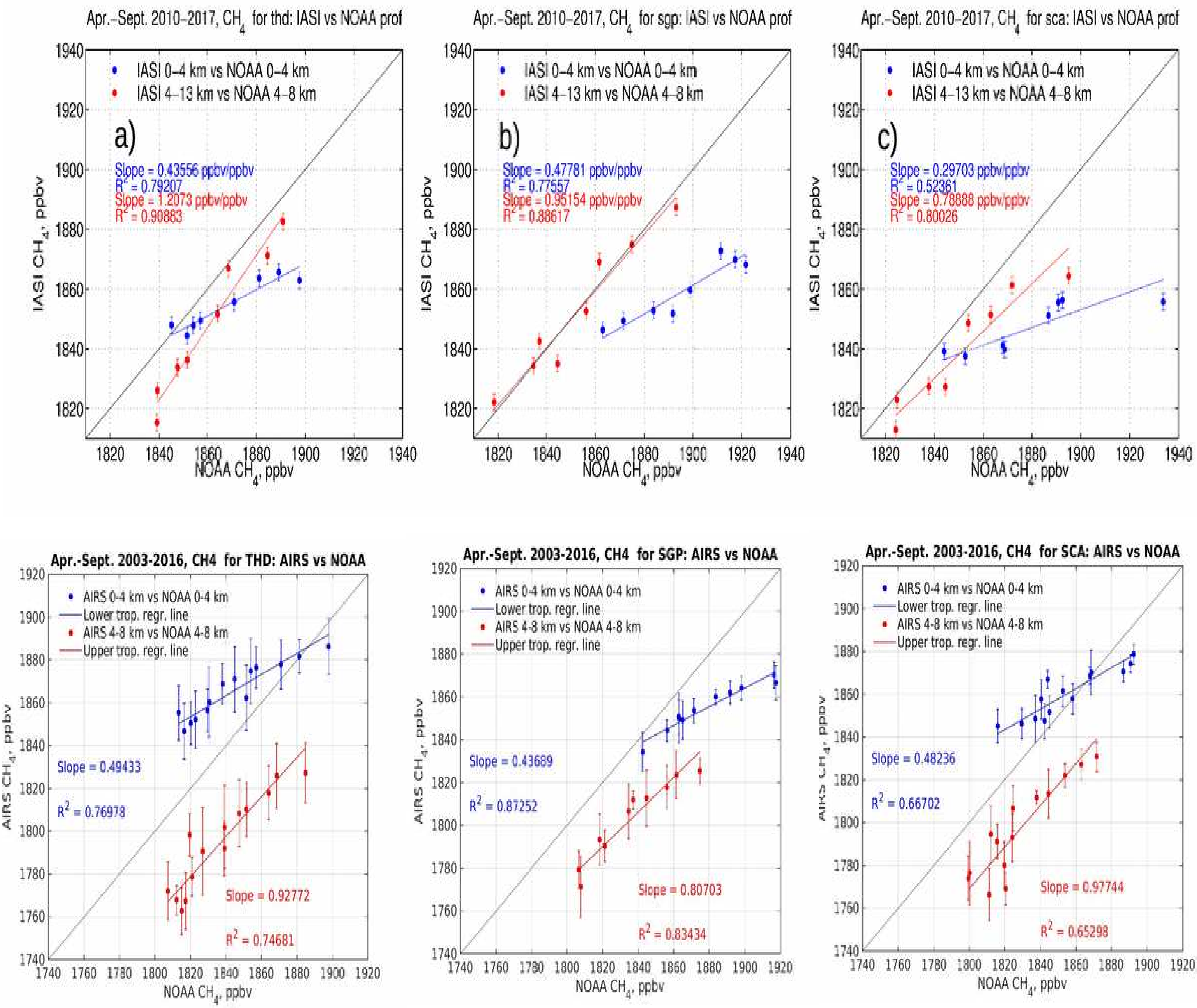


Left: November-December all CH4 data averaged over 8-years-long period, a corridor for the cross-section is shown and domains are numbered. Right: cross-sections for consecutive years along the corridor. Numbers correspond to the domains for averaging. Error bars are for 2*STD/sqrt(N).



Monthly IASI LT CH4 averaged over land, sea and together in the latitudinal belt 45 N – 85 N between January, 2010 and January 2019. Land CH4 has a maximum in August-September, minimum – in April-May. CH4 over sea has a maximum in November-December-January, minimum – in June. Winter data over Siberia and Canada are mostly lacking, as well as data over thick ice around the Pole.

IASI and AIRS validation with aircraft ESRL data



April-September IASI (top) and AIRS (bottom) CH4 data for low troposphere (below 4 km) and upper troposphere (above 4 km) compared with concentrations measured from NOAA aircrafts at 3 sites in USA: Trinidad Height, CA (THD), Southern Great Plain, OK (SGP), and Charleston, SC (SCA). Slopes for LT are in the range 0.4-0.5, for MUT they are in the range 0.8 – 1.2. The plots evidence lower sensitivity of both instruments to LT layers. The airborne data courtesy Colm Sweeney (GMD/ESRL).