

1 Large XCH₄ anomaly in summer 2013 over Northeast 2 Asia observed by GOSAT

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16 Abstract

17 Extremely high levels of column-averaged dry-air mole fractions of atmospheric
18 methane (XCH₄) were detected in August and September 2013 over Northeast Asia
19 (~20 ppb above the averaged summertime XCH₄ over 2009-2012, after removing a
20 long-term trend), as being retrieved from the Short-Wavelength InfraRed (SWIR)
21 spectral data observed with the Thermal And Near-infrared Sensor for carbon
22 Observation - Fourier Transform Spectrometer (TANSO-FTS) onboard Greenhouse
23 Gases Observing Satellite (GOSAT). Similar enhancements of XCH₄ were also
24 observed by the ground-based measurements at two Total Carbon Column Observing
25 Network (TCCON) sites in Japan.

26 The analysis of surface CH₄ concentrations observed at three monitoring sites
27 around the Japan islands suggest that the extreme increase of XCH₄ has occurred in a
28 limited area. The model analysis was conducted to investigate this anomalously high

1 XCH₄ event, using an atmospheric transport model. The results indicate that the
2 extreme increase of XCH₄ is attributed to the anomalous atmospheric pressure pattern
3 over East Asia during the summer of 2013, which effectively transported the CH₄-rich
4 air to Japan from the strong CH₄ source areas in East China. The two Japanese
5 TCCON sites, ~1,000 km east-west apart each other, coincidentally located along the
6 substantially CH₄-rich air flow from East China. This analysis demonstrates the
7 capability of GOSAT to monitor an XCH₄ event on a synoptic scale. We anticipate
8 that the synoptic information of XCH₄ from GOSAT data contributes to improve our
9 understanding of regional carbon cycle and the regional flux estimation.

10

11 **1 Introduction**

12 Atmospheric methane (CH₄) is the second important anthropogenic greenhouse gas
13 after carbon dioxide (CO₂), contributing about 20 % of the total radiative forcing from
14 the major well-mixed greenhouse gases (Forster et al., 2007). Methane has multiple
15 natural and anthropogenic sources in the Earth's surface while being mainly removed
16 through reaction with hydroxyl radical (OH) in the troposphere and by photolysis in
17 the stratosphere. The atmospheric CH₄ level has more than doubled since the onset of
18 the industrial revolution in the 18th century (Etheridge et al., 1998). Its growth rate
19 has been considerably variable over the past few decades (Dlugokencky et al., 2009).
20 On a global scale, the causes of recent changes in the CH₄ growth rate remain
21 unknown (e.g. Kirschke et al., 2013; Dlugokencky et al., 2009), and on a regional
22 scale, significant discrepancies have been found in the emission estimates between
23 bottom-up and top-down approaches (e.g. Miller et al., 2013). On the other hand,
24 given the larger radiative forcing than carbon dioxide, it has been argued that reducing
25 anthropogenic CH₄ emission might be a mitigation of possible severe impact of global
26 warming (e.g. Hansen and Sato, 2004). Therefore, to elucidate the drivers of changes
27 in atmospheric CH₄ concentrations and to quantify the regional source distributions
28 are challenging tasks.

29 The temporal variations of observed atmospheric CH₄ are complicated due to
30 various sources on the Earth surface, interactions between the emission sources and
31 the atmospheric transport, and removal in the atmosphere. To improve the regional

1 CH₄ flux estimates on the Earth surface, it is needed to better understand the relative
2 contribution of atmospheric transport to the observed variations of atmospheric CH₄.

3 In the past decades, the investigations of the spatiotemporal variability in
4 atmospheric CH₄ concentrations and the inverse modeling estimates of surface CH₄
5 flux estimates had been mainly based on the ground-based measurements including
6 aircraft and shipboard measurements (e.g. Bousquet et al., 2006; Bergamaschi et al.,
7 2010; Miller et al., 2013). However, the current ground-based measurements of CH₄
8 are still sparse. In the recent years, the measurements from the satellites have been
9 providing the large spatial and temporal coverage to help better understand the
10 variations of atmospheric CH₄. Greenhouse gases Observing SATellite (GOSAT)
11 was launched in January 2009, providing column-averaged dry-air mole fractions of
12 atmospheric CH₄ (XCH₄) that are retrieved from Short-Wavelength InfraRed (SWIR)
13 solar spectra observed onboard Thermal And Near infrared Sensor for carbon
14 Observation - Fourier Transform Spectrometer (TANSO-FTS) instrument (Yokota et
15 al., 2009; Yoshida et al., 2013). The GOSAT TANSO-FTS aims at measurements of
16 atmospheric CH₄ concentrations in three-month averages with an accuracy of better
17 than 2 % at 100-1,000 km spatial resolution (Kuze et al., 2009). GOSAT XCH₄ is
18 preceded by the several previous and on-going satellite projects, for example, the
19 Infrared Atmospheric Sounding Interferometer (IASI, Crevoisier et al., 2009), and the
20 Tropospheric Emission Spectrometer (TES, Wecht et al., 2012) and the SCanning
21 Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY,
22 Schneising et al., 2011). Among them, XCH₄ retrievals from SCIAMACHY
23 instrument onboard ENVISAT launched in 2003 was pioneering, but the
24 communication with ENVISAT was lost in April 2012. These satellite data have been
25 used for the inversion studies of surface CH₄ emissions. Most of the satellite-based
26 inversions are focused on the global-scale estimates (e.g., Bergamaschi et al. 2007;
27 2009; 2013; Fraser et al., 2013; Moteil et al, 2013; Cressot et al, 2014; Houweling et
28 al., 2014; Alexe et al., 2015). Recently the satellite data have been applied for the flux
29 estimation on a regional- and local-scale at a higher spatial resolution. For example,
30 Wecht et al. (2014) compared the multiple observational constraints including
31 GOSAT and TES to optimize methane emission in California. Turner et al. (2015)
32 estimated North American methane emission at a resolution of up to 50 km × 50 km
33 using GOSAT data. Kort et al. (2014) demonstrated that satellite-based observations

1 can quantify localized anthropogenic CH₄ emissions in the Southwest USA using the
2 SCIAMACHY and Total Carbon Column Observing Network (TCCON) data.

3 Here, we report the extremely high XCH₄ event observed by GOSAT in August
4 and September 2013 over Northeast Asia. Similar high XCH₄ event were also
5 detected by the ground-based measurements at the two Japanese TCCON sites in
6 Tsukuba and Saga. Given the spacing and temporal frequency (three-day recurrence)
7 of GOSAT sampling, along with possible retrieval biases of XCH₄ retrievals, it is
8 interesting that the GOSAT detected the synoptic-scale variation of XCH₄ that is
9 coherent with the ground-based measurements. This GOSAT-detected XCH₄ event
10 suggests the potential of GOSAT XCH₄ analysis in higher temporal and spatial
11 resolution. The capability to capture synoptic-scale variations of atmospheric CH₄
12 leads to better regional flux estimation because the synoptic-scale variations of
13 atmospheric CH₄ can carry the information on regional surface fluxes. On the other
14 hand, the atmospheric CH₄ concentrations are highly changeable with the atmospheric
15 transport as well as surface fluxes. Toward improving regional flux estimation, it is
16 essential to observe better a synoptic-scale variation of the atmospheric CH₄ and
17 quantify the attribution of such variations.

18 In this study, we analyse the extremely high XCH₄ observed by GOSAT in the
19 summer of 2013 and investigate the attributions of such a significant increase of
20 XCH₄. We discuss how capable GOSAT XCH₄ is to monitor synoptic-scale XCH₄
21 variations.

22

23 **2 Observations**

24 **2.1 GOSAT XCH₄**

25 GOSAT is a joint project of the Japanese Ministry of the Environment (MOE), the
26 National Institute for Environmental Studies (NIES) and the Japan Aerospace
27 Exploration Agency (JAXA) to monitor the global distribution of atmospheric CO₂
28 and CH₄ from space (Yokota et al., 2009). The retrieved XCH₄, as a part of NIES
29 GOSAT Level 2 (L2) product (v02.xx), has been reported to have a mean bias of -5.9
30 ppb and mean standard deviation of 12.6 ppb against the XCH₄ at selected TCCON
31 sites (Yoshida et al., 2013). In this study, we analysed NIES GOSAT L2 XCH₄

1 (v02.21) without any bias correction. The description of the latest updated retrieval
2 procedures and the auxiliary information can be found at GOSAT User Interface
3 Gateway (<https://data.gosat.nies.go.jp>).

4 We analysed GOSAT XCH₄ over two regions in Northeast Asia separately
5 (Fig. 1). One is over northeastern China-Korea (115°E-130°E, 30°N-40°N), and the
6 other is over Japan (130°E-145°E, 30°N-40°N). The northeastern China-Korea region
7 covers highly populated and industrialized areas with large anthropogenic CH₄
8 sources in the Eurasia continent. The Japan region has small CH₄ sources, but located
9 downwind of the continental CH₄ emissions. Time-series of XCH₄ data from June
10 2009 to March 2014 over the two regions with monthly means are shown in Fig. 2. It
11 is noted that we used only the XCH₄ over land to minimize possible errors depending
12 on sounding observation mode (Fig. 3). In fact, since a few soundings over ocean
13 around East Asia were retrieved, removed XCH₄ data through this criterion are less
14 than 5% of the total. A long-term trend component in each XCH₄ dataset derived
15 through a digital filtering of two-year cutoff period (Nakazawa et al., 1997) is also
16 plotted in Fig. 2. To focus the seasonal variations, the trend components were
17 removed, and the detrended XCH₄ time-series are further analysed.

18 The GOSAT XCH₄ retrievals over Northeastern China-Korea have clear
19 seasonality with high peaks in summer and low peaks in winter. The summertime
20 high XCH₄ appear to be influenced by the seasonal biogenic CH₄ emissions from rice
21 paddies and natural wetlands underneath in East China and Korea. The summer peak
22 in 2013 was more prominent than the preceding two years, 2011 and 2012. Also, the
23 summertime XCH₄ retrievals over Northeastern China-Korea in 2009 and 2010 were
24 relatively high while no significantly high XCH₄ was observed over Japan. Since
25 there is a limited number of retrieval available over Japan for the first two years of the
26 GOSAT operation, it is difficult to discuss the XCH₄ difference over the two regions
27 for 2009 and 2010. We thus leave this topic for a future investigation.

28 The seasonality of the GOSAT XCH₄ retrieval over Japan is overall similar to
29 Northeastern China-Korea. Although the seasonal cycle varies largely year-to-year,
30 XCH₄ retrievals of August and September in 2013 were outstandingly high. Japan is
31 located downwind of strong anthropogenic and natural biogenic CH₄ emissions in the
32 continent, and then the signals of the continental CH₄ emissions are lowered as the air
33 is transported. However, it is noticeable that, in the summer of 2013, the XCH₄

1 retrievals over both Japan and Northeastern China-Korea regions reached the almost
2 same high levels. This comparable XCH₄ levels in the two regions indicates there
3 was a mechanism of fast atmospheric transport in 2013 to bring CH₄-rich air to Japan
4 with less diffusion than the preceding years.

5 The number of GOSAT retrievals over Japan increased in 2013 and 2014,
6 compared with those in the previous years. This increase is due to the change of the
7 observation strategy to increase available GOSAT retrievals over Japan. The initial
8 regular schedule, there were fewer soundings over lands, but most soundings were
9 over oceans or land-ocean mixed locations. The soundings over ocean or mixed
10 locations are difficult to be retrieved. As a result, a few retrievals remained over
11 Japan after screening. Aiming at increasing the retrievals over Japan, the observation
12 locations were moved inland from ocean and mixed locations as much as possible.
13 The observation strategy change was made as a concerted decision by GOSAT Project
14 terms among the three agencies NIES, JAXA, and MOE. This observation change
15 was implemented on May 6, 2013.

16 The spatial maps of GOSAT retrievals we used in this study are shown in Fig. 3. As
17 seen in Fig. 3a, most of the soundings were taken at lower surface elevation; more
18 than 80% is below 100 m, 95% is below 500 m. The spatial maps of the retrievals in
19 August and September in 2012 and 2013 are shown in Fig. 3b and Fig. 3c,
20 respectively. Kort et al. (2014) applied the elevation correction to SCIAMACHY XCH₄ over
21 in the Southwest USA. The elevation impact is mainly exhibited over the Rocky Mountains
22 because of low CH₄ air in the stratosphere. Surface elevation/topography could be one of
23 potential biases in GOSAT XCH₄. However, for the GOSAT data we used, we found no
24 statistically significant correlation with surface elevation ($r = -0.11$) and also the GOSAT
25 XCH₄ data sampled at elevation of more than 1000m is a few (~0.1% out of total). Therefore,
26 no elevation correction was applied to the GOSAT XCH₄ in this study. In Figs. 3 (b) and (c),
27 the observation strategy change mentioned above is noticeable, the coverage of
28 retrievals over Japan was dramatically increased in 2013, compared with 2012. As far
29 as the China-Korea, there is no significant difference between 2012 and 2013.

30 **2.2 TCCON XCH₄**

31 Inside the Japan region of this study, ground-based XCH₄ measurements have been
32 conducted at two TCCON sites, Saga (33.24°N, 130.29°E) and Tsukuba (36.05°N,

1 140.12°E) as shown in Fig. 1. TCCON is a worldwide network of ground-based high-
2 resolution FTSs, which record spectra of the direct sunlight in the near-infrared, and
3 provides accurate and precise column-averaged dry-mole fractions of atmospheric
4 constituents including CO₂, CH₄, N₂O, HF, CO, H₂O, and HDO retrieved from these
5 spectra absorbed by them (Wunch et al., 2011). The TCCON XCH₄ measurements
6 have an estimated uncertainty of 7 ppb (2σ) (Wunch et al., 2010). TCCON data play a
7 critical role in the validation of space-based measurements. The Saga TCCON site is
8 in Kyushu Island, operated by JAXA since June, 2011. The Tsukuba TCCON site is
9 located ~50 km north of Tokyo in the Japan main island, operated by NIES since
10 2009. These two Japanese TCCON sites are apart ~1,000 km longitudinally. In this
11 study, we use the TCCON data processed by GGG 2012.

12 Figure 4 shows XCH₄ retrievals at Saga and Tsukuba TCCON sites during the
13 period for 2011 to 2014. We processed the both TCCON XCH₄ time-series in the
14 same manner with the GOSAT XCH₄ to obtain the long-term trends that are shown in
15 blue lines in Fig. 4. It is interesting that, before the summer 2013, XCH₄ retrievals at
16 Tsukuba overall are lower than at Saga. Since Saga is located closer to the continent
17 than Tsukuba, Saga is considered to be influenced by the continental anthropogenic
18 CH₄ emissions more strongly than Tsukuba. In the summer of 2013, extremely high
19 XCH₄ retrievals both at Saga and Tsukuba were observed, reaching almost a same
20 level. This XCH₄ enhancement observed at the ground-based TCCON sites is
21 coincident with the high XCH₄ observed by GOSAT, and strongly supports our
22 speculation that the CH₄ rich air was transported quickly from the continent to Japan
23 for this period.

24 To focus on the seasonal and synoptic variations, we compared the detrended
25 XCH₄ time-series from GOSAT over Japan and the two Japanese TCCON sites.
26 Figure-5a shows that all the detrended XCH₄ data are overall in phase of seasonal
27 cycle with seasonal amplitude of ~20 ppb. Compared with TCCON XCH₄, GOSAT
28 XCH₄ shows large short-term variability. In 2013, both GOSAT and TCCON XCH₄
29 together rapidly increased in August and remained high in September. In 2012, both
30 GOSAT XCH₄ and TCCON XCH₄ show no clear tendency in August, and all of them
31 appear to be upward in September. On average, the XCH₄ level of GOSAT over Japan
32 in August and September 2013 is higher by ~15 ppb than 2012. The XCH₄ levels of

1 both TCCON sites in 2013 are higher by ~20 ppb than 2012. These enhancements of
2 XCH₄ are comparable to their seasonal amplitude.

3 To examine further how the synoptic variability of GOSAT is correlated with
4 TCCON, we removed the mean seasonal cycles from the detrended XCH₄ time-series
5 and took the monthly means (Fig. 5b). Except the months when the retrievals are
6 available for less than two days, the correlation coefficients (*r*) of the monthly means
7 between GOSAT and TCCON at Saga, and between GOSAT and TCCON at Tsukuba,
8 are 0.81 and 0.61, respectively. These correlation coefficient values exceed the 95%
9 significance level. Despite the large short-term variability, the synoptic variability of
10 GOSAT over Japan is overall correlated with the TCCON XCH₄ at two Japanese sites.
11 The enhancement of XCH₄ in the summer of 2013 is consistent among GOSAT and
12 TCCON. If the period is limited to May–December 2013, when the number of
13 GOSAT XCH₄ retrievals was increased due to the observation strategy change
14 mentioned earlier, the correlation coefficients (*r*) between GOSAT and TCCON are
15 improved to be 0.91 with Saga and 0.96 with Tsukuba. This implies that the increase
16 in the observations over Japan improves the capability of GOSAT to detect synoptic
17 variability in XCH₄.

18

19 **2.3 Ground-based surface CH₄ concentrations**

20 In order to see the relationship between the surface CH₄ concentration and the
21 enhancement of GOSAT XCH₄ over Japan, we analyzed the surface CH₄
22 concentrations observed at three ground-based monitoring stations in Japan, Cape
23 Ochi-ishi (COI, 43.16°N, 145.49°E), Ryori (RYO, 39.03°N, 141.82°E), and
24 Yonagunijima (YON, 24.47°N, 123.02°E). These site locations are shown in Fig. 1.
25 At all the stations, continuous measurements of atmospheric CH₄ are conducted.
26 Cape Ochi-ishi (COI) is a station operated by NIES, which is located at the east tip of
27 Hokkaido island (Tohjima et al., 2002). Ryori (RYO) is located inside the Japan
28 region defined in this study, where the monitoring of surface greenhouse gas
29 concentrations has been conducted by the Japan Meteorological Agency (JMA) as a
30 part of the Global Atmospheric Watch (GAW) program of the World Meteorological
31 Observation (WMO). RYO is on the west coast of the Japan main island, about 300
32 km north of Tsukuba and far away from direct influences of residential and industrial

1 pollutants. Yonagunijima (YON) is also one of JMA-operated GAW stations, which
2 is located far south of the Japan main island and east of ~110 km of Taiwan. The
3 details on RYO and YON are provided on the web page of WMO GAW World Data
4 Centre for Greenhouse Gases (WDCGG)
5 (<http://ds.data.jma.go.jp/gmd/wdcgg/introduction.html>).

6 The time-series of surface CH₄ concentrations at the three ground-based
7 stations are shown in Fig. 6, with their monthly means and long-term trends. Here we
8 analyzed the afternoon mean CH₄ (averaged hourly CH₄ over 12:00 - 15:00 local
9 time) from the respective data sets, assuming that the afternoon values are large-scale
10 representative. The observed CH₄ concentrations at all the sites show similar seasonal
11 cycles in timing. Seasonally the CH₄ values are low in July and August, and high in
12 winter to spring. In the winter, the westerly wind prevails and transports the CH₄-rich
13 air from the continent (mainly anthropogenic CH₄ emitted in East China) to Japan,
14 causing the rise of CH₄ concentrations. In the summer, the southeasterly wind is
15 dominant, bringing clean air to Japan from the Pacific Ocean, where is no major CH₄
16 source, so that the surface CH₄ concentrations become low.

17 In the summer of 2013, unseasonably high CH₄ concentrations were observed
18 at RYO with a sharp increase in the middle of August. The CH₄ concentrations at COI
19 started increasing earlier from its summer minimum than the previous year, 2012. At
20 YON, no significant CH₄ enhancement was seen in 2013 compared with the previous
21 years. Since no similar CH₄ change to RYO and COI was observed at YON, the
22 farthest southwestern island of Japan, this significant CH₄ enhancement event appears
23 to be spatially limited in the area around Japan main island and Hokkaido island. To
24 further examine the summer increase of surface CH₄ concentrations, we compared the
25 detrended CH₄ at RYO and COI for the two years of 2012 and 2013 (Fig. 7). The
26 timing and amplitude of seasonal cycles at RYO and COI overall agree well with each
27 other, except for the summer of 2013. In August and September of 2013, the
28 temporal variations of CH₄ at RYO and COI are different from those in the previous
29 year 2012 when the CH₄ concentrations were low over the summertime and started
30 rising at the end of September. In August 2013, the abrupt CH₄ increase by ~100 ppb
31 was observed at RYO, followed by COI with ~1 week delay. In September, the CH₄
32 at both sites lowered but stayed in the higher level than 2012. Given that the fact the
33 major CH₄ sources in East China, the sudden large increase of CH₄ in August 2013 is

1 probably caused by unseasonal transport of CH₄-rich air from the continent to Japan
2 though normally in August the wind with CH₄-low air from the Pacific Ocean is
3 prevailing over Japan.

4

5 **3 Model analysis**

6 The observational data analysis suggested that the atmospheric transport would be a
7 key factor of the extreme enhancement event of XCH₄ and surface CH₄ concentrations
8 in the summer of 2013 over Japan. To investigate how the inter-annually varying
9 atmospheric transport plays the role in the enhancement of XCH₄ and surface CH₄, we
10 conducted a forward model simulation using the global atmospheric transport model
11 of National Institute for Environmental Studies (NIES-TM) version 8.1i.

12 The NIES-TM has a horizontal resolution of 2.5°×2.5° with 32 vertical layers
13 (Belikov et al., 2013). The global wind fields used in this study were obtained from
14 the JMA Climate Data Assimilation System (JCDAS) (Onogi et al., 2007). The
15 planetary boundary layer height data are obtained from the European Centre for
16 Medium-Range Weather Forecasts (ECMWF) Interim Reanalysis dataset (Dee et al.,
17 2011). In order to examine the impact of time-varying atmospheric transport on the
18 seasonal cycles of atmospheric CH₄ and XCH₄ fields, the CH₄ emissions averaged
19 over 2009-2010 were repeatedly used during the entire model simulation period for
20 2009-2013. The CH₄ emissions comprise anthropogenic fluxes and natural fluxes.
21 The anthropogenic fluxes are from the Emissions Database for Global Atmospheric
22 Research (EDGAR) inventory, v4.2 FT2010 (<http://edgar.jrc.ec.europa.eu/>). The natural
23 CH₄ fluxes are biomass burning from Global Fire Emissions Database (GFED) v3.1
24 (van der Werf et al., 2010), wetland, rice paddy emissions and soil sinks from the
25 Vegetation Integrative Simulator for Trace gases (VISIT) (Ito and Inatomi, 2012), and
26 termites (Fung et al., 1991). Except the termites CH₄ emission, all the natural fluxes
27 are seasonal. We used the modeled methane loss and climatological OH fields
28 provided for a model inter-comparison project “TransCom-CH₄“ (Patra et al., 2011).

29 Figure 8 shows the simulated surface CH₄ concentration and XCH₄ fields for
30 August and September in 2012 and 2013. As a common feature, the high levels of
31 XCH₄ and surface CH₄ are found over East China, reflecting the spatial distribution of

1 the strong anthropogenic emissions around Beijing and Shanghai and biogenic CH₄
2 sources from rice cultivation in the southeastern China.

3 Different patterns are found in the XCH₄ and surface CH₄ fields between 2012
4 and 2013. In August 2012, both levels of XCH₄ and surface CH₄ over Japan are as
5 low as those over the Pacific oceans. In August 2013, higher concentrations of surface
6 CH₄ extended from the northeastern China and the Korean peninsula to the Japan
7 islands. The surface CH₄ concentration level in 2013 over Japan is increased by 40-
8 60 ppb from the level in 2012. The XCH₄ values over Japan are also enhanced by
9 ~20 ppb while the XCH₄ values over the southeastern China are lowered compared to
10 the 2012 level. The lower concentrations in August 2013 over the southeastern China
11 than 2012 indicate that the northward wind along the coast was so fast that CH₄ was
12 not much accumulated over the CH₄ source area in the southeastern China, but
13 transported away to the north. As a result, the areas of the highest levels of CH₄ and
14 XCH₄ shifted to the northeast, from the southeast China. In September 2013, XCH₄
15 level over the southeastern China is higher than August, but still lower than the level
16 of September 2012. Also the XCH₄ over Japan remains higher level than that of 2012.
17 The surface CH₄ concentration pattern in September 2013 is almost similar to the one
18 in 2012, but slightly higher values are found over Japan.

19 Figure 9 shows the time-series of modeled XCH₄ for GOSAT and TCCON,
20 compared with the observations. For GOSAT, the modeled XCH₄ co-located with the
21 GOSAT observations are sampled and averaged for comparison. The modeled XCH₄
22 produce the enhancement in summer 2013, in phase with the observations (Fig. 9).
23 Overall the temporal variations of modeled XCH₄ are correlated with the observations
24 ($r = 0.50 \sim 0.72$). These correlation coefficient values exceeded the 95% significance
25 level. The seasonal cycles of modeled XCH₄ for GOSAT and TCCON are in good
26 agreement with the observations, while the modeled GOSAT XCH₄ show less short-
27 term variability than the observations. The modeled surface CH₄ concentrations for
28 the three Japanese sites, COI, RYO, and YON, are shown in Fig. 10. Though the
29 modeled seasonal amplitude is slightly smaller than the observed, the modeled CH₄
30 overall capture the observed synoptic variations, as well as the abrupt increase in
31 August 2013 at COI and RYO. The model was run with cyclo-stationary surface CH₄
32 fluxes, which are seasonally varying but not inter-annually. Inside the model, only
33 the transport field is varying inter-annually. The model-observation comparison thus

1 provides supporting evidence that anomalous wind field in 2013 plays a key role in
2 the large XCH₄ event in 2013.

3

4 **4 Discussions**

5 **4.1 Characteristics of Atmospheric Circulation in the summer of 2013**

6 Forward modeling gives us insights into the contribution of atmospheric transport on
7 the enhancement of XCH₄ and surface CH₄ concentration in the summer of 2013 over
8 Japan. Here we examine the 2013 summertime atmospheric transport over the
9 northeastern Asia.

10 Japan's summer climate is governed by the Pacific High (a lower-level high-
11 pressure system) and the Tibetan High (an upper-level high-pressure system). These
12 pressure systems were reported to have been enhanced during July and August 2013
13 (Tokyo Climate Center News No.34 Autumn 2013, available at
14 <http://ds.data.jma.go.jp/tcc/tcc/news>). The Pacific High continued to expand westward
15 and largely developed over the western part of Japanese islands including Okinawa.
16 The Tibetan High expanded to the Japan main island in line with the northward
17 meandering of upper-level westerly winds (the subtropical jet stream). The enhanced
18 atmospheric transport from East China to Japan was probably attributed to those
19 anomalously developed high-pressure systems.

20 To see how the 2013 summertime atmospheric transport differs from the mean
21 transport pattern, Fig. 11 shows the wind fields at the surface and at 850 hPa pressure
22 level, from the JCDAS wind fields of August and September in 2013 over East Asia,
23 compared with those of the mean wind fields for the five years of 2009-2013.

24 At the surface level (Fig. 11a), the mean wind field clearly shows that, in
25 August the southeasterly wind from the Pacific Ocean prevails due to the
26 development of the Pacific High. In September the wind from the continent to Japan
27 start blowing as the Pacific High is retiring. In August 2013, as the Pacific High
28 expanded westward, the air moved northward along the coast of China, turned around
29 the Korean Peninsula, and flowed to Japan. This wind pattern suggests that the CH₄-
30 rich air was transported from East China to Japan in 2013, while the clean air is
31 normally transported from the Pacific Ocean. In September 2013, over the Pacific

1 Ocean, south of the Japan main island, easterly wind was still stronger than the
2 normal, but the wind pattern over Japan was almost back to the normal, which can be
3 characterized as a weak convergence of westerly wind from the continent and easterly
4 wind from the Pacific Ocean. This nearly normal wind pattern over northern Japan
5 would lower the CH₄ concentrations at the surface level as observed at RYO and COI.

6 At the 850 hPa level (Fig. 11b), it is notable that, in August 2013 the air
7 moved over the East China along the coast and turned around the Korean peninsula
8 sharply to the Japan islands. The anomalous westerly winds were stronger in the
9 upper levels than near the surface. Given the major CH₄ source distributions in East
10 Asia, the strong northward air flow along the coast could reduce local CH₄
11 accumulation, but transport the CH₄-rich air effectively to the north and then to Japan
12 as turning around the Korean peninsula. In September 2013, the wind speed over
13 Japan was much lower than August, but wind still blows westerly from the continent
14 to Japan. This slower westerly air flow could maintain the higher level of XCH₄ over
15 Japan during the September of 2013.

16 The wind patterns we examined above shows us how the atmospheric
17 transport field in 2013 differed from a climatological field on a regional-scale. In
18 order to narrow down the origins and the upstream patterns of the air masses to the
19 Japanese TCCON sites, we conducted back trajectory analysis using the Stochastic
20 Time-Inverted Lagrangian Transport (STILT) model (Lin et al., 2003), driven by
21 Global Data Assimilation System (GDAS) meteorology (1° × 1°). To obtain the
22 monthly mean features of the upstream, we released 100 particles from the height of
23 1500 m (approximately ~850 hPa) at Saga and Tsukuba, at every 12:00 noon local
24 time (= 3 UT) and traveled backward for 10-days. Every 30 minutes, the number of
25 particles was counted by a 1° × 1° air column and the total number of particles over
26 the 10 day duration was divided by the maximum number per column. Thus, we
27 obtained a normalized daily upstream pattern and averaged them over a month.
28 Figure 12 shows the monthly normalized trajectories for August and September in
29 2012 and 2013. There are distinct differences in the upstream patterns between 2012
30 and 2013. The patterns of the summer of 2012 are almost like climatological; in
31 August, the wind flows dominantly from the Pacific to the Japan, in September the
32 dominant wind direction is in transition; from southeasterly wind (from the Pacific)
33 to northwesterly wind (from the continent). On the other hand, in August 2013 the air

1 masses reached the Japanese TCCON sites from the west, after traveling over the
2 coastal side of East China. In September 2013, the westerly wind from the continent is
3 still dominant, especially for Saga. This backtrajectory result supports that the
4 anomalous wind field in the summer of 2013 brought the CH₄-rich air from China to
5 Japan, resulting in the high XCH₄ observed at the two Japanese TCCON sites and also
6 by GOSAT over Japan.

7

8 **4.2 Other possible factors**

9 Although we suggest that the atmospheric transport field probably attributes to the
10 enhancement of XCH₄ and CH₄ concentration observed in the summer of 2013, we
11 cannot entirely rule out other possible factors. Here we discuss two factors. One is
12 the surface emission changes. Though the temporal variations in XCH₄ do not
13 necessarily correlate with the surface emissions (e.g., Bloom et al., 2010), the surface
14 emission change is potential to impact on the change in XCH₄. The second is the
15 contribution of stratospheric methane.

16 The CH₄ emissions from rice cultivations and wetland in Southern China
17 might be enhanced under the hot summer condition in 2013. East Asia around China
18 experienced a hotter summer monsoon season (June-September) by more than 1 °C
19 than the season normal (Tokyo Climate Center News No.34 Autumn 2013), while less
20 than 60 % of the normal precipitation in eastern China was reported. A hot weather
21 condition increases the CH₄ emissions through the enhancement of photosynthesis
22 and methanogenic activity in inundated grounds such as wetlands and rice paddies;
23 while a dry condition reduces the CH₄ emissions from wetlands as the water table
24 levels in the ground become low. Thus, the hot and dry weather conditions have
25 opposite effects on the CH₄ emissions from wetlands. The time delay in the
26 correlation between CH₄ emissions and climate anomalies should be considered as the
27 groundwater plays an important role in wetland CH₄ emissions. Furthermore, since
28 rice cultivation is human-managed, multiple controlling factors on CH₄ emissions
29 from rice paddies should be considered. A further investigation of wetland and rice
30 CH₄ emission changes responding to the climate anomaly in East Asia is needed.

31 Another possibility is the contribution of stratospheric methane. Saad et al.
32 (2014) presented the analysis that the stratospheric methane causes short-term

1 fractionations in total column averaged CH₄ observed at several TCCON sites. The
2 contribution of stratospheric methane to the anomaly in summer 2013 is supposed to
3 be minor or less influential. Firstly the surface CH₄ concentrations at COI and RYO
4 increased in August 2013 when the XCH₄ anomaly occurred, suggesting the major
5 contributor on the anomaly is in the troposphere. Secondly, the order of the
6 stratospheric methane fractionation is smaller than ~3 ppb, which would not be
7 enough to produce the anomaly of an order of ~20 ppb.

8

9 **5 Conclusion**

10 In this study, we have examined the extremely high XCH₄ event over Northeast Asia
11 observed by GOSAT in August and September, 2013. Similar XCH₄ enhancements in
12 amplitude and timing were observed at the two Japanese TCCON sites, Tsukuba and
13 Saga. Furthermore, during the same period, the ground-based atmospheric CH₄
14 monitoring sites of Ryori and Ochi-ishi located in the northern part of Japan observed
15 the higher levels of surface CH₄. In particular, surface CH₄ concentrations at Ryori
16 showed the rapid increase in the middle of August 2013.

17 Our model analysis indicates that the significant enhancement of XCH₄ and
18 surface CH₄ are mainly attributed to the anomalous atmospheric pressure patterns of
19 Pacific High and Tibetan High over East Asia during the summer of 2013. The CH₄-
20 rich air was effectively-transported to Japan from the major CH₄ source area in East
21 China. The model analysis also indicates that the XCH₄ enhancement occurred in a
22 limited area of the northeastern China to the Japan main island. The two Japanese
23 TCCON sites, ~1,000 km apart from each other, happened to be located along the
24 anomalously CH₄-rich air flow from the Eurasian continent and coincidentally
25 observed the extreme increase of XCH₄. The GOSAT with three-day recurrence
26 successfully observed the high XCH₄ event. This data analysis study demonstrates the
27 capability of GOSAT to monitor the synoptic-scale XCH₄ event in the association
28 with the high-pressure system anomalies. The GOSAT capability to detect synoptic
29 variations could be helpful to quantify the relative contribution of atmospheric
30 transport, leading to better estimation of regional CH₄ fluxes.

31

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7

8

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23

1 **Figure Captions**

2 Figure 1. Two regions considered in this study: Northeastern China-Korea (115°E -
3 130°E, 30°N - 40°N, gray-shaded) and Japan (130°E - 145°E, 30°N - 40°N, blue-
4 shaded). The locations of the Saga and Tsukuba TCCON stations are marked by
5 closed circles. The open circles are indicated the locations of the surface monitoring
6 stations around Japan, Cape Ochi-ishi (COI), Ryori (RYO), and Yonagunijima
7 (YON).

8
9 Figure 2. Temporal variations of GOSAT XCH₄ over the two regions of Northeast
10 Asia: (a) Northeastern China – Korea (115°E - 130°E, 30°N - 40°N), and (b) Japan
11 (130°E - 145°E, 30°N - 40°N). GOSAT XCH₄ data are shown in grey dot. The
12 monthly means are plotted in red solid circle and line, whereas monthly means in
13 open circles indicate less than two retrievals available per month. Blue lines indicate
14 the long-term trends. The histogram in the bottom show the number of GOSAT XCH₄
15 data per month.

16
17 Figure 3. Locations of GOSAT soundings with the surface elevations, in the two
18 regions considered in this study. The locations of three capital cities, Tokyo, Seoul
19 and Beijing are also shown in black markers. (a) All soundings of GOSAT data used
20 for 2009-2014. (b) Same with (a) but in August and September 2012. (c) Same with
21 (a) but in August and September 2013.

22
23 Figure 4. Temporal variations of TCCON XCH₄ at (a) Saga (130.29°E, 33.24°N)
24 and (b) Tsukuba (140.12°E, 36.05°N), Japan. TCCON XCH₄ data are shown in grey
25 dot, daily means in green dots. The monthly means are plotted in red solid circle and
26 line, whereas monthly means in open circles indicate less than two observation days
27 per month. Blue lines indicate the long-term trends. The histograms at the bottom
28 show the number of observation day per month.

29
30 Figure 5. (a) Detrended XCH₄ for 2012 to 2013 at Saga and Tsukuba, Japan and
31 GOSAT over Japan. (b) Same with (a) but also minus mean seasonal cycles. The

1 monthly means of the individual XCH₄ time series are shown in solid lines and circles.
2 The open circles for TCCON indicate that observation days in a month are less than
3 two days. The discontinuity of GOSAT in July 2012 indicates no GOSAT XCH₄
4 retrieval. Long-term components in individual XCH₄ time series are removed by low
5 pass digital filter of cutoff frequency of two years. Mean seasonal cycles are
6 composed of two harmonics of year and a half year cycles. August and September of
7 both 2012 and 2013 are highlighted.

8

9 Figure 6. Temporal variations of atmospheric CH₄ concentrations observed at the
10 ground-based monitoring sites around Japan, (a) Cape Ochi-ishi (COI, 43.16°N,
11 145.49°E), (b) Ryori (RYO, 39.03°N, 141.82°E), and (c) Yonagunijima (YON,
12 24.47°N, 123.02°E). The site locations are shown in Figure 1. Afternoon means of
13 hourly CH₄ concentrations are shown in grey lines. The monthly means are plotted in
14 red solid circle and line. Blue lines indicate the long-term trends.

15

16 Figure 7. Detrended CH₄ for 2012 to 2013 at Ryori (RYO) and Cape Ochi-ishi (COI)
17 in Japan. Long-term components in individual CH₄ time series are removed by low
18 pass digital filter of cutoff frequency of two years. August and September of both
19 2012 and 2013 are highlighted.

20

21 Figure 8. Spatial distribution of monthly mean modelled (a) CH₄ and (b) XCH₄ in
22 August and September of 2012 and 2013.

23

24 Figure 9. Model simulated XCH₄ time-series in comparison with the observed
25 GOSAT XCH₄ over the two target regions of (a) Northeastern China-Korea and (b)
26 Japan, and with the observed TCCON XCH₄ at (c) Saga and (d) Tsukuba. For
27 GOSAT, modeled XCH₄ outputs are sampled at corresponding model grids and
28 averaged by region. August and September of both 2012 and 2013 are highlighted.

29

30 Figure 10. Model simulated CH₄ time-series in comparison with the observed CH₄ at
31 (a) Cape Ochi-ishi (COI, 43.16°N, 145.49°E), (b) Ryori (RYO, 39.03°N, 141.82°E),

1 and (c) Yonagunijima (YON, 24.47°N, 123.02°E). August and September of both
2 2012 and 2013 are highlighted.

3

4 Figure 11. Monthly mean wind fields of August and September at (a) surface and (b)
5 850hPa. The left panels are the wind fields averaged over the five years of 2009-2013,
6 and the right panels are the monthly mean wind fields of the year 2013.

7

8 Figure 12. Monthly mean ten-day backward trajectories from (a) Saga and (b)
9 Tsukuba at 12:00 noon local time (= 3:00 UT). The trajectories started at an altitude
10 of 1500 m (approximately 850 hPa). 100 particles are released every day for a month.
11 To normalize the number density of particles, the particles passed at each 1° x 1° grid
12 air column are counted, and the total numbers are divided by the maximum number
13 per grid.

Figure 1

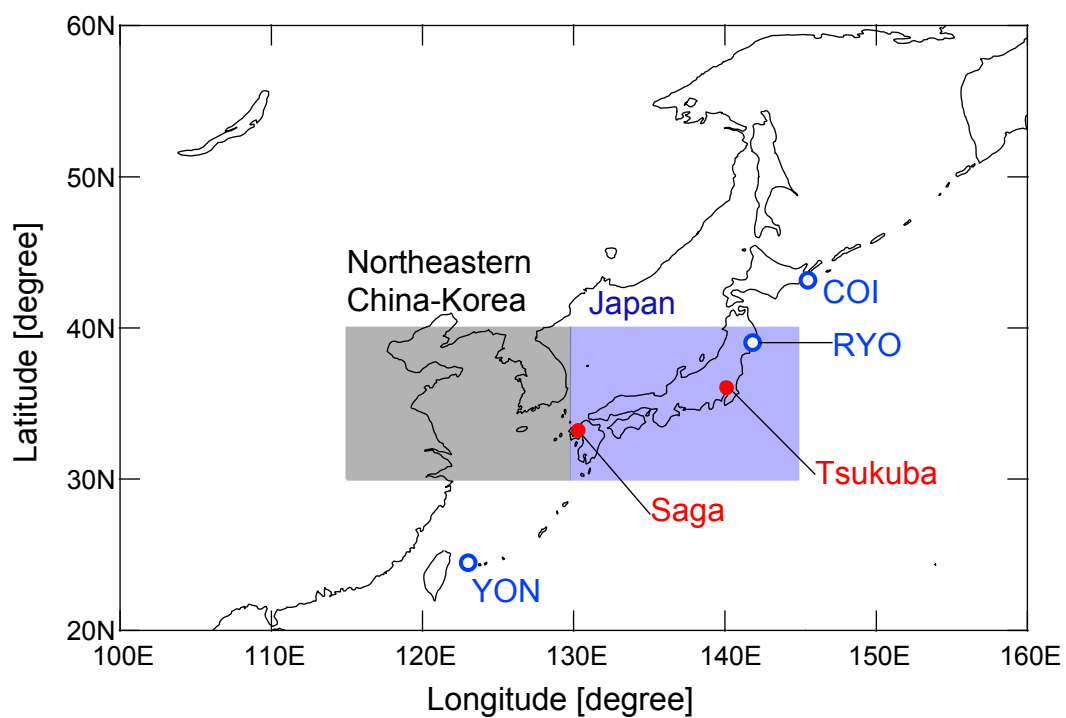


Figure 1. Two regions considered in this study: Northeastern China-Korea (115°E -130°E, 30°N -40°N, gray-shaded) and Japan (130°E-145°E, 30°N-40°N, blue-shaded). The locations of the Saga and Tsukuba TCCON stations are marked by closed circles. The open circles are indicated the locations of the surface monitoring stations around Japan, Cape Ochi-ishi (COI), Ryori (RYO), and Yonagunijima (YON).

Figure 2

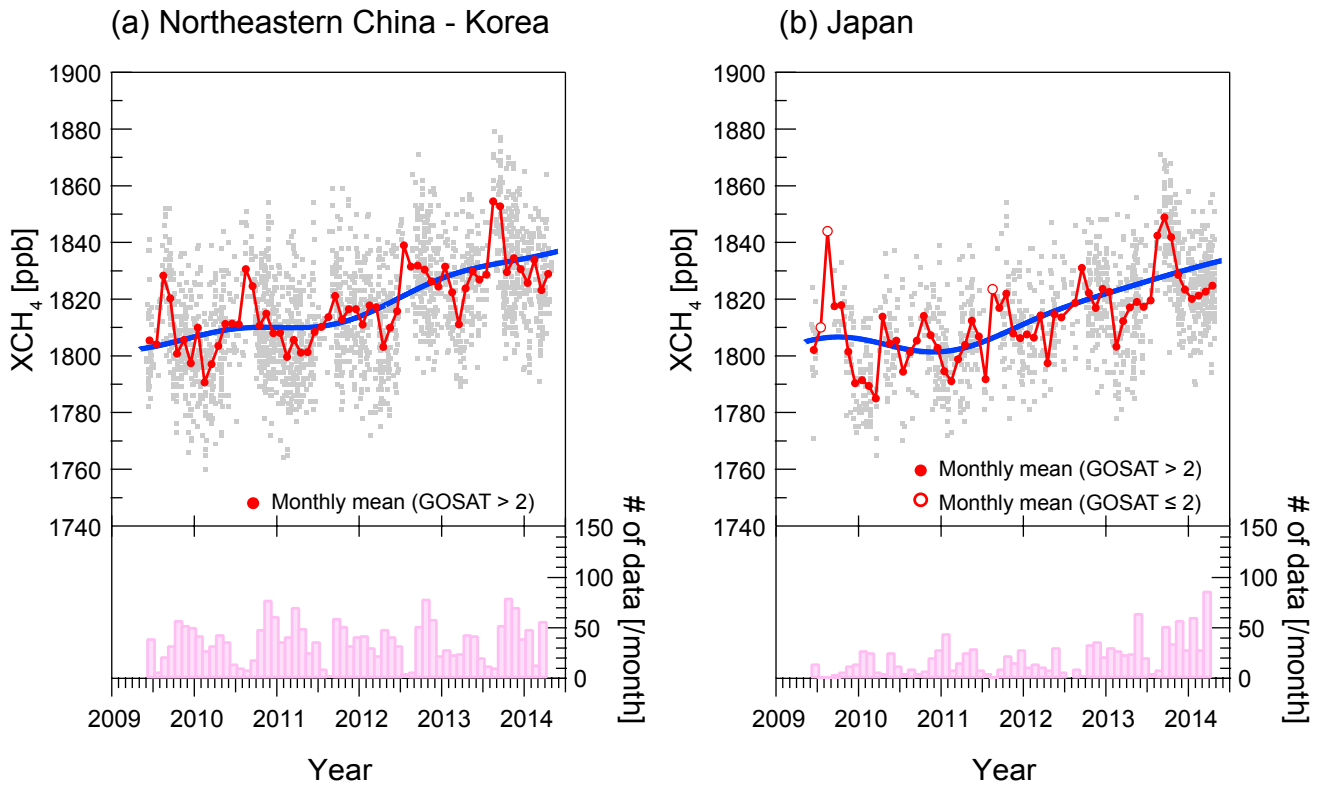


Figure 2. Temporal variations of GOSAT XCH₄ over the two regions of Northeast Asia: (a) Northeastern China – Korea (115°E-130°E, 30°N-40°N), and (b) Japan (130°E-145°E, 30°N -40°N). GOSAT XCH₄ data are shown in grey dot. The monthly means are plotted in red solid circle and line, whereas monthly means in open circles indicate less than two retrievals available per month. Blue lines indicate the long-term trends. The histogram in the bottom show the number of GOSAT XCH₄ data per month.

Figure 3

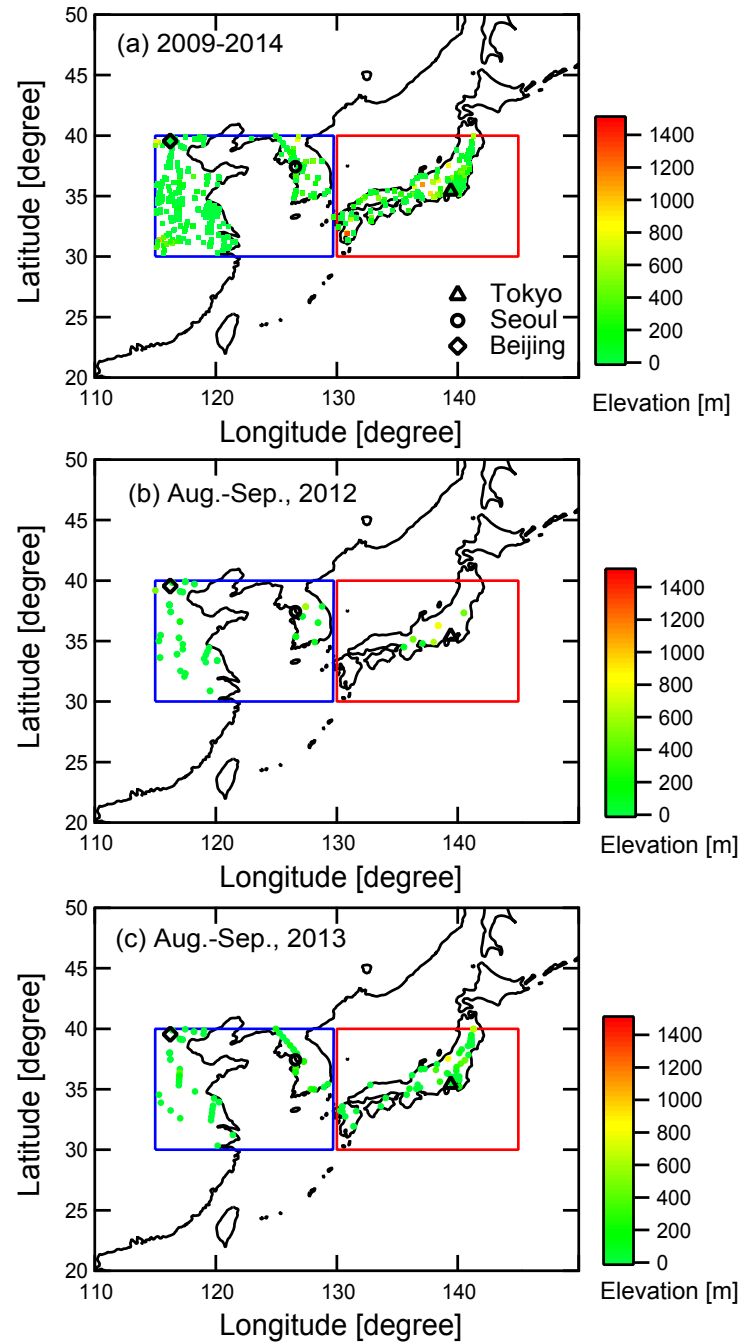


Figure 3. Locations of GOSAT soundings with the surface elevations, in the two regions considered in this study. The locations of three capital cities, Tokyo, Seoul and Beijing are also shown in black markers. (a) All soundings of GOSAT data used for 2009-2014. (b) Same with (a) but in August and September 2012. (c) Same with (a) but in August and September 2013.

Figure 4

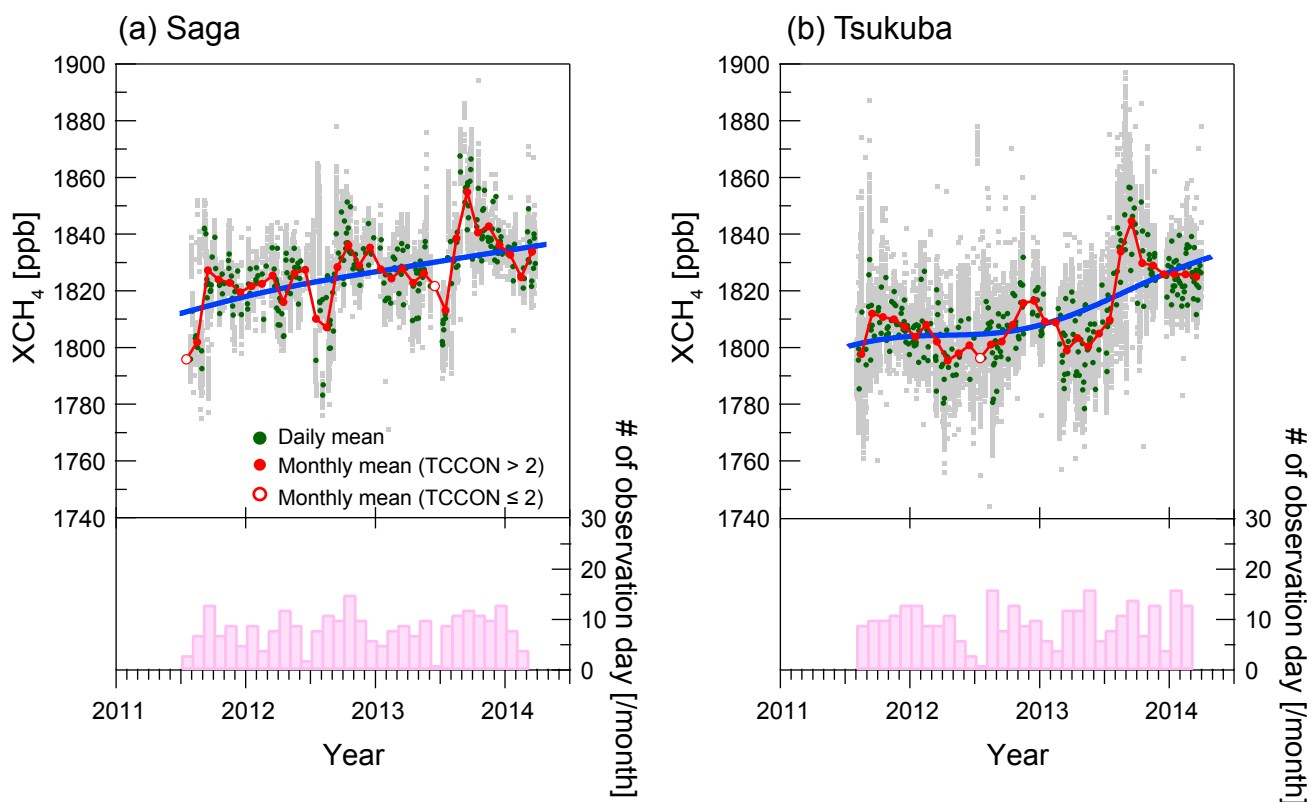


Figure 4. Temporal variations of TCCON XCH₄ at (a) Saga (130.29°E, 33.24°N) and (b) Tsukuba (140.12°E, 36.05°N), Japan. TCCON XCH₄ data are shown in grey dot, daily means in green dots. The monthly means are plotted in red solid circle and line, whereas monthly means in open circles indicate less than two observation days per month. Blue lines indicate the long-term trends. The histograms at the bottom show the number of observation day per month.

Figure 5.

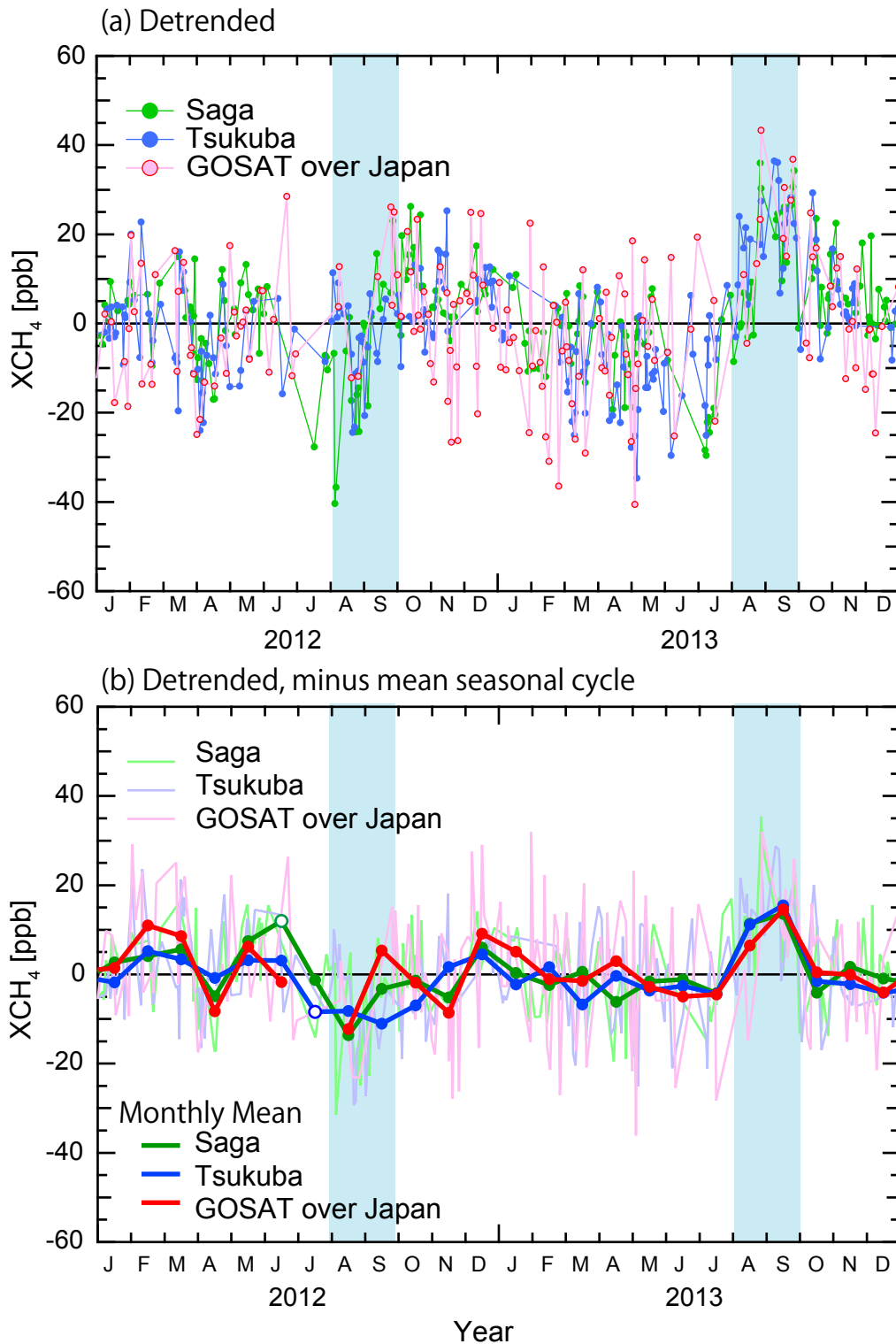


Figure 5. (a) Detrended XCH₄ for 2012 to 2013 at Saga and Tsukuba, Japan and GOSAT over Japan. (b) Same with (a) but also minus mean seasonal cycles. The monthly means of the individual XCH₄ time series are shown in solid lines and circles. The open circles for TCCON indicate that observation days in a month are less than two days. The discontinuity of GOSAT in July 2012 indicates no GOSAT XCH₄ retrieval. Long-term components in individual XCH₄ time series are removed by low pass digital filter of cutoff frequency of two years. Mean seasonal cycles are composed of two harmonics of year and a half year cycles. August and September of both 2012 and 2013 are highlighted.

Figure 6

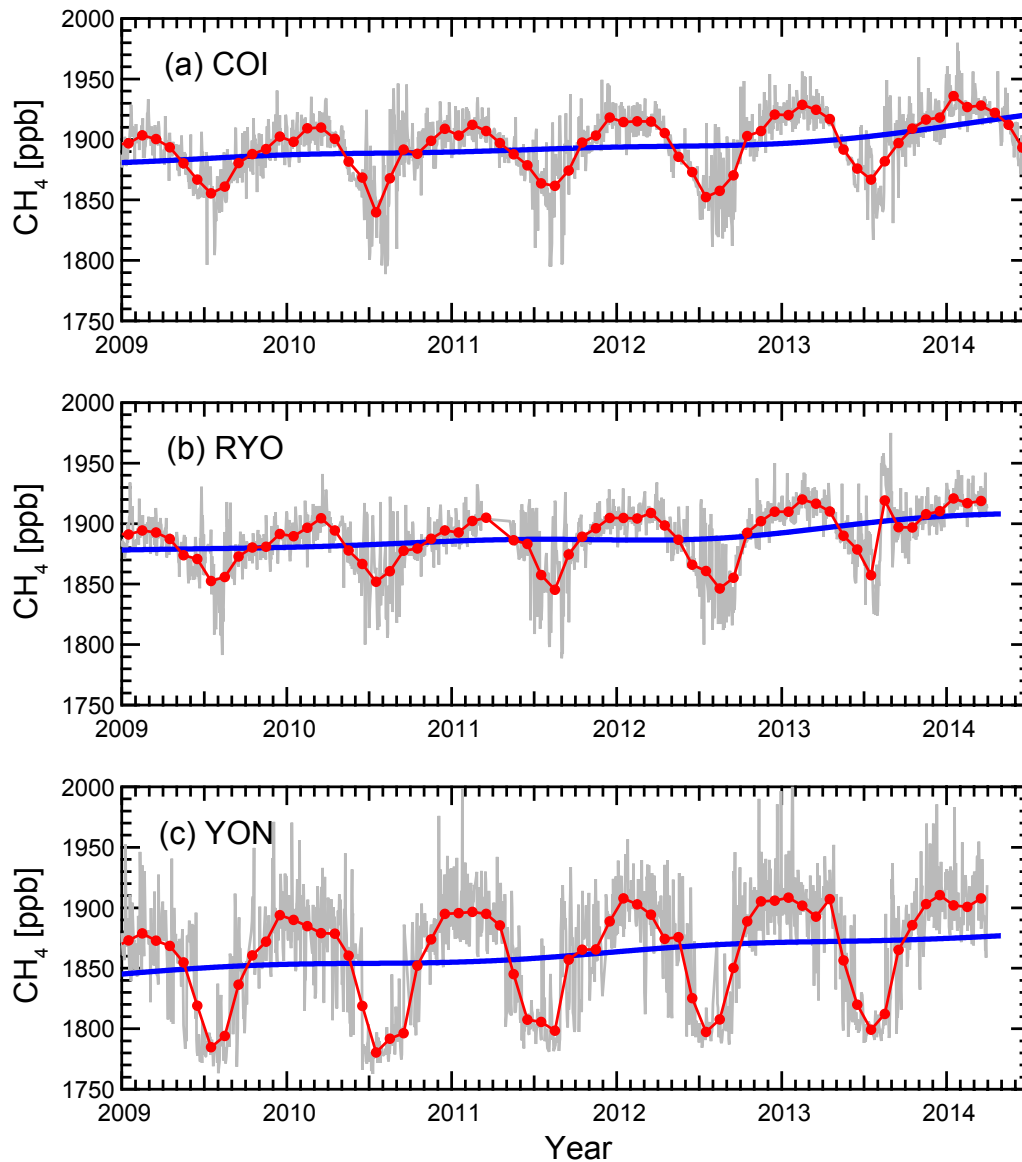


Figure 6. Temporal variations of atmospheric CH₄ concentrations observed at the ground-based monitoring sites around Japan, (a) Cape Ochi-ishi (COI, 43.16°N, 145.49°E), (b) Ryori (RYO, 39.03°N, 141.82°E), and (c) Yonagunijima (YON, 24.47°N, 123.02°E). The site locations are shown in Figure 1. Afternoon means of hourly CH₄ concentrations are shown in grey lines. The monthly means are plotted in red solid circle and line. Blue lines indicate the long-term trends.

Figure 7

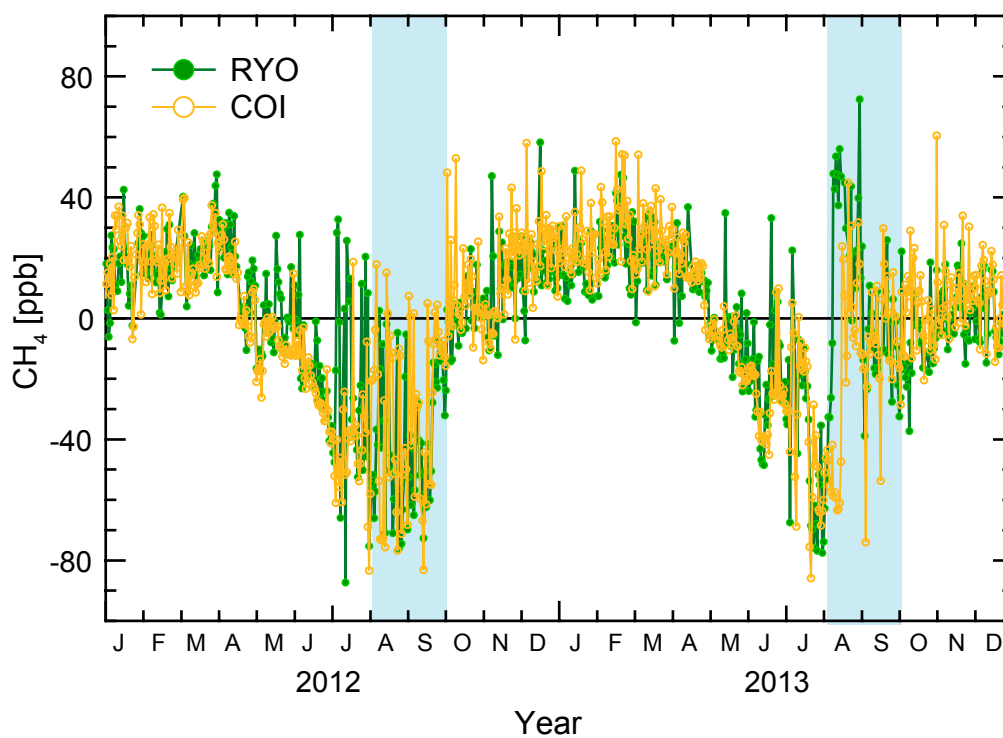
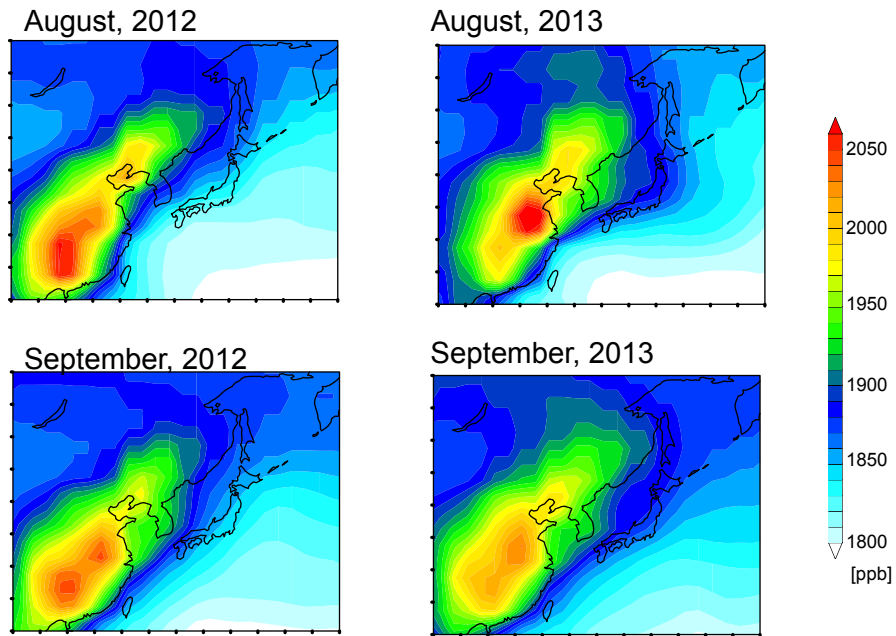


Figure 7. Detrended CH₄ for 2012 to 2013 at Ryori (RYO) and Cape Ochi-ishi (COI) in Japan. Long-term components in individual CH₄ time series are removed by low pass digital filter of cutoff frequency of two years. August and September of both 2012 and 2013 are highlighted.

Figure 8

(a) Surface CH_4



(b) XCH_4

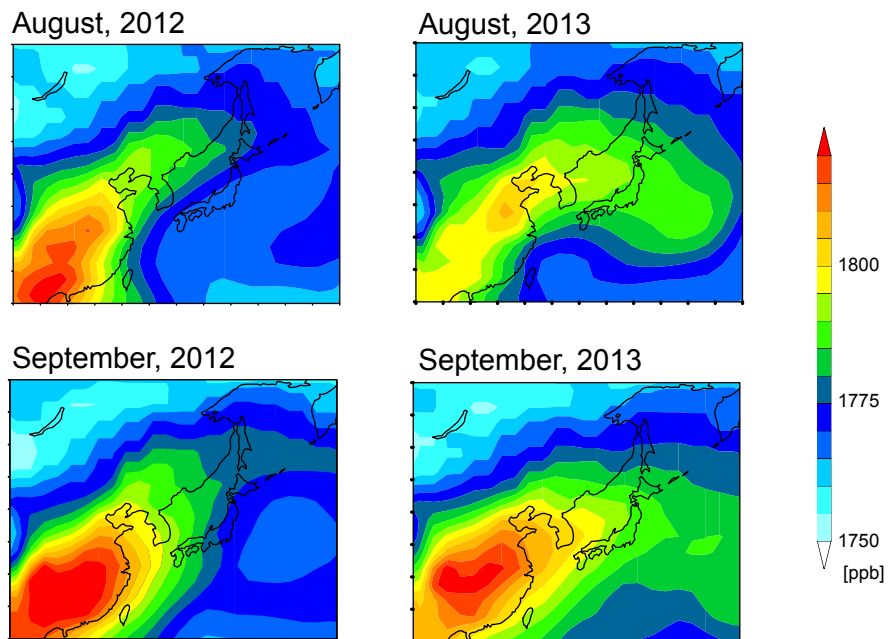


Figure 8. Spatial distribution of monthly mean modelled (a) CH_4 and (b) XCH_4 in August and September of 2012 and 2013.

Figure 9.

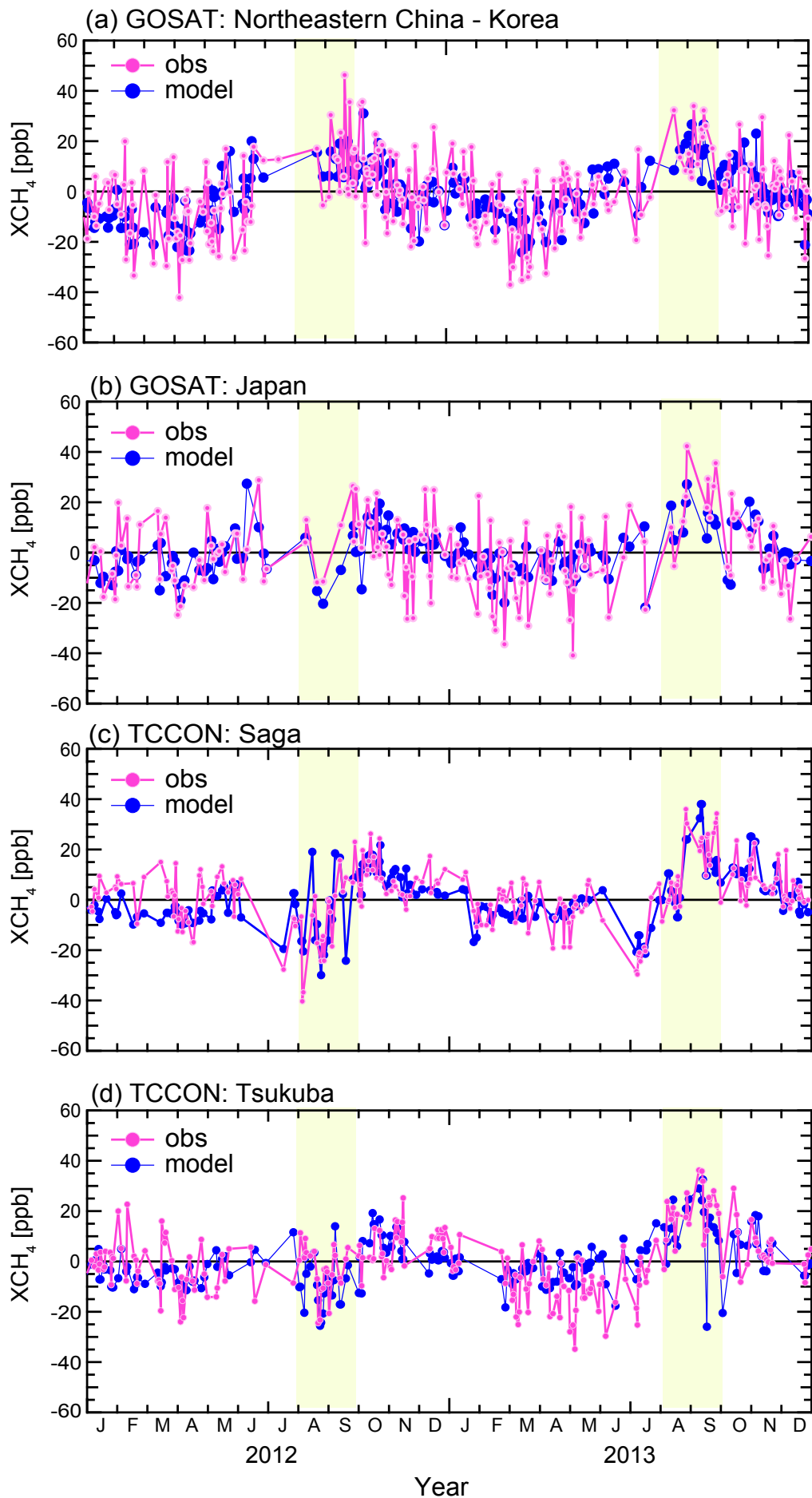


Figure 9. Model simulated XCH_4 time-series in comparison with the observed GOSAT XCH_4 over the two target regions of (a) Northeastern China-Korea and (b) Japan, and with the observed TCCON XCH_4 at (c) Saga and (d) Tsukuba. For GOSAT, modeled XCH_4 outputs are sampled at corresponding model grids and averaged by region. August and September of both 2012 and 2013 are highlighted.

Figure 10

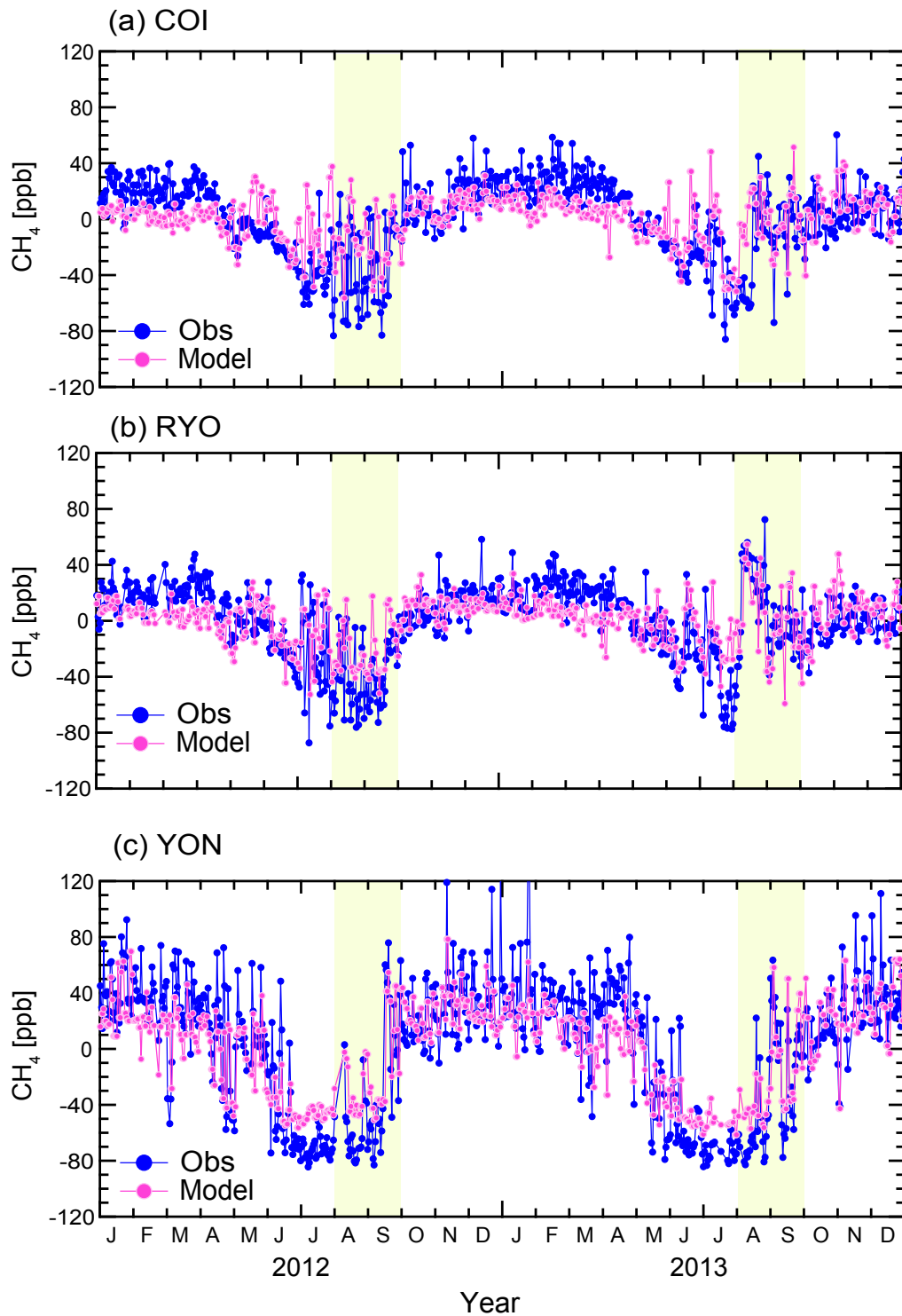
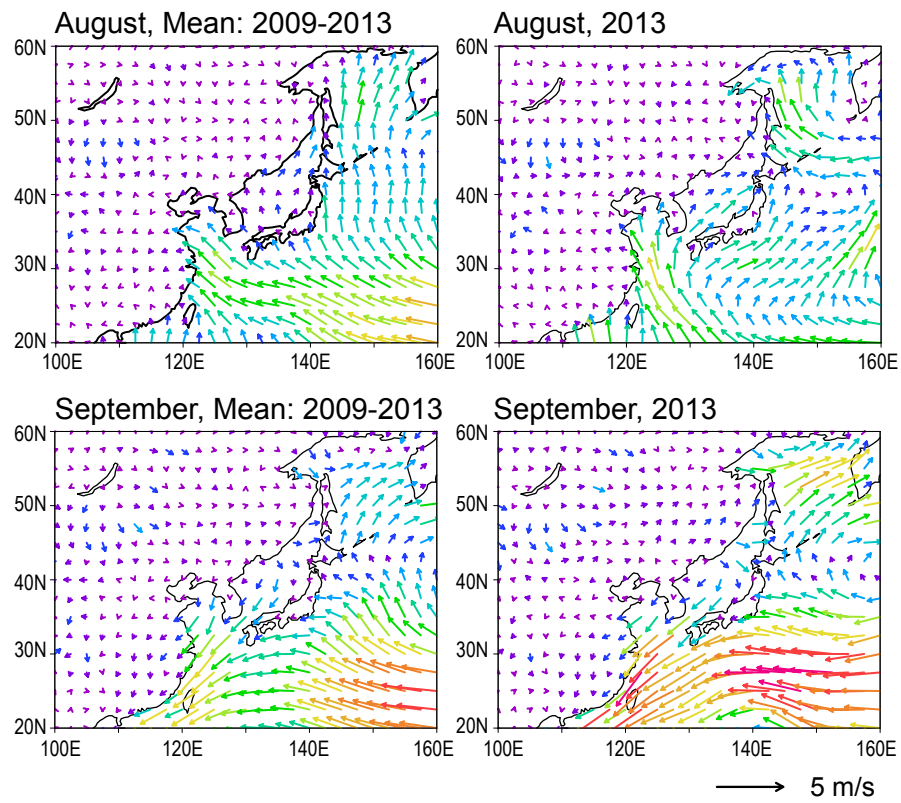


Figure 10. Model simulated CH₄ time-series in comparison with the observed CH₄ at (a) Cape Ochi-ishi (COI, 43.16°N, 145.49°E), (b) Ryori (RYO (39.03°N, 141.82°E), and (c) Yonagunijima (YON, 24.47°N, 123.02°E). August and September of both 2012 and 2013 are highlighted.

Figure 11

(a) Surface



(b) 850 hPa

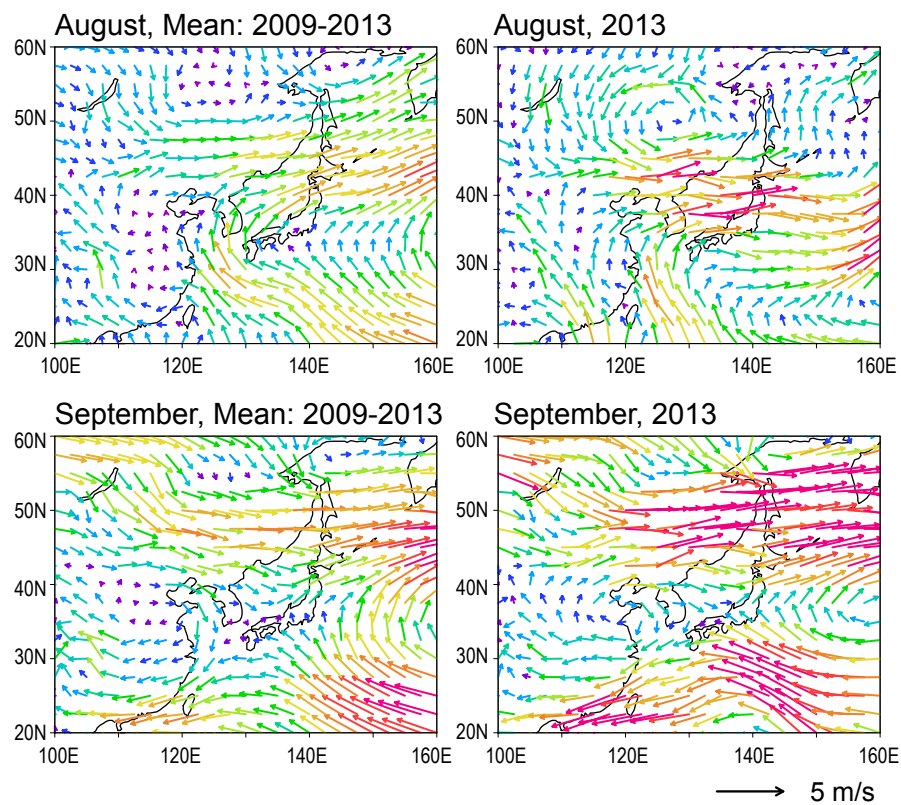


Figure 11. Monthly mean wind fields of August and September at (a) surface and (b) 850hPa. The left panels are the wind fields averaged over the five years of 2009-2013, and the right panels are the monthly mean wind fields of the year 2013.

Figure 12.

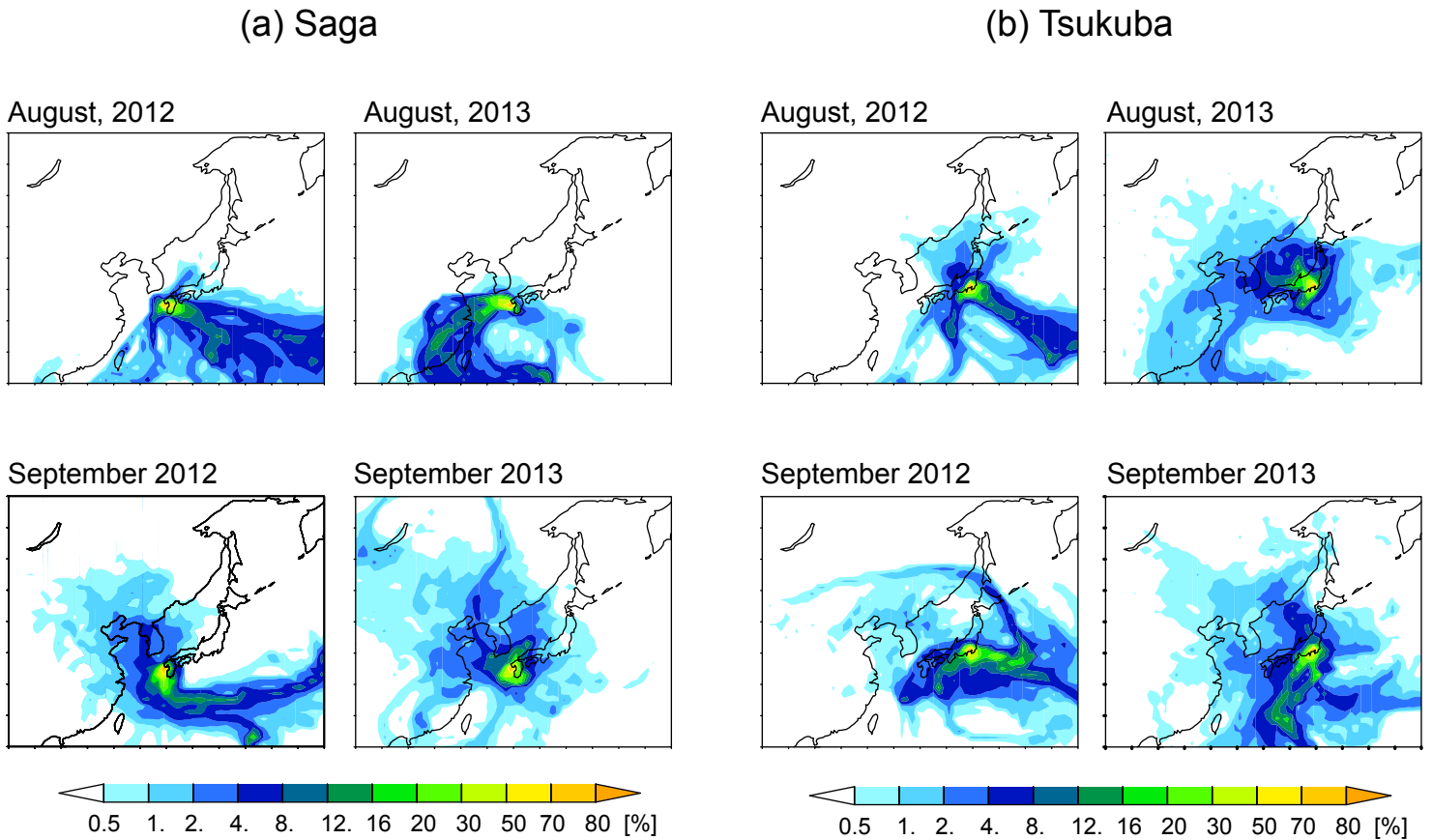


Figure 12. Monthly mean ten-day backward trajectories from (a) Saga and (b) Tsukuba at 12:00 noon local time (= 3:00 UT). The trajectories started at an altitude of 1500 m (approximately 850 hPa). 100 particles are released every day for a month. To normalize the number density of particles, the particles passed at each $1^\circ \times 1^\circ$ grid air column are counted, and the total numbers are divided by the maximum number per grid.