Reply to reviewer 1

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The authors would like to thank Reviewer 1 for the helpful comments. We have addressed the issues raised by the reviewer in the following. The review comments are copied in red and our responses are in black normal font and changes in black bold font below. The references and figures which have been added/modified in the manuscript are attached at the end.

Ishizawa et al.'s manuscript identify anomalously high GOSAT observations over Japan in summer 2013. They present surface observations (total column and in situ) that seem to corroborate these GOSAT observations and use model simulations to interrogate the source of the methane anomaly. The manuscript is fairly well written. However, I have some serious concerns with the manuscript. In particular, the authors need to account for the seasonal cycle and topography before the data can be used to answer their questions.

## 2 Major comments:

### 2.1 Source of the anomaly

The source attribution for this anomaly is severely lacking. The wind patterns do not seem to fit with their discussion. It appears that only the 850 hPa winds in August 2013 would have actually brought high-CH4 airmasses from China (and only to the northern part of Japan). The only thing that seems to really stand out in the rest of the panels is the flow from the Pacific towards southern Japan, why would Saga have a large anomaly from this air? Hysplit runs would be more convincing for showing the sources of the airmasses. As for the CTM results, does the CTM capture the duration of this anomaly? What does a 2012 and 2013 timeseries of XCH4 in the Japan region look like? Does the anomaly show up? What about the simulated surface sites?

Following the reviewer’s suggestions and questions, firstly we conducted the backward trajectory calculation to see where air traveled before reaching the two Japanese TCCON sites. These results have been included in the paper **(Fig.12)**. Secondly, the model-observation comparisons of $XCH_4$ and surface $CH_4$ time-series have been added in the manuscript **(Figs. 9 and 10)** to show how well the CTM captures the anomaly of $XCH_4$.

We employed the STILT modeling system that has been developed based on HYSPLIT coupling with a Lagrangian dispersion component. To see the upstream feature of the summer months, August and September, 100 particles were released from the height of 1500 m of the TCCON sites at every 12:00 noon local time ( = 3 UT). The trajectory results have included in Sec 4.1 (**Fig. 12)** after the discussion of wind patterns **(now Fig. 11)**. There are distinct differences in the back trajectory between 2012 and 2013. The trajectory pattern in summer 2012 is very climatological; in August, the wind from the Pacific to the Japan prevails, in September the dominant wind is in a transition from southeasterly wind (from the Pacific) to northwesterly wind (from the continent). On the other hand, in August 2013, the air masses reach the Japanese TCCON sites, after traveling over the coastal side of East China. In September 2013, the westerly wind from the continent is still influential, especially to Saga. These back trajectory results visualize that, the anomalous wind field in summer 2013 brought the $CH_4$-rich air from China to Japan, resulting in the high $XCH_4$ observed at the two Japanese TCCON sites and also by GOSAT over Japan.

The CTM simulation results of $XCH_4$ have been included in the manuscript (**Fig. 9**), compared with the observations. For GOSAT, the modeled $XCH_4$ values co-located with the GOSAT observations are averaged for comparison. The model simulations are in agreement with the observations, of which correlation coefficients (r) are 0.50-0.72. These correlation coefficient values exceeded the 95% significance level. Furthermore, the model simulations produced the enhancements of $XCH_4$ in summer 2013. The model was run with cyclo-stationary surface $CH_4$ fluxes, which are seasonally varying but not inter-annually. Inside the model, only the transport field is varying inter-annually. The model result thus provides supporting evidence that anomalous wind field in 2013 plays a key role in the large $XCH_4$ event in 2013.

The graphs of the modeled surface $CH_4$ concentrations for the three Japanese sites, COI, RYO, and YON, have been included (**Fig. 10)**. Though the modeled seasonal amplitude is slightly smaller than the observed, the modeled $CH_4$ overall captured the observed synoptic variations, as well as the abrupt increase in August 2013 at COI and RYO.

## 2.2 Real or noise?

The authors claim that GOSAT is able to detect synoptic-scale XCH4 enhancements. It's not clear to me that GOSAT was actually able to pick this up and that it's not just an artifact of the analysis. There are other periods in the record that GOSAT seems to do quite poorly compared to TCCON. For example, June/July 2012 in Figure 4 seems to be a ~20 ppb anomaly in TCCON that GOSAT misses. Why is the former anomaly real and the latter just noise? The two sets of TCCON data are the only thing that makes me think this "anomaly" was real and I'm not convinced that GOSAT actually observed it.

More GOSAT $XCH_4$ retrievals are available in 2013 than 2012. The increase of available retrievals of GOSAT-$XCH_4$ over Japan improves the correlation between GOSAT and TCCON and also enables GOSAT to detect the 2013 summer anomaly clearly. TCCON-$XCH_4$ appears to be anomalously low in August 2012 (though the reviewer mentioned June/July 2012). GOSAT-$XCH_4$ over Japan lowered in August 2012 to the same extent as the TCCON-$XCH_4$ (see also the **Fig 9**). This study is focused on the anomaly in 2013, using the TCCON $XCH_4$ as an observational verification. The scope of this study does not mean that the low $XCH_4$ observed at TCCON sites in summer 2012 is noise.

In addition to the TCCON observations, we have included more model analysis as supporting evidence on the GOSAT-observed anomaly in summer 2013.

Additionally, the authors claim that the modeled XCH4 in August 2013 are lower than 2012 because of these strong zonal winds. However, the GOSAT observations don't seem to support this (Figure 2). Why would the GOSAT observations pick up the Japan high anomaly but not the China low anomaly?

We apologize for the confusion. We need to clarify our explanation of the model result. As the reviewer noticed, the observed GOSAT-$XCH_4$ in Northeastern China-Korea is not lower in 2013 than in 2012, but rather higher. The wind pattern in 2013 altered the spatial distribution of atmospheric $CH_4$ over East China. In 2012, the highest concentration appeared over the southeastern China, while in 2013 the hot spot was shifted to the north and also the level of the highest $XCH_4$ was lower than 2012, as explained below.

First of all, to help distinguish the spatial difference of modeled $XCH_4$ between 2012 and 2013 in Fig. 7 (**now Fig. 8**), we have changed the color scales. What we emphasize here is how the inter-annually varying wind field alters the spatial distributions of surface $CH_4$ concentration and $XCH_4$. In August 2012, the highest $XCH_4$ appear around the southeastern China, while in August 2013 the highest $XCH_4$ area shifts northward. Furthermore, the highest $XCH_4$ level in 2013 is lower than in 2012. Given the same fluxes were used in the model for the both years, these differences between 2012 and 2013 indicate the strong wind carries the $CH_4$-rich air northward, resulting in less accumulation of $CH_4$ around the source area in the southeastern China. This tendency is also seen in September. Regarding the $XCH_4$ in Northeastern China-Korea, the $XCH_4$ level in 2013 is expected to be higher than that in 2012, as $CH_4$-rich air masses are transported from the southeastern China more in 2013 than 2012. The time-series of observed GOSAT $XCH_4$ in Northeastern China-Korea are shown in **Fig. 9**, compared with the modeled $XCH_4$. The $XCH_4$ in September 2013 is higher by ~3ppb than in 2012.

2.3 XCH4 in different parts of a region are not directly comparable
Figure 1 shows a simple example of how topography can impact the XCH4. This is why papers like Kort et al. (2014; GRL) computed anomalous methane by removing the bias due to topography. By averaging GOSAT observations over a large region you could be inducing a sampling bias. For example, if you have a higher density of GOSAT observations over Korea in 2012 and then in 2013 you have more observations over Bejing you will almost certainly have a higher regional" XCH4 simply due to topography. This effect can be up to 20 ppb in parts of Japan (near Mt. Fuji).

The topography bias seems to have less impact our analysis. We have included the location maps of GOSAT $XCH_4$ retrievals we used in this study, including the surface elevation information **(Fig. 3)**. For the entire period 2009-2014 we shown in Fig.2, the surface elevation of $XCH_4$ over Japan ranges up to 1350m, and ~90% of the data is below 500m. The highest surface elevation of $XCH_4$ over Japan is ~850 m in 2012/2013. Since we used the NIES L2 $CH_4$ for General User (GU) which has been applied screening (https://data.gosat.nies.go.jp), there is few retrieval available for the mountainous area in the central Japan (near Mt. Fuji). For Northeastern China-Korea region, the number of observation over Korea was increased in 2013 than 2012. Beijing is located almost at the northwest corner of the target region. There is no significant difference in the observation number around Beijing between 2012 and 2013.

Kort et al. (2014; GRL) analyzed the persisting $XCH_4$ signal at a higher spatial resolution from the multi-year Satellite data. Our analysis is on the temporal signal on a regional scale, which was detectable at the two TCCON sites, ~1000km apart. The topography bias would be critical when analyzing a signal on a local scale, like an anthropogenic large point source.

Additionally, in 2013 and 2014 you see an increase in GOSAT observations over Japan (bottom panel of Figure 2b). If these happened to be over Tokyo (lower elevation) it could explain part of this "Large XCH4 anomaly". What is the spatial distribution of the GOSAT observations? A figure showing the location of the GOSAT observations would be helpful (maybe observation density).

As we mentioned earlier, the location maps of GOSAT observation have been included in the manuscript. As the reviewer noticed, the number of GOSAT observations over Japan was increased in 2013 and 2014 compared to the previous years. This observation increase did not happen only over Tokyo, but all over the Japan islands. It is due to the observation schedule change by the GOSAT project teams among NIES, JAXA and MOE. In the initial regular schedule, there were fewer soundings over only lands, but most soundings were over oceans or land-ocean mixed locations. The soundings over ocean or mixed locations are difficult to be retrieved. To increase the retrievals over Japan, the observation locations were moved to inland from ocean and mixed locations as much as possible. This observation change was implemented on May 6, 2013.

### 2.4 Seasonal cycle

I've got a few issues with the treatment of the seasonal cycle:

1. Remove the seasonal cycle in your data. The anomalies seem to be on the order of 20 ppb, this is comparable to the peak-to-peak amplitude of the seasonal cycle. How much of this is seasonal?

Following the reviewer’s comment, we have added the time-series removed the mean seasonal cycles **(Fig. 5b)**. The amplitude of mean seasonal cycles is around ~20 ppb, comparable with the anomaly we discuss.

2. The seasonal cycle in XCH4 is not necessarily reflective of emissions. The seasonal cycle in the total column does not always follow the seasonal cycle in the emissions (cf. the Bloom et al., 2010 discussion of SCIAMACHY columns and wetland emissions in the Amazon). Changes in stratospheric methane induce higher order harmonics that don't peak when emissions peak. Figure 5 from Saad et al. (2014; AMT) is a nice illustration of this. So statements like, "The summertime high XCH4 must be partially attributed to the seasonal biogenic CH4 emissions from rice paddies and natural wetlands underneath East China and Korea." are not well founded.

Yes, the seasonal cycle in $XCH_4$ does not only reflect the surface emissions, but also other factors such as, the atmospheric mixing in the troposphere and contribution of stratospheric methane.

We appreciate the reviewer's comment on the seasonality in $XCH_4$ and emissions. We have made the sentences more moderate, including the reference to Bloom et al. (2010, Science). Firstly, we have changed the sentence from "*The summertime high XCH4 partially attributed to the seasonal biogenic $CH_4$ emissions from rice paddies and natural wetlands underneath East China and Korea*." to "**The summertime high $XCH_4$ appear to be influenced by the seasonal biogenic $CH_4$ emissions from rice paddies and natural wetlands underneath East China and Korea.**" Secondly, we have referred to Bloom et al. (2010, Science), as adding the sentences below, when we discuss the possibility of contribution from the surface emission change in Sec. 4.2. Other possible factors:

**Here we discuss two factors. One is the surface emission changes. Though the temporal variations in $XCH_4$ do not necessarily correlate with the surface emissions (e.g., Bloom et al., 2010), the surface emission change is potential to impact on the change in $XCH_4$.**

Regarding the contribution of stratospheric methane, we have added the paragraph below, refereeing to Saad et al. (2014, AMT):

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

**fractionations in total column averaged CH4 observed at several TCCON sites. The contribution of stratospheric methane to the anomaly in summer 2013 is supposed to be minor or less influential. Firstly the surface CH4 concentrations at COI and RYO increased in August 2013 when the $XCH_4$ anomaly occurred, suggesting the major contributor on the anomaly is in the troposphere. Secondly, the order of the stratospheric methane fractionation is smaller than ~3 ppb, which would not be enough to produce the anomaly of an order of ~20 ppb.**

3. Figure 7 is presented as “CH4 and XCH4 in August and September 2012 and 2013, with respect to surface CH4 and XCH4 at South Pole". This does not make sense to me. Why would the authors present this as the difference between the Asia and the South Pole? They have different seasonal cycles. CH4 concentrations at 40_N and the South Pole are 6-months out of phase (Northern hemisphere peaks when the Southern hemisphere is at a minimum). This makes interpretation of the plot nearly impossible. Are differences between years due to changes in a different (not shown) hemisphere? Are changes between August and September due to changes in the Southern hemisphere?

To present a spatial distribution with the respect to South Pole is one of typical ways to show the relative spatial distribution. However, to avoid any confusion, we have shown the absolute values of the modeled $CH_4$ and $XCH_4$.

3 Minor comments:
Incomplete literature review
The authors don't seem to have cited any of the previous literature on this topic. The last paragraph on page 24997 briefly mentions a couple studies that used in situ observations to estimate methane fluxes but completely neglects the satellite studies (which are the more relevant studies to this work). Examples of relevant studies: Bergamaschi et al. (JGR 2007, JGR 2009, JGR 2013), Fraser et al. (ACP 2013), Monteil et al. (JGR 2013), Wecht et al. (JGR 2014), Kort et al. (GRL 2014), Cressot et al. (ACP 2014), Houweling et al. (ACP 2014), Turner et al. (ACP 2015), and Alexe et al. (ACP 2015) to name a few.

We thank you for pointing out our lack of the literature review. The references of the flux inversion studies using Satellite data have been included, as adding the sentences below in introduction section in the manuscript.

**These satellite data have been used for the inversion studies of surface CH4 emissions. Most of the satellite-based inversions are focused on the global-scale estimates (e.g., Bergamaschi et al. 2007; 2009; 2013; Fraser et al., 2013; Moteil et al, 2013; Cressot et al, 2014; Houweling et al., 2014; Alexe et al., 2015). Recently the satellite data have been applied for the flux estimation on a regional- and local-scale at a higher spatial resolution. For example, Wecht et al. (2014) compared the multiple observational constraints including GOSAT and TES to optimize methane emission in California. Turner et al. (2015) estimated North American methane emission at a resolution of up to 50 km×50 km using GOSAT data. Kort et al. (2014) demonstrated that satellite-based observations can quantify localized anthropogenic $CH_4$ emissions in the Southwest USA**

Page 24997, Lines 23: Miller et al. (2013) also use aircraft data.

When categorizing the measurements into two types, ground-based and satellite-based, aircraft measurements belong to ground-based. Therefore, we referred to Miller et al. (2013) there. To describe specifically, we have changed “*ground-based measurements*” to **“ground-based measurements including aircraft and shipboard measurements”**.

Page 24999, Lines 16-17: How are you deducing the large methane sources in China? Bottom-up inventories, EDGAR, something else?

The major $CH_4$ source in China is anthropogenic. We used the EDGAR v4.2 for anthropogenic emissions, except the rice cultivation. The $CH_4$ emissions from rice cultivation are from VISIT-$CH_4$. All the $CH_4$ emission sources we used in the model run are described in Sec. 3 Model analysis.

Page 25000, Lines 16-18: As I mentioned in the major comment, you can't compare the XCH4 values. There are biases due to topography, for example, that you have not accounted for.

As we answered in our response to the major comment (2.3), the topography biases are not supposed to affect the analysis.

Page 25003, Lines 18-21: Wind patterns don't seem to support this.

As we answered to the major comment (2.1), we have conducted the trajectory analysis, and the results have been included in the manuscript. We keep the wind patterns to help interpret the model simulations and trajectory analysis results.

Page 25004, Line 3: This is very coarse resolution, is this resolution sufficient to resolve these sort of spatial patterns? I'd rather see this plotted without the spatial interpolation, that way we can see the actual model resolution.

We have included the model-observation comparison of time-series as we mentioned earlier in our response to the major comment (2.1).

Page 25005, Line 4-8: However this isn't seen in the GOSAT data. So if this argument were true and GOSAT can pick up the synoptic event then why isn't it seeing this lower XCH4 over China?

Thank you for pointing out the confusion. The summertime $XCH_4$ over Northeastern China-Korea in 2013 was not lower, but higher than 2012. This statement has been modified to explain clearly, as answered to the major comment earlier. The mode simulation also captures the higher $XCH_4$ in 2013 (**Fig. 9a**).

Figure 4: Does this have the seasonal cycle removed?

Figure 4 (**now Fig. 5a**) showed the detrended observations which long--term trend components are removed. Now we have added **Fig. 5b**, which are removed mean seasonal cycles from the detrended time-series in **Fig. 5a**.

Figure 8: Shouldn't surface observations in Sept 2013 be lower than average since the air is mostly coming from the Pacific? How is this air coming from China? Especially at Saga.

As we answered to the major comment (2.1), we have conducted the back trajectory analysis and included the results in Sec. 4.1**.** In September 2013, the most of airmasses traveled from China/the continent to Saga (**Fig. 12a**). For Tsukuba, in September

2013 some were from the Pacific, but the air mass from China were more influential than in September 2012.

4 References:

Bloom et al.: Large-scale controls of methanogenesis inferred from methane and gravity spaceborne data. Science 327, 322-5, 2010.

We have added.

Kort et al.: Four corners: The largest US methane anomaly viewed from space. Geophys. Res. Lett. 41, doi:10.1002/2014GL0615053, 2014.

We have added.

Saad et al.: Derivation of tropospheric methane from TCCON CH4 and HF total column observations, Atmos. Meas. Tech., 7, 2907-2918, doi:10.5194/amt-7-2907-2014, 2014.

We have added.

## References added

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Figure 3

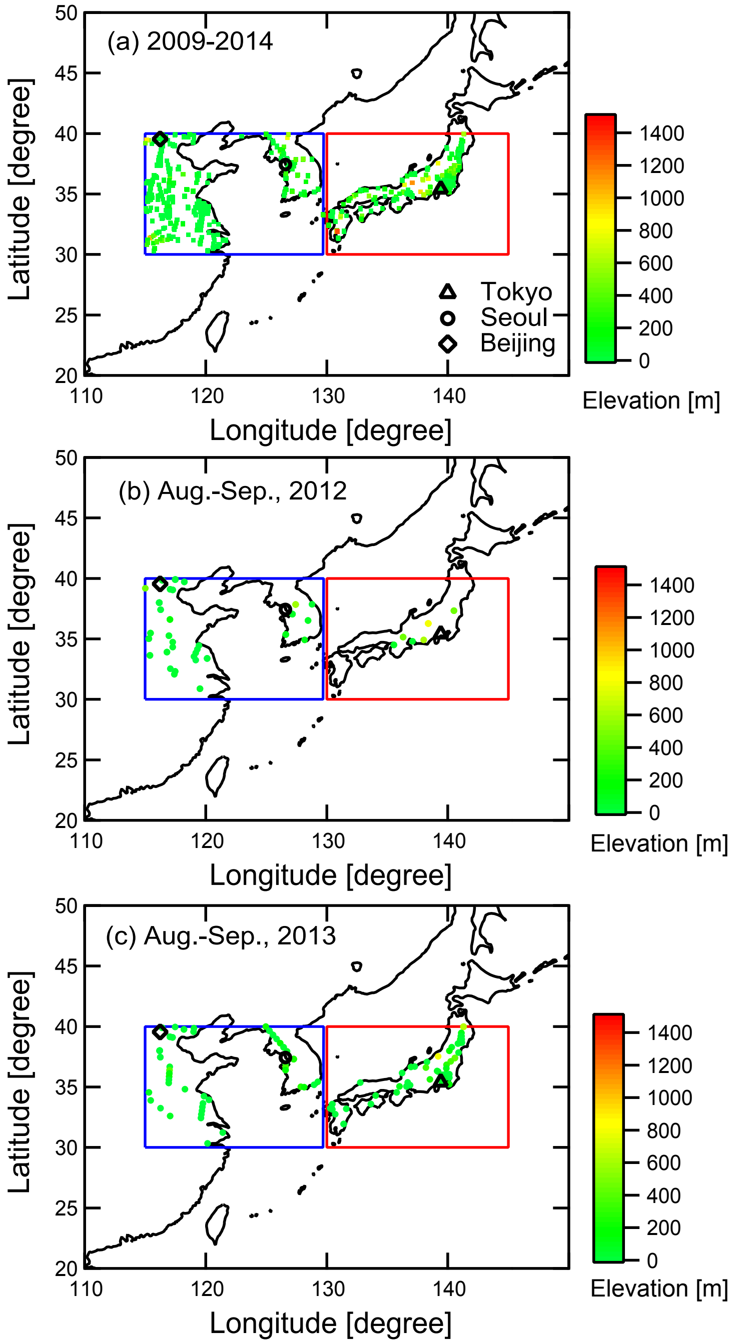
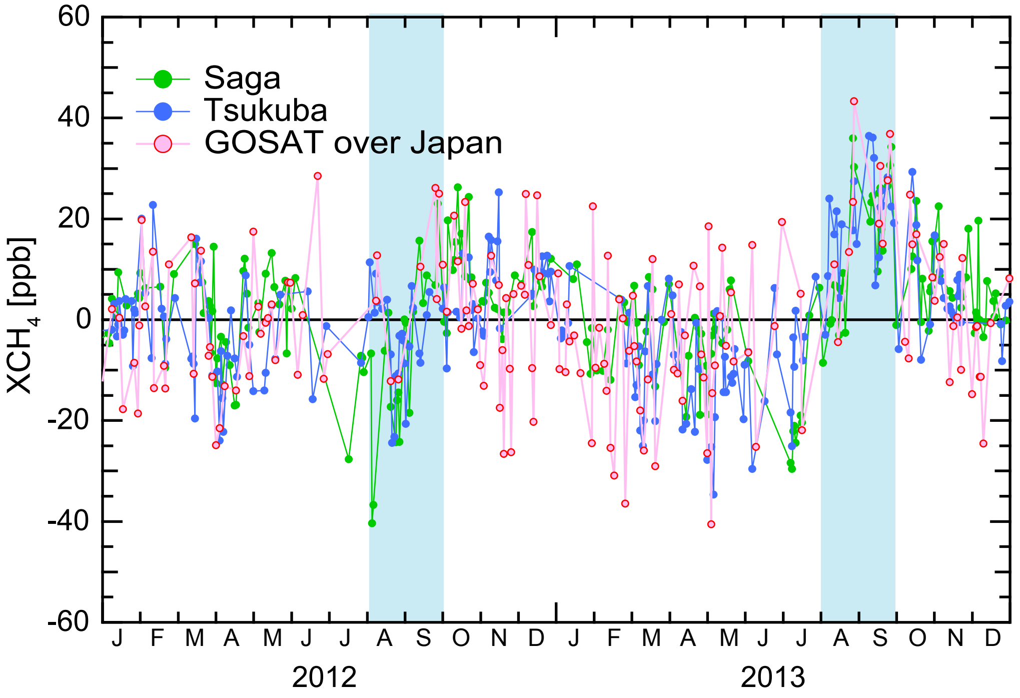


**Figure 3.** Locations of GOSAT soundings with the surface elevations, in the two regions considered in this study. The locations of three capital cities, Tokyo, Seoul and Beijing are also shown in black markers. (a) All soundings of GOSAT data used for 2009-2014. (b) Same with (a) but in August and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

**Figure 5.**

(a) Detrended



(b) Detrended, minus mean seasonal cycle

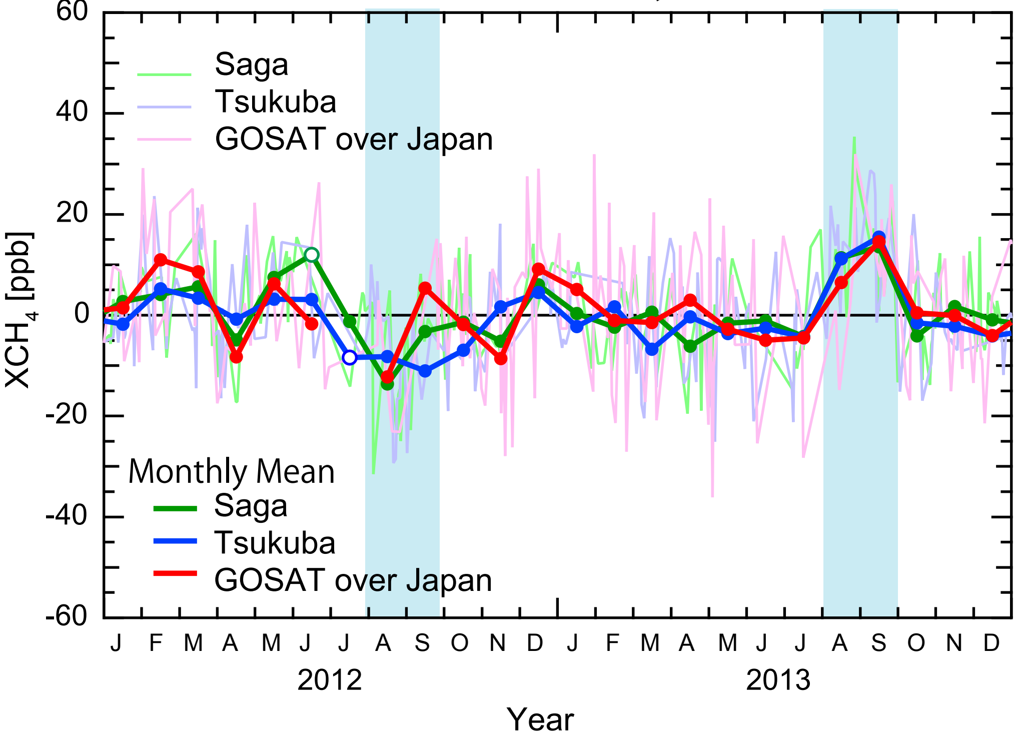
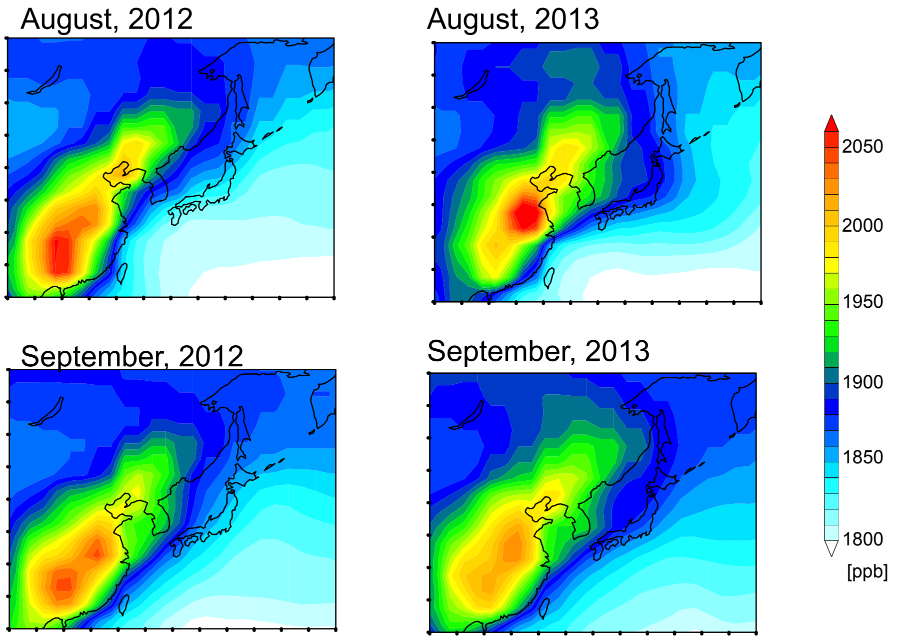


**Figure 5.** (a) Detrended XCH$_4$ for 2012 to 2013 at Saga and Tsukuba, Japan and GOSAT over Japan. (b) Same with (a) but also minus mean seasonal cycles. The monthly means of the individual XCH$_4$ time series are shown in solid lines and circles. The open circles for TCCON indicate that observation days in a month are less than two days. The discontinuity of GOSAT in July 2012 indicates no GOSAT XCH$_4$ retrieval. Long-term components in individual XCH$_4$ 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 8

(a) Surface $CH_4$



(b) $XCH_4$

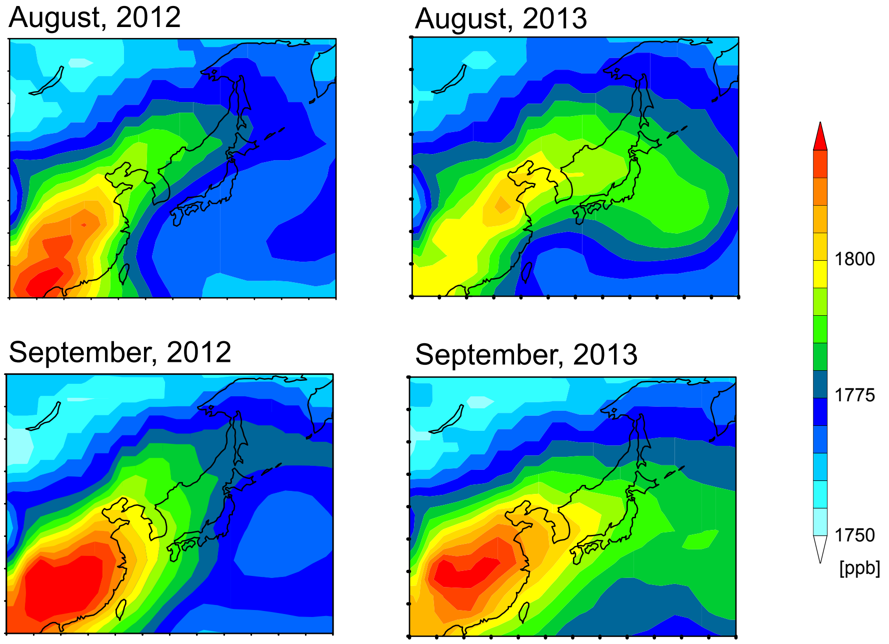
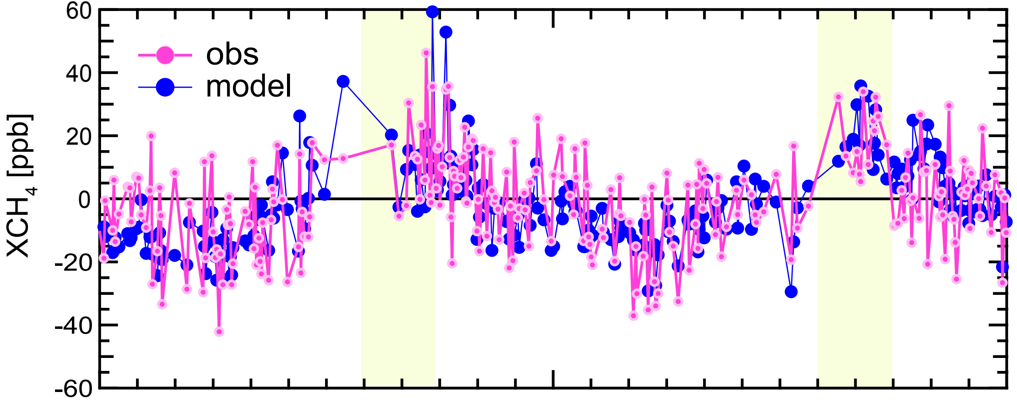


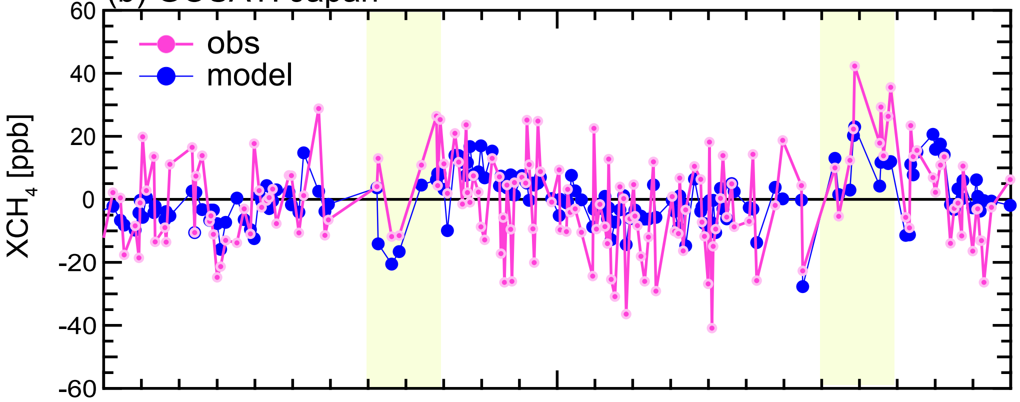
**Figure 8.** Spatial distribution of monthly mean modelled (a) $CH_4$ and (b) $XCH_4$ in August and September of 2012 and 2013.

Figure 9.

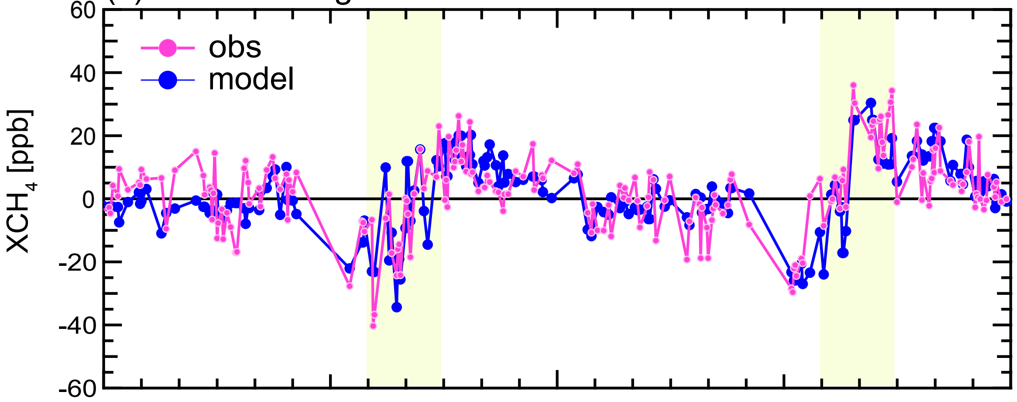
(a) GOSAT: Northeastern China - Korea



(b) GOSAT: Japan



(c) TCCON: Saga



(d) TCCON: Tsukuba

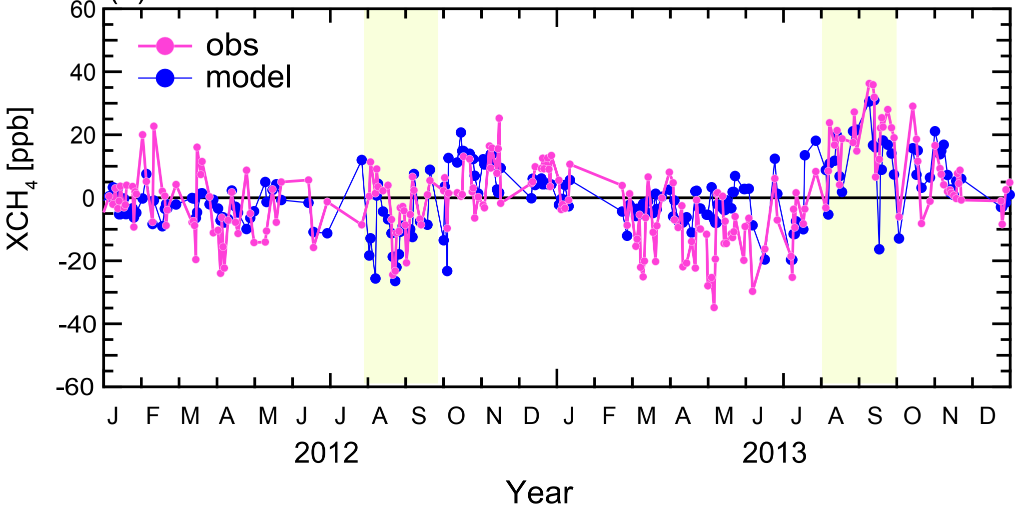


**Figure 9.** Model simulated XCH$_4$ time-series in comparison with the observed GOSAT XCH$_4$ over the two target regions of (a) Northeastern China-Korea and (b) Japan, and with the observed TCCON XCH$_4$ at (c) Saga and (d) Tsukuba. For GOSAT, modeled XCH$_4$ outputs are sampled at corresponding model grids and averaged by region. August and September of both 2012 and 2013 are highlighted.

Figure 10

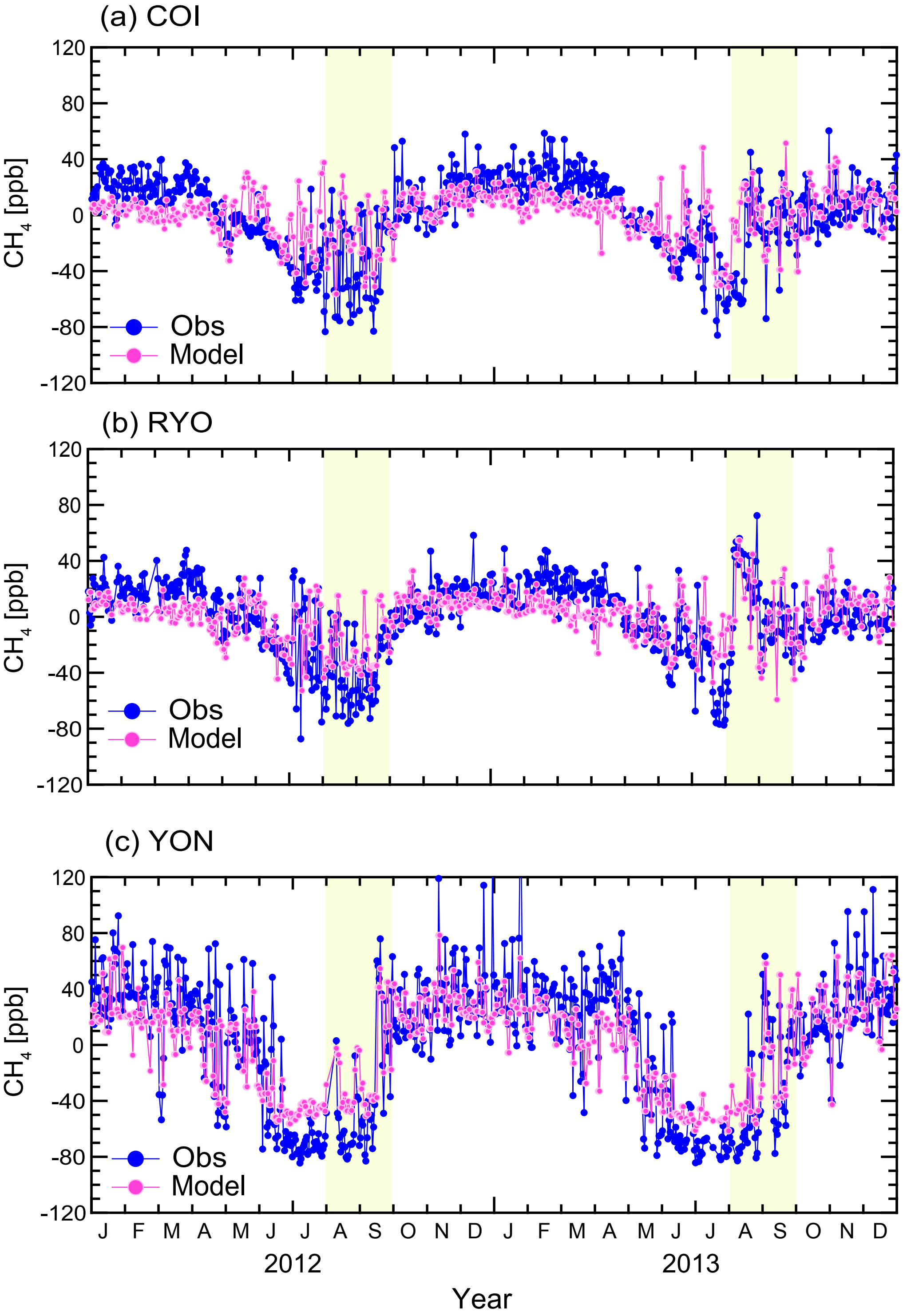
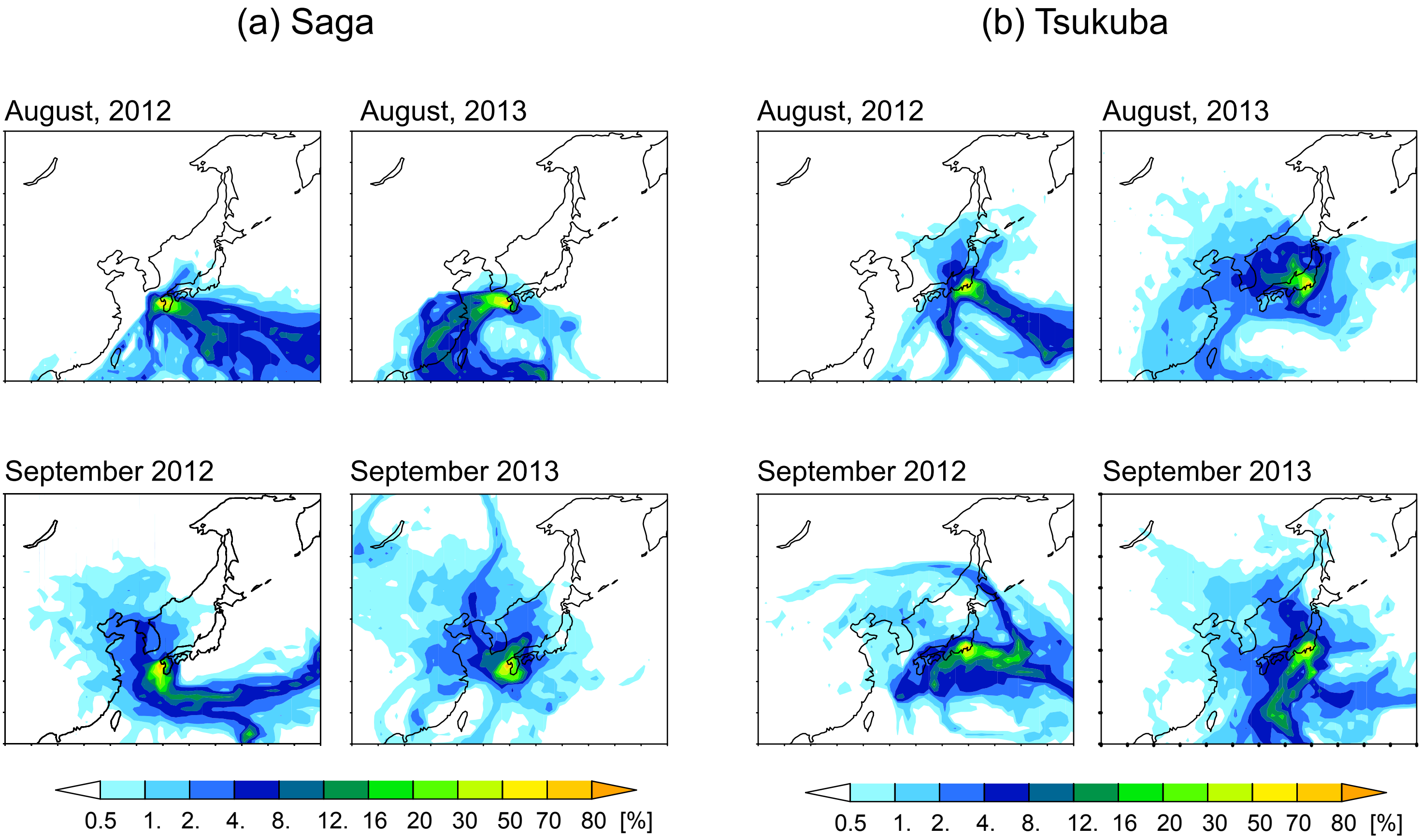


**Figure 10.** Model simulated $CH_4$ time-series in comparison with the observed $CH_4$ at (a) Cape Ochi-ishi (COI, 43.16˚N, 145.49˚E), (b) Ryori (RYO (39.03˚N, 141.82˚E), and (c) Yonagunijima (YON, 24.47˚N, 123.02˚E). August and September of both 2012 and 2013 are highlighted.

Figure 12.


(a) Saga

(b) Tsukuba

August, 2012

August, 2013

September 2012

September 2013

**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˚x1˚ grid air column are counted, and the total numbers are divided by the maximum number per grid.