General.

We would like to thank the anonymous Referee #1 for providing comments to improve and clarify our manuscript. We will revise the text by fully taking the comments into account. Please find our responses to the specific comments and questions below. Our response is written in bold. The revised parts of the manuscript are highlighted in bold red.

5 Comments of Referee #1 and our responses to them

General comments

This paper combines surface and aircraft measurements of atmospheric methane, together with modeling estimates to generate a reference dataset of methane column over a large region over the pacific ocean, south-east of Japan. The resulting time series are analyzed to discuss the growth rate and seasonal cycle.

10 The paper is very well written. The method is clearly described and the various uncertainties are discussed in detail. The paper can be published with minimal changes. I nevertheless offer some suggestions to the authors below :

Specific comments

Comment 1

Line 27: I recommend to use ppb, consistently with the rest of the text, rather than %

15 Response

We added the value in ppb to be consistent with the rest of the text. The revised sentence is as follows:

Lines 26–27: Depending on the models, the difference can be more than 12 ppb (0.6 %), showing the importance for the appropriate choice.

Comment 2

20 Line 36: You could say that methane is the second most important anthropogenic GHG after CO2 (rather than "one of the most")

Response

25

Thank you, we clarified the sentence as follows:

Lines 37–38: Methane (CH_4) is the second most important anthropogenic greenhouse gas (GHG) in the atmosphere after carbon dioxide (CO_2).

Comment 3

Line 208: "Instantaneous lifetime as short as one year" for the summer condition is not clear. Rather, you could provide the oxydatation fraction per month

Response

- 30 Thank for pointing this out. The expression "Instantaneous lifetime" is often used by modelers. Compared to the global atmospheric lifetime, it describes the lifetime at a specific time and location. The global atmospheric lifetime of CH₄ is 9.1 years (Szopa et al., 2021). But looking at the troposphere of the northern midlatitudes during the summer month July, the lifetime of CH₄ can be short as 1 year, as shown in Fig. 14 of Patra et al. (2009) below. During the same month, the instantaneous lifetime of CH₄ of the southern hemisphere is longer.
- 35 Figure 14 of Patra et al. (2009) illustrates the different instantaneous lifetimes in boreal winter, upper plot (a), in comparison with boreal summer, lower plot (b). At 30° N, the lifetime at the lower troposphere in January is about 4 to 8 years, but in July 1 to 2 years.



Patra et al., 2009, Figure 14. Latitude-pressure distribution of monthly-average instantaneous CH₄ lifetime (=1.0 / [K_{OID}×O¹D + 40 K_{OH}×OH + K_{CI}×Cl]) at model grids during (a) boreal winter and (b) boreal summer of 2000.

The main sink is the oxidation with OH radicals, which is primarily produced by the photolysis of ozone in the presence of water vapor (Saunois et al., 2020). That means, during summer, higher temperature and more sunlight can lead to higher concentration of OH. But other factors like atmospheric circulation impact the lifetime essentially

45 (Patra et al., 2009).

In total, the global lifetime remains the same, but at specific locations and times, the instantaneous lifetime can vary depending on the environmental conditions like concentration of OH radicals, atmospheric circulation pattern etc.

Therefore, knowing the instantaneous lifetime, we cannot simply derive the oxidation fraction of methane per month, because we would need to know the concentration of OH, the presence of other atmospheric gases, environmental

50 conditions at that given month and location etc.

However, Chandra et al., 2021, Fig. 11a, simulated the average monthly removal rate of CH₄ over the course of one year. At 30° N, the removal rate is about 60–40 ppb per month.



Fig. 11. Latitude-height distributions of annual (2010) average rate of change in CH₄ concentration (tendency) due to the chemical loss (a) and three transport terms (b, c, d: due to advection, convection, and diffusion, respectively) as simulated by the MIROC4-ACTM. The height (*y*-axis) is shown as mean pressure at model levels, normalized by the surface pressure, as MIROC4-ACTM follows hybrid sigma and pressure coordinate, respectively, below and above 329 hPa or model level 14.

55

Since the oxidation fraction per month for the summer month is not crucial to the understanding of our new approach, we didn't include it in the revised text. However, we clarified the sentence and terminology "instantaneous lifetime" as follows:

Lines 210–214: During boreal summer, a higher OH concentration contributes to an increased CH4 removal by oxidation

60 at our study region (Travis et al., 2020). Including other atmospheric factors, such as atmospheric circulation pattern, models estimate the instantaneous lifetime of CH₄ for July to be as short as 1 year (Fig. 14 in Patra et al., 2009).

Comment 4

Figures 3 and 4 are not clear. I suggest to not show the shaded areas, but only the best estimates together with a single bars for the full period that would indicate the typical uncertainty range

65 Response

80

We revised Fig. 3 and 4 by only showing the 16 ppb uncertainty range of the best result, approach 3 (blended obs. XCH₄), and ACTM_{XCH₄} as grey area. Furthermore, we removed the comparison with the TCCON stations from Fig. 3 to make the comparison of the approaches clearer. Instead, we added a new Fig. 4 which only shows the results of approach 3 in comparison with those of the two TCCON stations. As pointed out by Referee 3#, the missing legend of

70 the linear fit was added to the new Fig. 4.

We also revised Fig. 5 (now Fig. 6) and Fig. A3 (now Fig. A4) in order to have the same color depth. In addition, we revised the caption of the new Fig. 6 and new Fig. A4 by adding the description of the uncertainty range:

Lines 443–445: *Figure 6: Temporal variation of the blended obs. XCH*₄ (*ACTM*_{XCH4}, *black*) *in comparison with GOSAT* 75 *XCH*₄ *retrievals from NIES* (*orange*), *RemoTeC* (*blue*), *and OCFP* (*green*) *at the latitude range 30–40° N* (*a*) *and 20–30° N* (*b*). *The grey area is the 16 ppb uncertainty of the blended obs. XCH*₄.

Lines 511–513: Figure A4: Temporal variation of the blended obs. XCH_4 ($ACTM_{XCH4}$, black) in comparison with GOSAT XCH_4 retrievals from NIES (orange), RemoTeC Heidelberg (HD) (magenta), RemoTeC SRON (blue), and OCFP (green) at the latitude range $g1 = 30-40^{\circ} N$ (a) and $g2 = 20-30^{\circ} N$ (b). The grey area is the 16 ppb uncertainty range of the blended obs. XCH_4 .



Figure 3: Temporal variation of monthly averaged XCH₄ obtained by approach 1 (simple obs. XCH₄, green), approach 2 (obs. XCH₄, orange), and approach 3 (blended obs. XCH₄, black) at the latitude range $30-40^{\circ}$ N (a) and $20-30^{\circ}$ N (b). The uncertainty ranges are 22 ppb, 20 ppb, and 16 ppb for approach 1, 2, and 3 respectively. Only the 16 ppb uncertainty range of approach 3 is shown as grey area. Uncertainty ranges of the other approaches are not shown for readability.



Figure 4: Temporal variation of monthly averaged XCH4 obtained by approach 3 (blended obs. XCH4, black), and from the TCCON station in Saga (green) and Tsukuba (orange) at the latitude range 30–40° N (a) and 20–30° N (b). The grey area is the 16 ppb uncertainty range of approach 3; error bars are the standard deviations of TCCON. Also shown is the linear least-square regression (deep blue line) with a 90% confidence interval on the slope and intercept (deep blue dashed line) of approach 3.

← ACTM_{XCH4} → CAMS_{XCH4} → CAMSinv_{XCH4}



Figure 5: Comparison between the blended obs. XCH4 (approach 3) derived from CH4 profiles using the MIROC4-ACTM (ACTM_{XCH4}, 95
 black), CAMS (CAMS_{XCH4}, green), and CAMSinv (CAMSinv_{XCH4}, orange) for the stratospheric column at the latitude range 30–40° N (a) and 20–30° N (b). The uncertainty range of all results is 16 ppb. The grey area is the uncertainty of ACTM_{XCH4}. Uncertainty ranges of the other results are not shown for readability.

Comment 5

The conclusion is more a summary than a conclusion. It would be better to offer a real conclusion to the reader

100 Response

Thank you for the comment. It is true that we rather provided a summary of the results of our study than a conclusion. However, we keep the summary part, because we believe, it helps the readers to understand the main results of the study. Based on the summary, we added a real conclusion at the end as shown as response to the following Comment 6.

105 We changed the chapter heading to "5 Summary and Conclusion" to clarify that we give a summary of our results and a conclusion at the end. Beside the main conclusion at the end, we concluded each summary paragraph with one or two sentences as follows:

Line 446: 5 Summary and Conclusion

Lines 461–463: Based on the lowest uncertainty and difference towards TCCON, approach 3, defined as blended observation-based XCH₄ (blended obs. XCH₄), is the most suitable for evaluating satellite observations over oceans.

Lines 468–471: MIROC4-ACTM and CAMSinv consider chemical losses in the stratosphere, where MIROC4-ACTM additionally uses an optimized atmospheric transport model. We conclude that for accurately deriving XCH4, a well modelled stratosphere is necessary that includes CH4 sinks. Therefore, either CAMSinv or MIROC4-ACTM is suitable for our approach of which CAMSinv is publicly available.

115 Lines 478–479: These observations show that using the blended obs. XCH₄ dataset, CH₄ trends and seasonal variations can be detected, and satellite observations evaluated.

Comment 6

In addition, the last paragraph is not a conclusion but rather a discussion. Please correct

Response

120 We revised the last paragraph as follows:

Lines 480–494: Having an uncertainty range lower than the mission targets of GOSAT and TROPOMI, the accuracy of satellite derived XCH₄ over oceans can be accessed by our best approach 3. While the blended obs. XCH₄ dataset is not suitable for detecting small scale variations of CH₄ like those from point sources and sinks, spatial pattern and large-scale long-term trends can be evaluated and used for carbon cycle studies. Furthermore, our ship-aircraft

- 125 based approach has the potential to quickly create long-term dataset in areas where other highly precise reference data, such as from measurement campaigns like HIPPO flights or TCCON stations, are not available. Uncertainties and limitations caused by limited in situ data will be reduced in the near future. This includes the re-start of aircraft observations by CONTRAIL over the western Pacific Ocean, probably within the next 2 years, and the spatial extension of other aircraft projects like that of the In-service Aircraft for a Global Observing System (IAGOS) project. As a complement
- 130 to established validation networks we can contribute with our ship-aircraft derived XCH₄ dataset to the validation of TROPOMI, GOSAT-GW and other upcoming satellite missions in future.

References

135 Chandra, N., Patra, P. K., Bisht, J. S. H., Ito, A., Umezawa, T., Saigusa, N., Morimoto, S., Aoki, S., Janssens-Maenhout, G., Fujita, R., Takigawa, M., Watanabe, S., Saitoh, N., and Canadell, J. G.: Emissions from the oil and gas sectors, coal mining and ruminant farming drive methane growth over the past three decades, Journal of the Meteorological Society of Japan, 99, 309–337, https://doi.org/10.2151/jmsj.2021-015, 2021. K. Patra, P., Takigawa, M., Ishijima, K., Choi, B.-C., Cunnold, D., J. Dlugokencky, E., Fraser, P., J. Gomez-Pelaez, A., Goo,

- 140 T.-Y., Kim, J.-S., Krummel, P., Langenfelds, R., Meinhardt, F., Mukai, H., O'Doherty, S., G. Prinn, R., Simmonds, P., Steele, P., Tohjima, Y., Tsuboi, K., Uhse, K., Weiss, R., Worthy, D., and Nakazawa, T.: Growth Rate, Seasonal, Synoptic, Diurnal Variations and Budget of Methane in the Lower Atmosphere, Journal of the Meteorological Society of Japan. Ser. II, 87, 635–663, https://doi.org/10.2151/jmsj.87.635, 2009.
- Saunois, M., Stavert, A., Poulter, B., Bousquet, P., Canadell, J., Jackson, R., Raymond, P., Dlugokencky, E., Houweling, S.,
 Patra, P., Ciais, P., Arora, V., Bastviken, D., Bergamaschi, P., Blake, D., Brailsford, G., Bruhwiler, L., Carlson, K.,
 Carrol, M., Castaldi, S., Chandra, N., Crevoisier, C., Crill, P., Covey, K., Curry, C., Etiope, G., Frankenberg, C.,
 Gedney, N., Hegglin, M., Höglund-Isaksson, L., Hugelius, G., Ishizawa, M., Ito, A., Janssens-Maenhout, G., Jensen,
 K., Joos, F., Kleinen, T., Krummel, P., Langenfelds, R., Laruelle, G., Liu, L., Machida, T., Maksyutov, S., McDonald,
 K., McNorton, J., Miller, P., Melton, J., Morino, I., Müller, J., Murguia-Flores, F., Naik, V., Niwa, Y., Noce, S.,
 O'Doherty, S., Parker, R., Peng, C., Peng, S., Peters, G., Prigent, C., Prinn, R., Ramonet, M., Regnier, P., Riley, W.,
 Rosentreter, J., Segers, A., Simpson, I., Shi, H., Smith, S., Steele, L. P., Thornton, B., Tian, H., Tohjima, Y., Tubiello,
 F., Tsuruta, A., Viovy, N., Voulgarakis, A., Weber, T., van Weele, M., van der Werf, G., Weiss, R., Worthy, D.,
 Wunch, D., Yin, Y., Yoshida, Y., Zhang, W., Zhang, Z., Zhao, Y., Zheng, B., Zhu, Q., Zhu, Q., and Zhuang, Q.: The
 Global Methane Budget 2000–2017, Earth Syst Sci Data, 12, 1561–1623, https://doi.org/10.5194/essd-12-1561-2020,

155

2020.

- Szopa, S., Naik, V., Adhikary, B., Artaxo, P., Berntsen, T., Collins, W. D., Fuzzi, S., Gallardo, L., Kiendler Scharr, A., Klimont, Z., Liao, H., Unger, N., and Zanis, P.: Short-Lived Climate Forcers, 817–922 pp., https://doi.org/10.1017/9781009157896.008, 2021.
- Travis, K. R., Heald, C. L., Allen, H. M., Apel, E. C., Arnold, S. R., Blake, D. R., Brune, W. H., Chen, X., Commane, R.,
 Crounse, J. D., Daube, B. C., Diskin, G. S., Elkins, J. W., Evans, M. J., Hall, S. R., Hintsa, E. J., Hornbrook, R. S.,
 Kasibhatla, P. S., Kim, M. J., Luo, G., McKain, K., Millet, D. B., Moore, F. L., Peischl, J., Ryerson, T. B., Sherwen, T.,
 Thames, A. B., Ullmann, K., Wang, X., Wennberg, P. O., Wolfe, G. M., and Yu, F.: Constraining remote oxidation
 capacity with ATom observations, Atmos Chem Phys, 20, 7753–7781, https://doi.org/10.5194/acp-20-7753-2020,
 2020.