Review's comments

Manuscript Number: *acp-2022-600*

Title: Measurement report: atmospheric CH_4 at regional stations of the Korea Meteorological Administration/ Global Atmosphere Watch Programme: measurement, characteristics and long-term changes of its drivers
Authors: Lee H., Seo W.-I., Li S., Lee S, Kenea S, and Joo S.

The authors of this study present high-quality record of the atmospheric CH_4 mole fractions measured at three WMO/GAW stations in Korea: Anmyeondo (AMY), Jeju Gosan Suwolbong (JGS), and Ulleungdo (ULD). To confirm the data quality in detail, the authors evaluate the total measurement uncertainties at the three sites, which are less than GAW's compatibility goal for atmospheric CH_4 measurement (±2 ppb). Then, they investigate the characteristic features of the CH_4 variations by precisely analyzing the excess CH_4 mole fractions from the background variations, the diurnal cycles, the seasonal cycles, and the growth rates. From PSS analysis and the $^{13}CH_4$ isotopic data obtained at nearby site (TAP), they also discuss the temporal changes in the source region and the dominant source categories from a 5-year period of 2000-2006 to that of 2016-2020. Although I think the discussion based on the PSS and isotopic analyses have some problems, I believe that the data presented in this report are quite reliable and contribute to the global modeling studies especially for the CH_4 emission analysis from the east Asia.

I found that the paper contains material that should be published in Atmospheric Physics and Chemistry. However, I think that the authors should make more efforts to improve the manuscript as is suggested in followings before publication.

General comments:

1) I'm not sure why the authors discuss in depth the influence of water vapor on the CRDS analyzer in Section 2.3 and 3.1. The CRDS analyzer provides dry CH_4 mole fraction by measuring the water vapor simultaneously. The study of Rella et al. (2013) revealed that the preliminary reduction of water vapor of sample air less than

1% allow CRDS analyzer to measure the CH_4 mole fraction within the level of GAW's compatibility goal (±2 ppb). The authors describe that the air samples are dried to −50℃. I believe that the laboratory standard gases and working standard gases uses in this study are sufficiently dried (I think that the absolute dew points should be clarified in the text). In addition, if the air samples are dried by a cold trap of −50℃, the dilution effect is comparable to the reproducibility of the CRDS analyzer. Therefore, I think it would be better to simplify the description about H_2O influence on the CRDS measurement.

2) I'm very curious about the diurnal cycles of the L3 hourly data. If the data selection method described in Section 2.4.2 effectively remove the local effect, there is no statistically significant diurnal cycles in the L3 data. Such information would be useful to make the L3 data more reliable.

3) I don't agree with the discussion based on the PSS and the $\delta^{13}CH_4$ analyses in Section 3.5.

As for the PSS analysis, I'm not sure how to interpret the plots shown in Figs 9a and 9b. Although the authors seem to consider that the plots reflect the CH_4 flux distributions, the flux distribution is considerably different from the previously reported flux distributions (e.g., Ito et al., 2019). Additionally, I cannot believe that such large temporal change in the flux distribution occurred between the two periods. So, I think that the authors should clarify what the plots based on the PSS analysis mean. The authors only show the plots based on the observation at AMY. But I'm curious about the similar plots based on the data at JGS and ULD. If the distributions based on JGS and ULD are similar to Fig. 9b, such result would convince us to some extent that the PSS analysis is reliable.

I'm not sure what the isotopic analysis means. The authors would like to suggest that influences of the biogenic sources increased from 2006-2010 to 2016-2020 because the $\delta^{13}C_{(CH4)}$ values for the latter period shows faster decreasing rates than those for the former period. However, the background $\delta^{13}C_{(CH4)}$ data also show the secularly decreasing trend (for example, you can see the recent $\delta^{13}C_{(CH4)}$ change in NIWA's home page: https://niwa.co.nz/atmosphere/our-data/trace-gas-plots/methane). So, how do the

authors distinguish the regional influence on the $\delta^{13}C_{(CH4)}$ values from the background change? Additonally, why don't the authors use Miller-Tans plot to evaluate of the $\delta^{13}C$ values for the CH_4 regional sources? On the other hand, the values of the intercept of the Keeling plot for CS and KL increased from 2006-2010 to 2016-2020, suggesting the influence of the biogenic sources relatively decreased. This result is inconsistent with the above result.

I'm also curious about the $\delta^{13}C_{(CH4)}$ data during the period from 2010 to 2016. Why don't the authors discuss the period from 2010 to 2016?

Specific comments:

Page 2, line 28: "Kim et al., 2014" should be "Kim et al., 2015".

Page 2, line 30-32: "Li et al., 2020" and "Li et al., 2022" are not listed in References.

Page 2, line 36: "Turnbull et al., 2015" is not listed in References.

Page 3, line3: "Watanabe et al. 2000" is not listed in References.

Page 4, line 17: What's the "−90℃" mean? If it's not the trap temperature, it should be removed to avoid confusion.

Page 5, line 6: I believe that the four laboratory standards are kept in high-pressure cylinders. I think that it would be better to clarify what kind of cylinders are used for the laboratory standards (volume and material).

Page 5, line 13-20: I'm a little bit confused by this part. I think that the CRDS analyzer (Picarro) give dry mol fractions of the atmospheric CH_4. So, why does the authors discuss the dilution effect of water vapor?

Page 5, line 17 (Eq. 1): It should be better to clarify what the $C_{dilution}$ and C_{dry} mean. And the definition of H_{act}, "H_2O difference between standard gases and samples", is rather confusing. Is it "H_2O difference between laboratory standard gases and working standard gases"?

Page 7, line 10-11: It should be better to clarify the detail of the fitting function (order of polynomial and order of harmonics) and the cut-off frequency for the digital filtering based on Thoning et al. (1989).

Page 7, line 12: It should be better to clarify how to compute the monthly data, long-term trend, and seasonal amplitude in detail.

Page 7, Table 2: There are no values of MS for AMY and ULD. Does it mean that MS criteria were not applied to the data for AMY and ULD?

Page 7, line 25-26: The geological locations of WLG and RYO are already given in page 3, line 1-3.

Page 8, line 6-8: It should be better to add the reference for the CH_4 isotopic measurement at INSTAR.

Page 8, line 5 and 9: It says here that the distance between TAP and AMY is 24 km. But other parts in this manuscript, the distance is 28 km. Which value is correct?

Page 8, line 9-10: It should be better to add a figure showing synoptic scale variations of CH_4 observed at AMY and TAP.

Page 8, line 35-page 9, line 4 (2.7 PSS analysis): Since I'm not sure PSS analysis, I tried to read the cited papers, Remann et al., 2004, 2008; Li et al., 2017. But these papers are not listed in References. If the single line for each backward trajectory is used for the calculation of $C_{(i,j)}$, the sensitivity of the potential source strength increase with the distance from the start point in comparison with the reality. This effect would result in the overestimation of the potential source strength with increasing the distance from the station, wouldn't it?

Page 12, line 8-9: Are the characteristics of the bivariate polar plots for the three stations (Figs. 3-5) same with those for the other years?

Page 18, line 10-11: The seasonal cycle of the atmospheric CH_4 is also influenced by the atmospheric transport.

Page 18, line 19: The distance between TAP and AMY is repeatedly described in this manuscript.

Page 18, line 20: “during” what?

Page 19, Figure 7b: Why don’t the authors plot the negative growth rate in the figure?

Page 19, line 12: Was the annual growth rate calculated from the annual means based on the monthly means, listed in Table 4? If so, it would be helpful for the readers to describe that in the manuscript.

Page 19, line 15: CH_4 at AMY, JGS, JLD, and RYO showed significant increases from 2016 to 2017, but small (or negative) increases from 2017 to 2018.

Page 20, line 1: There is relatively large differences in the CH_4 growth rate between WLG and the WMO global mean in 2017 and 2020.

Page 20, line 5-6: There is no description about the $\delta^{13}CH_4$ data at AMY. Please add the explanations for the data. In addition, since the plot shown in Fig. S3 is not Keeling plot but Miller-Tans plot, the slopes of the plots represent the $\delta^{13}C$ of the CH_4 sources.

Page 21, line 1: “Sources affecting CS and KL were paddy fields and …” ?

Page 21, line 13: I’m not sure what “this assumption” means.

Page 21 line 3-14: Are all the flask data collected at TAP plotted in Figs 9? Are the source regions of all the flask data classified into three sectors, CS, CN, and KL? How are they classified?

Page 26, line 21-23: “Shuang-Xi et al., 2013” is not cited in the text.

Page 27, line 3-5: “Winderlich et al., 2010” is not cited in the text.