

Review's comments

Manuscript Number: *acp-2022-600*

Title: Measurement report: atmospheric CH₄ at regional stations of the Korea Meteorological Administration/ Global Atmosphere Watch Programme: measurement, characteristics and long-term changes of its drivers

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I have gone through the response to my comments and the revised manuscript and found the manuscript has been improved to some extent. However, I think that there remain some problems which should be resolved before acceptance. The authors should respond the following comments before publication.

- 1) I'm not sure what the sentence, "However, isotopic signature using $\delta^{13}\text{CH}_4$ explained that CH₄ sources are becoming mixture of not only biogenic but also fossil fuel sources in East Asia from 2006 to 2020", means. I believe that the CH₄ sources in East Asia included fossil fuel-related source also before 2006.
- 2) The added sentence in the revised manuscript, "... since generic water correction cannot be applied to each instrument in different environments" (page 4, line 21) is a little bit misleading. The previous studies including Rella et al. (2013) and Hazan et al. (2016) indicated that the generic manufacturer's water correction function does not necessarily provides appropriate values for wet air samples. However, as Rella et al. (2013) indicated, the preliminary reduction of water vapor of sample air less than 1% allow us to measure the CH₄ mole fraction within the level of GAW's compatibility goal (± 2 ppb). So, I think it would be better for the authors to simply mention that the sample air is dried to reduce the humidity effect less than the detection limit instead of correcting the humidity effect based on the generic manufacturer's water correction function or independently determined water correction function.
- 3) In the revised manuscript, the authors mentioned "AMY started flask sampling for CH₄ and $\delta^{13}\text{C}_{(\text{CH}_4)}$ in December 2013 with same method like TAP and these data were used only for characterization of CH₄ at AMY in section 3.5" (page 8, line

- 22-23). But there is no discussion based on the $\delta^{13}\text{C}_{(\text{CH}_4)}$ data for AMY in this paper.
- 4) The authors should explain how to prepare the Miller-Tans plots shown in Fig. 9b, Fig. 6S, and Fig. 7s because most readers are not necessarily familiar with the plot. In particular, the basic equation for the Miller-Tans plot should be described in the manuscript. And then, the authors should clarify how to determine the background values of the CH_4 concentration and isotope ratio. Additionally, a proper reference should be cited in the manuscript for the Miller-Tans plot.
 - 5) I'm not sure the discussion based on the regional isotopic signatures from the Miller-Tans plots in page 22 line 12-20. The isotopic values can give us the information about the mix of the isotopically different CH_4 sources of the regional emissions causing the excess CH_4 at TAP. Therefore, the temporal and spatial changes in the isotopic ratio probably reflect the changes in the mix of the sources. But there is no information about emission strength in the isotopic ratio for the regional emissions. Therefore, I think it is difficult to discuss the variation in the CH_4 growth rate from the result based on the isotopic analysis in this study. For example, I cannot understand why the authors conclude "This suggested that CH_4 growth rate in East Asia was affected not only biogenic but also pyrogenic sources, unlike global." (page 22, line 12-13). If the authors want to discuss the cause of the trend variation in East Asia, the isotopic ratios for the base line should be discussed. But the authors also mentioned "The long-term trend at AMY was very similar to the global trend" (page 21, line 21-22). So, I think the trend of isotopic ratio at AYM is also similar to the global trend. The similar discussion about the trend in East Asia based on the isotopic analysis is also described in Section 4 (page 24, line 14-15).
 - 6) I'm curious about whether there is seasonality in the source regions (CN, CS, and KL) affecting the observation at TAP. If there is seasonality, does it explain to some extent the differences in the isotopic ratios for the source regions shown in this study? For example, if the air masses are transported from CN to TAP during winter, it is expected that the isotopic signature would be relatively higher because the biogenic CH_4 emissions are reduced during winter.
 - 7) I found several mistakes in English in the manuscript as some examples are shown in the followings:
Page 1, line 20-21: "... growth rate increased 3.3 ppb yr⁻¹ ..." → "... growth rate increased by 3.3 ppb yr⁻¹ ..."

Page 1, line 30: "...compared with that of other long-lived greenhouse gases..."
 → "...compared with those of other long-lived greenhouse gases..."

Page 7, line 18: "... days moving median of of HA..."

Page 10, line 2: "The BLH can was obtained ..."

Page 22, line 13: "... was affected not only biogenic but also pyrogenic..."
 → "...was affected by not only biogenic but also pyrogenic ..."

Therefore, I think that it should be better to check the language in general.

- 8) Added sentences for Eq. 1 (page 6, line 7-9) are rather confusing. The Eq. (1) simply expresses the dilution effect caused by the water vapor (H_{act}). But I cannot understand what the sentences, "Here H_{act} is the H_2O difference between laboratory and working standards. C_{dry} is the CH_4 mole fraction of laboratory standard while $C_{dilution}$ is the expressed CH_4 mole fraction when H_{act} existed.", mean. Do the authors consider the water mole fraction in the laboratory standard is zero? If not, the exact relationship among the H_2O differences between laboratory and working standards ($H^W - H^L$), the CH_4 mole fraction of standard with H^L ($C_{dilution}^L$) and H^W ($C_{dilution}^W$) is expressed by the following equations:

$$C_{dilution}^W = C_{dilution}^L - 0.01(H^W - H^L) \cdot C_{dry}$$

Or

$$C_{dilution}^W / C_{dilution}^L = (1 - 0.01 \cdot H^W) / (1 - 0.01 \cdot H^L),$$

where C_{dry} represents the dry CH_4 mole fraction of the standard.

However, I think above discussion is too detailed. In the manuscript, it would be better to simply examine the absolute H_2O mole fraction of the standard gases and the dried air samples and discuss the dilution effects according to Eq. (1). I believe that the resultant changes in the CH_4 mole fractions caused by the H_2O dilution effect are negligible.

- 9) Page 5, line 28: The URL <https://gml.noaa.gov/ccl/service.html> should be <https://gml.noaa.gov/ccl/services.html>.
- 10) Page 23, Fig. 9(a): It would be better to clarify the boundaries of the northern China (CN), southern China and Korea local (KL) in the map.