

First of all, we would like to thank Vasili Petrenko for his time and his valuable comments. We thank him in the dedicated part of the manuscript.

The authors present a new multiyear record of $\Delta^{14}\text{CH}_4$ and $\Delta^{14}\text{CO}_2$ from the high-altitude Jungfrauoch, Switzerland observatory that spans from 2019 - 2024. The $\Delta^{14}\text{CO}_2$ record shows the expected trend (gradual decline), attributable to continuing fossil CO_2 emissions. In my review, I focus on $\Delta^{14}\text{CH}_4$, as this is the main focus of the paper and also more within my expertise.

$\Delta^{14}\text{C}$ of atmospheric CH_4 is useful for constraining the fossil fraction of methane emissions; this fraction is uncertain on both the regional and global scales. While the interpretation of $\Delta^{14}\text{CH}_4$ is complicated by direct nuclear emissions (something the authors discuss in detail), $\Delta^{14}\text{CH}_4$ can be used in conjunction with methane stable isotope measurements for assessing regional and global CH_4 budgets with higher confidence. The new record from Laemmel et al. fills a void in published recent direct atmospheric $\Delta^{14}\text{CH}_4$ measurements. It also captures a recent gradual increase in $\Delta^{14}\text{CH}_4$ in the free troposphere over Europe. This is novel and valuable; the measurements appear to be of high-quality. I therefore consider this study to be a good fit for ACP, and recommend its publication after some minor revisions.

We are thankful for this positive comments.

Detailed comments:

Impressive precision (as evaluated by repeated measurements of two ambient air standards) of 8 ‰ is achieved for such small samples – well done!

The passive sampling into a large volume steel tank, and nighttime time-averaged sampling are nice innovations well suited for the purpose of getting high-quality background measurements of $\Delta^{14}\text{CH}_4$ at this location.

How stable is CH_4 in the large volume Tecobags, and was this tested for time intervals representative of sample storage? A brief mention / description of this would be useful.

We are thankful for this comment. The extraction in the laboratory was generally done within one week after sampling and most of the time the day after. We added this information in section 2.2. Information about the stability of CH_4 and CO_2 concentrations in the transfer bags were also added in section 2.3. During part of the sampling period presented here, we could use a Picarro analyzer at the Swiss Federal Institute of Metrology METAS in Bern to measure CH_4/CO_2 concentrations in the remaining sampled air from the transfer bags. We added an acknowledgement to both contact people at METAS.

Has simple “memory” (residual carbon from prior sample affecting the next sample) been tested for the laboratory extraction system? This would be useful to describe briefly as well.

What are the typical % yields of the extraction (i.e., what % of the CH_4 carbon is in the end recovered as CO_2 , as compared to the expected amount based on CH_4 mole fraction and air volume processed)? This would be useful to describe briefly as well.

We are grateful for this comment. Information about memory (= cross-contamination) and extraction yield were added in section 2.2 based on tests made by Espic et al. (2019) in 2018-2019. Since then, the extraction system has been continuously optimized and we believe that these values have still been improved even if no new dedicated tests were done for that.

The form of Equation 1 is not familiar to me, and I think the equation needs to be better explained / supported. What is C_obs? What is 14C_n? I don't see this equation derived in the reference that is given to support it (Graven et al., 2019). Considering this, I recommend deriving the equation starting with the most basic quantities (number of 14CH4 molecules, number of total CH4 molecules) in the manuscript (perhaps in the supplement).

We are thankful for this comment. We removed equation 1 from the main manuscript and added a detailed calculation of $\Delta^{14}C^{Nuc}$ at the end of the supplement material.

Line 324. note that Hmiel et al (2020) did not report any Antarctic ice core 14CH4; for Antarctica they only reported firn air measurements

We are grateful for this remark. We corrected this point by saying “previously reported measurements from samples (ice cores and/or firn air) of Greenland and Antarctica”.

Figure 2 caption. Please clarify what exactly the uncertainty bars on panel B represent. Is this the statistical uncertainty from individual AMS measurements? Or combined uncertainty from measurements and other aspects of processing?

We are thankful for this comment. Black error bars show the statistical uncertainty of the individual $\Delta^{14}CH_4$ measurements. We added this information in the caption of Figure 2.

Figure 3 caption – same comment as above regarding uncertainty bars.

We are grateful for this comment . Black error bars show the statistical uncertainty of the individual $\Delta^{14}CH_4$ measurements. Blue error bars represent the standard deviation of the nuclear influence. We completed this information in the caption of Figure 3.

I am not sure that trying to correct the $\Delta^{14}CH_4$ results for regional nuclear power plant influence makes sense, for two reasons. First, as the authors themselves note in another part of the manuscript, $\Delta^{14}CH_4$ releases are episodic rather than constant, so model-based corrections for individual samples are unlikely to be accurate. Second, I think the uncorrected measurements are overall more valuable, as they represent well – mixed tropospheric air at this location; this is what a global transport model would need for interpretation. That said, since the authors are comparing with predictions from a 1-box model, I can see the value of trying to remove the more regional nuclear influence. Perhaps some caveats along above lines could be added when discussing this correction and the corrected results.

We are thankful for this comment. We added some discussion about these points in the first paragraph of the conclusion part.

Figure 4 legend: Hmiel et al (2020), not 2019

We are grateful for this remark. We replaced 2019 by 2020 in Figure 4.