

RC1

First of all, we would like to thank the anonymous reviewer for her/his time and for the valuable comments.

Major comments

In this study Laemmle et al. presented ~5 years' worth of very high quality $\Delta^{14}\text{CH}_4$ and $\Delta^{14}\text{CO}_2$ measurements from Jungfraujoch, Switzerland. Excluding reconstruction from ice cores/firn air (e.g., Hmiel et al., 2020) and sporadic atmospheric $^{14}\text{CH}_4$ measurements conducted during limited duration measurement campaigns (e.g., Sparrow et al., 2018) this study represents the first $\Delta^{14}\text{CH}_4$ measurements from clean background sites for the purpose of long-term global atmospheric monitoring probably since the Barring Head measurements from New Zealand presented by Lassey et al. (2007) almost 20 years ago. Due to the lack of measurements, for years we've been using +350‰ as "background" $\Delta^{14}\text{CH}_4$. This study presents a very welcome update to that value, and very interestingly the authors found that atmospheric $\Delta^{14}\text{CH}_4$ have been increasing since 2019 probably due to the proliferation of PWR (pressurized water reactor) nuclear power plants.

Beyond the novelty of the $\Delta^{14}\text{CH}_4$ measurements, this study also shows that the Bern Radiocarbon laboratory (LARA) can produce state-of-the-art high quality atmospheric ^{14}C data that are comparable to the background measurements made within the ICOS network and other international laboratories that handle background air samples. The passive sampling system using a large volume container (JASS) is very innovative and provides a blueprint for large volume sampling that I hope can be adapted by other global monitoring observatories. Given the study site (in Central Europe), interference from nuclear power plants is unavoidable, and I think the authors did their best to correct for the influences of local nuclear power plants. The paper is very well written; I highly recommend the paper for publication and congratulate the authors on a great manuscript/study.

We are thankful for this positive comments.

I just have several general comments out of genuine curiosity for additional details that I think will help other readers, as well as some slightly nitpicky comments regarding choice of references, mostly in the introduction section (see minor comments).

1. In line 346-348 the authors mentioned: "*The standard deviation of the $\Delta^{14}\text{CH}_4$ value for all 40 CH_4 measurements over 15 months for each PAB bottle (so about 80 CH_4 measurements in total) is 8 ‰ (Fig. S4a,b), which is lower than the instrumental uncertainty of a single $\Delta^{14}\text{CH}_4$ measurement (12 ‰) indicating the satisfactory temporal reliability of our $\Delta^{14}\text{CH}_4$ measurements.*" Similar mention is repeated for $\Delta^{14}\text{CO}_2$ in line 438-441. First, I would replace "*temporal reliability*" with "*long-term reproducibility*." I'm also interested in how the instrumental uncertainty/single sample precision is determined. For this, hopefully the authors are willing to share the composition of samples/standards for the 39 position MICADAS "wheel" (I guess you don't use wheel in MICADAS, so batch/magazine/cartridges). How many OX-2s are used for normalization and how many replicates of the surveillance gas (the PABs cylinders) are put in each sample batch. What was the average stdev of however many (4,6,8,10) primary OX-2 standards used for normalization within a single sample batch, and whether the average stdev of primary standards was bigger/smaller than the single sample precision derived from counting statistics. Also, were the samples measured together in small amounts of 39 position sample batches/magazines (like within 6-7 batches at the minimum to account for the 130-something samples and 80 surveillance samples, or are they more spread out over larger amounts of sample

batches. Knowing the composition of the 39 position sample within a single MICADAS batch will give us a good sense of the inter-batch/magazine reproducibility.

We are thankful for this comment. Concerning the use of “long-term reproducibility” instead of “temporal reliability”, we agree with the reviewer and made the changes. Concerning the information about instrumental uncertainty/single sample precision, we added them in the section 2.2.

2. Moving to integrated and automated sampling with the JASS system is clearly very beneficial in terms of sampling consistency and reducing labor needed to do the grab samples. But I have a couple of follow-up questions regarding the JASS system. The grab samples clearly show some highly elevated $\Delta^{14}\text{CH}_4$ ($>420\text{‰}$ cutoff) – likely given the sporadic nature of $^{14}\text{CH}_4$ emissions from PWR venting. Would it be fair to say/acknowledge that with integrated sampling it is harder to correct for potential elevation from sporadic PWR venting? The footprint-based correction relies on annual, quarterly, and monthly NPP emissions – this might not capture the sporadic emissions from venting so there’s still some upside to grab samples. Second, in line 132-134 the authors mentioned that “*air sampling should occur passively, i.e., without any mechanical pumps in contact with the sampled air, thereby minimizing the risk of contamination from membrane outgassing.*” This is a noble principle to strive for, but is it fair to also acknowledge that one still needs to transfer the sample from the large volume container into the transport container? Reading the manuscript, in line 169-171 the authors wrote: “*At this stage, the transfer pump was evacuating the tank and after ~30 s of line flushing, a bag was connected to the line for the transfer of the sampled air. After about one hour, enough air was transferred to the bag for further ^{14}C analyses.*” So, the sample air is still in contact with mechanical pump, arguably even longer (1hr) compared to the 22-55 min needed to compress enough air during the manual grab samples (line 124). As such, the benefit of JASS sampling scheme in terms of sample cleanliness is only valid if the KNFN022 pump used for grab samples is dirtier than the KNF N922 pump used to transfer the sample from the large volume reservoir to the transfer bag.

We are grateful for these comments. Concerning the discussion grab vs integrated samples, we added some comments in the *Discussion* section. Concerning the sample cleanliness regarding the pumps, as correction, we exchanged the order of both principles (line 135) and added “as far as possible” for the passive sampling. As mentioned, taking grab samples took 20-55 min; with the JASS, it is possible to passively sample air over 84 hours (6 hours over 14 days) and transfer it into a bag within one hour. So calculating the ratio between pumping time and sampling time, we got 1 for the grab sampling method (pumping time = sampling time) and $1/84 \sim 0.01$ for the JASS method meaning that the potential influence of the pumping time on the sampling time is much lower for the JASS method. Moreover, we chose the KNF N922 pump especially for its high chemical resistance because of many mobile parts in PTFE or PTFE-coated (eg. pump head and diaphragm).

3. Between the $\Delta^{14}\text{CH}_4$ with $>420\text{‰}$ cutoff, Radon filter, and Radon filter + footprint based nuclear correction (Table 1) – I personally think that the Radon filter is the most straightforward/foolproof representative of a well-mixed background mid-latitude $\Delta^{14}\text{CH}_4$, given the sporadic nature of $^{14}\text{CH}_4$ emissions from PWR that is not captured by emission inventories. The authors take a more agnostic approach to this, which is fine, but I think at least the Radon filtered data should also be on Figure 4. I will also welcome an additional discussion if the authors have any inclination one way or another regarding which of the 3 annual averages they think is most representative of a true mid-latitude background value for $^{14}\text{CH}_4$.

We are grateful for these comments. We added the mean annual Rn-filtered $\Delta^{14}\text{CH}_4$ values to Figure 4. To complete Table 1, we extended the first column with annual average of raw $\Delta^{14}\text{CH}_4$ values. Furthermore, as a remark, in the first version of the manuscript, $\Delta^{14}\text{CH}_4^{\text{Rn-filt}}$ values were $\Delta^{14}\text{CH}_4$ values lower than 420 ‰ with corresponding $\text{Rn} \leq 1.5 \text{ Bq m}^{-3}$. In this new version, we simplify the definition of $\Delta^{14}\text{CH}_4^{\text{Rn-filt}}$ by only using the Rn-filtering ($\text{Rn} \leq 1.5 \text{ Bq m}^{-3}$); the values selected remained finally the same. In the conclusion part, we added a quick discussion about the different annual averages.

4. Since the footprints are already made, it would be interesting if the authors could show comparison of footprints/catchment area for the grab samples vs. integrated JASS sampling, maybe as a supplementary figure. Either pick two footprints that are somewhat representative of the average for the two sampling schemes, or sum the footprints and provide a contour of influence for the two sampling schemes.

We are grateful for this suggestion. We included a concentration footprint analysis for the different sampling strategies into a supplement (new Fig. S2). The analysis confirms the expectation that integrated nighttime sampling improves the representativeness of the sampled air mass as compared to biweekly grab sampling. We added a reference to the new Figure in section 2.1.

Minor comments

Line 30: “*biweekly*” is ambiguous as it can literally mean twice a week or once every two weeks. Be more specific and change it to either “*once every two weeks*” or “*fortnightly*”.

In the whole manuscript (figures included), we replaced “*biweekly*” by “*fortnightly*”.

Line 34: Add “*continued*” before “*dilution*.” This is just to draw the contrast that the continued decrease in $^{14}\text{CO}_2$ is pretty much expected and has been a trend that’s been observed in all global sites, unlike the increase in $^{14}\text{CH}_4$, which is a novel discovery of this study.

We added “*continued*” before “*dilution*”.

Line 37: “... *the mean nuclear ^{14}C contribution to our individual measurements was estimated to be $7 \pm 9 \text{ ‰}$...*” This is slightly misleading, as from my understanding not all measurements are corrected for local nuclear contribution, only measurements with high Radon. I would instead say something along the line of “*To assess the nuclear ^{14}C contribution to our individual measurements, we use a combination of in situ Radon-222 measurements and Lagrangian particle dispersion model convolved with bottom-up inventory of ^{14}C emissions from nuclear power plants.*” Probably also put this sentence before line 31 “*Over this period, $\Delta^{14}\text{CH}_4$ values showed an increase...*”

We are thankful for this comment. We agree that not all measurements are corrected for local nuclear contribution, but only measurements with low radon values (and not high radon values as mentioned by the reviewer). In the abstract, we removed the figures “ $7 \pm 9 \text{ ‰}$ ” and added the sentence suggested by the reviewer. Furthermore, in the conclusion, we refined the sentence presenting these figures by adding “ ^{222}Rn -filtered” before $\Delta^{14}\text{CH}_4$.

Line 47: The Hmiel et al. (2025) citation here is not appropriate. Please cite the original Law Dome ice core CH_4 mole fraction paper (MacFarling Meure et al., 2006).

We are thankful for this comment; we updated the citation.

Line 52: A more appropriate citation for the ^{14}C half-life of 5700 years is probably a combination of (Kutschera, 2013) – where the compilation of previous half-life measurements were made,

(National Nuclear Data Center, Brookhaven National Laboratory, www.nndc.bnl.gov), and the most “recent” effort by Roberts and Southon (2007).

We are grateful for this comment; we updated the citations.

Line 61-62: “*Atmospheric $^{14}\text{CO}_2$ have a long history. Since the first measurements focused on the atmospheric $^{14}\text{CO}_2$ bomb peak in the 1950-60s...*” The most appropriate citation for this statement, even before Levin et al. (1985) is obviously the original Rafter and Fergusson (1957) paper.

We are grateful for this comment; we updated the list of citations.

Line 69-72: “*Since the bomb peak in the middle of the last century, $\Delta^{14}\text{CO}_2$ values have been declining mostly due to the emissions of ^{14}C -free fossil fuel CO_2 , which depletes the atmospheric $\Delta^{14}\text{CO}_2$ signal (Levin et al., 2010).*” I understand what the authors meant to say, but this is not 100% correct, atmospheric $^{14}\text{CO}_2$ is coming down from the bomb peak also (or even mostly) because of uptake by plants and the surface ocean. Maybe cite something like the Oeschger et al. (1975) paper or something more recent regarding the partitioning of land/ocean sink and revise the sentence accordingly.

We agree with this comment; we updated the sentence and added the citations of Oeschger et al. (1975) and Graven et al. (2024).

Line 85: Consider giving acknowledgements to the very first pioneering atmospheric $^{14}\text{CH}_4$ measurements by Lowe et al. (1988) and Wahlen et al. (1989).

We agree with this comment and added a sentence in that sense with both citations.

Line 94-95: “*Fujita et al. (2025) estimated 30% lower global CH_4 emissions from the fossil-fuel industry compared to previous isotope-based studies.*” Clarify that the “previous isotope-based studies” specifically referred in this sentence are d^{13}C - CH_4 stable isotope only inversion like Basu et al. (2022) which does not include ^{14}C consideration.

We are grateful for this comment; we updated the sentence accordingly.

Line 128: I don’t quite understand the use of UTC throughout the manuscript - I think this can be a bit confusing because JFJ local time is not UTC, and when trying to sample the free troposphere local time matters more and absolute time. I would recommend just using local time and mention (UTC+1/UTC+2).

We are thankful for this comment. We understand that the use of UTC can be confusing but we still would like to let it like this. We chose to use the Raspberry Pi module controlling the JASS in UTC to avoid any datetime issues because of local time change (UTC+1/UTC+2). In addition, the whole modelling work was done on UTC basis. In total, UTC is used only 3 times in the manuscript and we believe that the readers will be fine with that. We added the local time information “(i.e. 08:00 and 12:00 local time, UTC+1/UTC+2)” at the end of the second paragraph in section 2.1.

Line 213: “*... and in equation 29 in Stenstrom et al. (2011).*” I’m a big fan of Stenstrom et al. (2011), but unfortunately it is not appropriate to cite as this was an internal report that was not peer-reviewed.

We understand this comment and removed this citation.

Line 227-229: “*In this study, R_n was used as a proxy to distinguish atmospheric conditions mostly influenced by free tropospheric conditions from conditions mostly influenced by the planetary*

boundary layer (i.e., with recent land contact).” This sentence is a bit awkward, I would just say something along the lines of “*In this study, R_n was used as a proxy to distinguish free tropospheric samples from those originating from the planetary boundary layer (i.e., with recent land contact).*”

We are thankful for this comment and replaced the original sentence by the suggested one.

Line 288: I think “convolve” might be the more correct mathematical term instead of “convolute”. Footprints are convolved with flux to produce mole fraction enhancement at receptor site.

We are thankful for this comment; we replaced “convoluted” by “convolved”.

Line 452-453: “*The offset in 2019 was probably due to a small fossil contamination in our early $\Delta^{14}\text{CO}_2$ measurements that was remedied during the year.*” Not that I don’t believe this explanation, but as it stands by reading this manuscript alone it is not very clear that the “true” European background $\Delta^{14}\text{CO}_2$ in 2019 is closer to the ICOS/Heidelberg values than the LARA values. Maybe show/at least mention Mace Head (MHD) measurements as independent dataset to support the idea that LARA sampling stream was slightly contaminated in 2019?

We are grateful for this comment. We added annual mean $\Delta^{14}\text{CO}_2$ values from the Mace Head (MHD) Atmospheric Research Station for 2019-2022 in Table 2. The good consistency of ICOS JFJ and MHD data in 2019 and 2020 strengthens the idea that our early $\Delta^{14}\text{CO}_2$ measurements in 2019 were slightly biased by fossil contamination that was corrected afterwards.

Line 526: Very minor thing, I might have missed it but I think “ $\Delta\Delta^{14}\text{CO}_2$ ” is not defined. Just say “*difference between ICOS sodium hydroxide-based integrated sampling vs. our JASS system*” to reduce jargon.

We are grateful for this comment; we added ($\Delta\Delta^{14}\text{CO}_2$) in the caption of figure 6 and replaced $\Delta\Delta^{14}\text{CO}_2$ by the proposed sentence.

References

Basu, S., Lan, X., Dlugokencky, E., Michel, S., Schwietzke, S., Miller, J. B., Bruhwiler, L., Oh, Y., Tans, P. P., Apadula, F., Gatti, L. V., Jordan, A., Necki, J., Sasakawa, M., Morimoto, S., Di Iorio, T., Lee, H., Arduini, J., and Manca, G.: Estimating emissions of methane consistent with atmospheric measurements of methane and $\delta^{13}\text{C}$ of methane, *Atmospheric Chemistry and Physics*, 22, 15351–15377, <https://doi.org/10.5194/acp-22-15351-2022>, 2022.

Hmiel, B., Petrenko, V. V., Dyonisius, M., Buizert, C., Smith, A. M., Place, P. F., Harth, C. M., Beaudette, R., Hua, Q., Yang, B., Vimont, I., Michel, S. E., Severinghaus, J. P., Etheridge, D. M., Bromley, T. M., Schmitt, J., Fain, X., Weiss, R. F., and Dlugokencky, E. J.: Preindustrial $^{14}\text{CH}_4$ indicates that anthropogenic fossil CH_4 emissions are underestimated, *Nature*, 2020.

Kutschera, W.: Applications of accelerator mass spectrometry, *International Journal of Mass Spectrometry*, 349–350, 203–218, <https://doi.org/10.1016/j.ijms.2013.05.023>, 2013.

Lassey, K. R., Lowe, D. C., and Smith, A. M.: The atmospheric cycling of radiomethane and the “fossil fraction” of the methane source, *Atmospheric Chemistry and Physics*, 7, 2141–2149, <https://doi.org/10.5194/acp-7-2141-2007>, 2007.

Lowe, D. C., Brenninkmeijer, C. A. M., Manning, M. R., Sparks, R., and Wallace, G.: Radiocarbon determination of atmospheric methane at Baring Head, New Zealand, *Nature*, 332, 522–525, <https://doi.org/10.1038/332522a0>, 1988.

MacFarling Meure, C., Etheridge, D., Trudinger, C., Steele, P., Langenfelds, R., van Ommen, T., Smith, A., and Elkins, J.: Law Dome CO₂, CH₄ and N₂O ice core records extended to 2000 years BP, *Geophys. Res. Lett.*, 33, L14810, <https://doi.org/10.1029/2006GL026152>, 2006.

Oeschger, H., Siegenthaler, U., Schotterer, U., and Gugelmann, A.: A box diffusion model to study the carbon dioxide exchange in nature, *Tellus*, 27, 168–192, <https://doi.org/10.1111/j.2153-3490.1975.tb01671.x>, 1975.

Rafter, T. A. and Fergusson, G. J.: “Atom Bomb Effect”—Recent Increase of Carbon-14 Content of the Atmosphere and Biosphere, *Science*, 126, 557–558, <https://doi.org/10.1126/science.126.3273.557>, 1957.

Roberts, M. L. and Southon, J. R.: A Preliminary Determination of the Absolute ¹⁴C/¹²C Ratio of OX-I, *Radiocarbon*, 49, 441–445, <https://doi.org/10.1017/S0033822200042363>, 2007.

Sparrow, K. J., Kessler, J. D., Southon, J. R., Garcia-Tigreros, F., Schreiner, K. M., Ruppel, C. D., Miller, J. B., Lehman, S. J., and Xu, X.: Limited contribution of ancient methane to surface waters of the U.S. Beaufort Sea shelf, *Science Advances*, 4, eaao4842, <https://doi.org/10.1126/sciadv.aao4842>, 2018.

Stuiver, M. and Polach, H. A.: Discussion Reporting of ¹⁴C Data, *Radiocarbon*, 19, 355–363, <https://doi.org/10.1017/S0033822200003672>, 1977.

Wahlen, M., Tanaka, N., Henry, R., Deck, B., Zeglen, J., Vogel, J. S., Southon, J., Shemesh, A., Fairbanks, R., and Broecker, W.: Carbon-14 in Methane Sources and in Atmospheric Methane: The Contribution from Fossil Carbon, *Science*, 245, 286–290, <https://doi.org/10.1126/science.245.4915.286>, 1989.

References

Graven, H. D., Warren, H., Gibbs, H. K., Khatiwala, S., Koven, C., Lester, J., Levin, I., Spawn-Lee, S. A., and Wieder, W.: Bomb radiocarbon evidence for strong global carbon uptake and turnover in terrestrial vegetation, *Science*, 384, 1335–1339, <https://doi.org/10.1126/science.adl4443>, 2024.

RC2

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 Jungfraujoch, 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 Laemmle 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.