

We thank anonymous referee #1 and Greg Balco for their constructive comments. We have made the following revisions:

1. Following comments from referee #1, all figures have been adjusted to within the page margins and their resolution increased.
2. Following comments from Greg Balco, we adjusted the main emphasis of the paper. We are confident about our measured ^{14}C values and the *in situ* ^{14}C partitioning between CO_2 , CO and CH_4 . As discussed in the manuscript, the muogenic *in situ* ^{14}C production rates in ice and the partitioning between ^{14}C species are critical to disentangle the paleoatmospheric and cosmogenic ^{14}C signals in ice cores. Thus, we believe on their own these results have a lot of scientific merit.
3. Our data also imply that the Heisinger et al. production rates are too high. Following comments from Greg Balco, we added several paragraphs in the discussion section thoroughly examining the possibility of our measurements being biased. We also conducted the sensitivity analyses (which we summarize below). Unfortunately, we still do not have a good explanation behind the discrepancy between production rates implied by our data and that of Heisinger et al. ^{14}C in organic phases such as formaldehyde might explain some, but very likely not all of the discrepancy. As our author team lack the expertise in nuclear chemistry to further investigate the causes behind this disagreement, within the scope of this manuscript we now present the disagreement as more of a conundrum that requires further research.

Detailed responses to comments from Greg Balco:

We added a thorough discussion about the possibility that our measurements might be biased (page 15, line 471 to page 16, line 518). We separate this discussion between the ^{14}CO and $^{14}\text{CO}_2$ measurements (the CH_4 fraction of total ^{14}C is <1%).

We believe that our ^{14}CO measurements are robust as we use a well-established technique. In air, the analytical technique we use yields comparable results to independent atmospheric ^{14}CO measurements from other research groups, as well as expected atmospheric ^{14}CO concentrations based on our knowledge about the sources and sinks of ^{14}CO in the present atmosphere (Petrenko et al., 2021). We also have ^{14}CO measurements from firn air and relatively recent (preindustrial) ice from Summit, Greenland (Hmiel et al., 2020; Hmiel, 2020). The ^{14}CO data from Summit, Greenland agree within uncertainties with the inferred ^{14}CO production rates from Taylor Glacier (Hmiel, 2020). We also collected field procedural blanks to

characterize the system blanks. To conclude, to the best of our knowledge, we have no reason to believe that our measurements are biased or that we are systematically losing ^{14}CO .

We acknowledge that our $^{14}\text{CO}_2$ measurements used a more novel and previously untested sublimation technique. However, we carefully characterize the system blanks using our bubble-free ice samples while flowing two standard gases with “modern” and “dead” $^{14}\text{CO}_2$ activities. The results of these tests are presented in Table S8. We do not see significant alterations in the measured $^{14}\text{CO}_2$ over bubble-free ice – indicating that the process of sublimating ice and flowing gas through the system components recovers $^{14}\text{CO}_2$ as expected. As was also already shown in the supplement (Table S9, measured vs expected mass), we are getting close to the expected amounts of carbon from CO_2 (i.e., there is no systematic loss of CO_2 during ice processing).

Another strong indication that our measurements are robust is the good agreement with independent results from Van der Kemp et al. (2002) collected from a different study site (Scharffenbergbotnen) using entirely different analytical techniques. First, we find good agreement in the ratio of ^{14}C compounds ($^{14}\text{CO}_2$ fraction = 0.66 ± 0.12 in this study, 0.69 in Van der Kemp et al., 2002). The ^{14}C ratios are independent of the ablation rates and ^{14}C production model; they are strictly a measure of the analytical technique robustness. Van der Kemp et al. (2002) used a dry extraction system for their $^{14}\text{CO}_2$ and ^{14}CO measurements. We used a sublimation technique for $^{14}\text{CO}_2$ and a wet extraction (melting) for ^{14}CO . The agreement in the ratios suggests that our extraction methods (and theirs) were not systematically losing either ^{14}CO or $^{14}\text{CO}_2$ (which would then bias the $^{14}\text{CO}_2$ and ^{14}CO fraction). It is theoretically possible that both our measurements and Van der Kemp et al. (2002) are wrong, but it would require all 3 analytical systems from these studies to be systematically/coincidentally wrong in the same direction and by the same magnitude to produce the same $^{14}\text{CO}_2$ and ^{14}CO fractions; this seems unlikely.

Second, we also find good agreement with Van der Kemp et al. (2002) for the *in situ* muogenic production rates, for both negative muon capture and fast muon reaction. This agreement takes into account the ablation rates from both areas. The deepest sample from Scharffenbergbotnen study is 45m and in our analysis we take the $^{14}\text{CO}_2$ and ^{14}CO from this depth as the initial condition. Because the Scharffenbergbotnen ablation rate based on stake reading is 0.16 ± 0.04 cm/yr, with the 1D ablation model our calculation only covers ~280 years. As discussed in the revised manuscript, a 14-year-long observation study published around the same time (Sinisalo et al., 2003) showed no significant and systematic change in the ablation rate of Scharffenbergbotnen blue ice area.

Very large (factor of 3 or more) changes in the ablation rate at both Scharffenbergbotnen and Taylor Glacier would be required to reconcile our and Van der Kemp et al (2002) measurements with the Heisinger et al. (2002) estimates, with the long-term ablation rates being much higher than those measured by stakes in recent years. Ablation rate at blue ice areas is controlled by climate via a combination of temperature, insolation and wind (mainly catabatic). To get a much higher long-term ablation rate at both Taylor Glacier and Scharffenbergbotnen (which are on opposite sides of Antarctica), we would need either the temperatures to have dropped sharply in the last couple of decades (definitely not the case), the winds to have slowed dramatically (this would be surprising given the steady to extreme catabatic winds Taylor Glacier experiences now), or for insolation to sharply decrease (this also seems highly unlikely given our experience on Taylor Glacier, which is about 80% sunny during peak summer in November - January). Finally, the Kavanaugh et al (2009 a, 2009b) glaciological studies of Taylor Glacier indicated that the glacier is approximately at steady-state given the stake-measured ablation rates, also arguing against large recent changes in ablation rates. To conclude, a large decrease in ablation rates in recent years as compared to the long-term average does not seem to be a realistic explanation.

We conducted sensitivity analyses following the comments from Greg Balco. We increased the total ^{14}C by +25% to account for possible *in situ* ^{14}C in organics and used the high ablation rate / deep ice flow scenario (Fig. S8). In the first test, we set the ^{14}C production from negative muon to be 84% of the Heisinger et al. (2002) production rate. We then tuned the ^{14}C production rate from fast muons under this scenario and find that the best-fit ^{14}C production rate from fast muons is zero. However, the fit is still not very good (dashed red line, Fig. 6D). This shows that our data, even assuming 25% contribution from organics and high ablation rate / deep ice flow scenario (which correspond to high ^{14}C production rates) cannot be reconciled with negative muon capture ^{14}C production rate estimate from Heisinger et al. (2002).

Figure 1 (of this response letter, see below) further illustrates how the ^{14}C production rate from Heisinger et al. (2002) is not compatible with our data, even when ^{14}C production from fast muon is set to zero and the data is uniformly scaled by +100% to account for ^{14}C in organics. The Heisinger et al. (2002) negative muon capture production rate yields a much higher total ^{14}C especially between 6.85 – 20 m ice depth (depths where production from negative muon capture dominates). One simply cannot improve the model-data fit by uniformly scaling the measurements to account for ^{14}C in organics. The only way to fit the shape of the data/measurement curve is to lower the ^{14}C production rate from negative muon capture and add some ^{14}C production rate from fast muon to improve the fit at depths >30m. It is still theoretically possible to fit the data if the *in situ* production from negative muon capture and fast muon produce *in situ* ^{14}C -bearing organic materials at different ratios relative to total expected ^{14}C . In other words, if somehow a lot of ^{14}C

produced via negative muon capture becomes ^{14}C -bearing organics to compensate for model-data mismatch between 6.85 – 20 m ice depth. However, as our data show that the two muogenic reactions produce constant ^{14}CO , $^{14}\text{CO}_2$, and $^{14}\text{CH}_4$ ratios, we consider this explanation as unlikely.

We included more discussion of several hypotheses that could possibly explain the disagreement between the ^{14}C production rates inferred from our data and those of Heisinger et al. (2002). Although we consider it unlikely, it may in principle be possible that ^{14}C in organics might account for a very large fraction of *in situ* ^{14}C from muons (enough to compensate up to a factor of 6 for negative muon capture and a factor of 3 for fast muon reaction). Second, it may be possible that the overall probability for ^{14}C production from ^{16}O in ice is much lower than in quartz – as the laboratory irradiation done by Heisinger et al. (2002) used quartz (and not water/ice) as the target compound. Third, it may be possible that the power factor used to scale the production rates from high energy muons used in Heisinger et al. (2002) laboratory experiment to lower average energy muons in the natural setting is incorrect. Unfortunately, we lack the experimental setup to characterize ^{14}C in organics. We also do not have the expertise or the analytical capability within our current author team to properly explore the nuclear chemistry aspects of this. As a result, we present this discrepancy as an open question to the greater scientific community to be pursued in future studies and focus the manuscript more specifically on constraining the ^{14}CO , $^{14}\text{CO}_2$, and $^{14}\text{CH}_4$ production rates in ice from muon reactions.

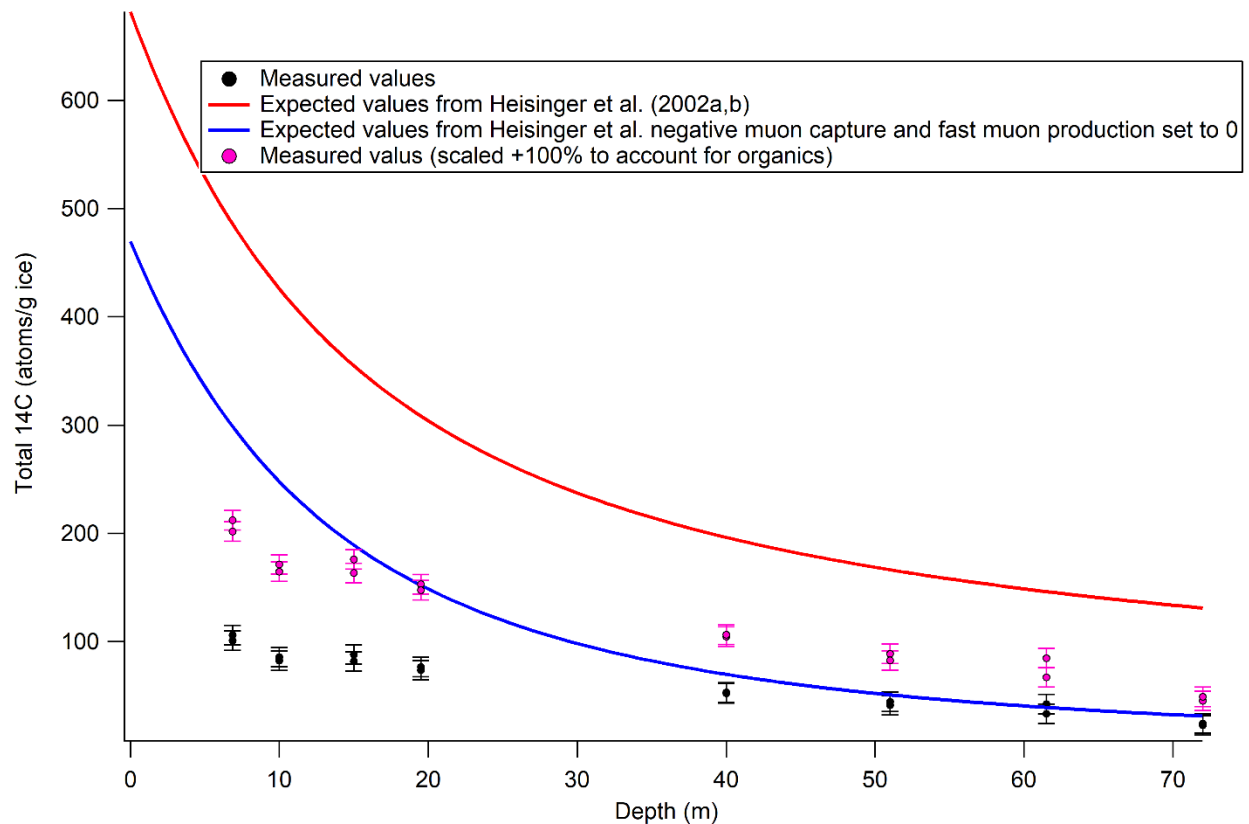


Figure 1. Model-data disagreement between measured total ^{14}C values, measured total ^{14}C values (+100% to account for organics), and expected total ^{14}C values from Heisinger et al., with and without ^{14}C production from fast muon mechanism.

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