Responses to referee

Major comments & summary

Greg Balco

Overall the paper is much better as regards exploring the differences between these data and other estimates of muon production rates. The only exception is the abstract. In this sentence:

"Assuming that the majority of in situ muogenic 14C in ice forms CO2, CO, and CH4, […] for negative muon capture and fast muon interactions, respectively"

the usage of "are too high" indicates that the authors are saying that the other measurements are wrong. In fact, this is not established, it is only established that different measurements using different approaches give very different results (as is well described in the last sentence of the abstract). Thus, this sentence should be reworded to say something like "our measurements disagree with previous measurements using different approaches" rather than "the other measurements are too high."

Nat Lifton

First, all isotopic results should be re-calculated using the methods of Hippe and Lifton (2014), that are specific to the in situ 14C AMS analyses as opposed to traditional radiocarbon measurements. The authors use measured pMC as the basis of their calculated 14C concentrations, which relies on a number of assumptions for organic radiocarbon that don't apply to in situ 14C systematics. Hippe and Lifton (2014) provides recipes for converting the pMC values to the raw measured isotopic ratios – the differences are often small but can be significant depending on the measurement details. Any older in situ 14C data to which these are being compared should also be recast appropriately per Hippe and Lifton (2014). This includes the Young et al. (2014) spallation production rate dataset and derived reference production rate.

Second, the authors should clarify whether production rate scaling is done using the LSDn nuclide-specific model from Lifton et al. (2014). If scaling does not use LSDn, then the authors should recalculate scaling factors using that model, as it accounts for the effects of both the cosmic ray flux and excitation functions for production of in situ 14C.

I also think the authors should also include the formulations of Balco (2017) in their discussions (given that it's in the reference list already) – a study that uses a different approach to fit the muon production proportions from the in situ 14C depth profile data of Lupker et al. (2015).

Finally, there are several points in the manuscript where the authors assert and/or imply that the Heisinger et al. (2002a, 2002b) muogenic production predictions are incorrect (as well as those from Lupker et al., 2015, and Balco, 2017, which support them) – Abstract, conclusions, and section 4.2 around Line 470, for example. I think it would be more correct to frame the comparison of this study's findings with those other predictions as a discrepancy that merits additional investigation, particularly with regard to comparisons of ice vs. rock measurement techniques. To their credit the authors do mention the need for future investigation to resolve this discrepancy, but I would just ask that they soften the assertions of correctness.

We thank both Greg Balco and Nat Lifton for their thoughtful and constructive comments. We have further adjusted the abstract, discussion, and conclusion of the manuscript to remove all suggestions that prior work in quartz may be incorrect and simply highlighting the apparent disagreement. We also further clarified the specific muogenic production model ("model 1A" from Balco 2017, which is a MATLAB implementation of the model first presented in Balco et al., 2008) that we used to constrain the 14C production parameters, as well as the scaling model we used to scale the surface muogenic production rates at our study site (Taylor Glacier) to the typically reported SLHL (sea level high latitude) production rates ("LSDn" – the nuclide-specific scaling provided in Lifton et al. 2014). With regards to ^{14}C calculations, we use the same method as Petrenko et al. (2016), and as shown in the supplement of Petrenko et al. (2016) this method is compatible with Hippe and Lifton (2014).

Detailed comments

Greg Balco

I have one other minor comment about the line 595+ area. This is a bit confusing, because it applies only to differences between ice measurements and lab irradiation measurements, but not to differences between ice and quartz data in natural settings. This paragraph requires clarification as to whether these possibilities are intended to explain differences between the ice data and the laboratory irradiations (which is possible), or between the ice data and the quartz data (which is not possible). Section 4.3 of the previous draft of the paper is much clearer on this subject.

We removed the mention of f_C and f_D in this section (like Greg Balco, we also cannot find a good reason to believe that f_C values are incorrect; and the Lupker data do seem to support the f_D value) and instead added some discussion on the possible effect of α, following Nat Lifton's comments.

Nat Lifton

"First, all isotopic results should be re-calculated using the methods of Hippe and Lifton (2014) […]" and other references to Hippe and Lifton (2014)

We have added in page 9 line 275

"The amount of ^{14}C molecules per gram ice for ^{14}CO, $^{14}CH_4$, and $^{14}CO_2$ (Table 1) is calculated using the same method as in Petrenko et al. (2016) and is consistent with Hippe and Lifton (2014) formulations for in situ ^{14}C concentrations."

I also think the authors should also include the formulations of Balco (2017) in their discussions (given that it's in the reference list already) – a study that uses a different approach to fit the muon production proportions from the in situ 14C depth profile data of Lupker et al. (2015).

We added discussions and references to Balco (2017) in this response (see below), page 4 line 117 in the Introduction and later in page 17 line 560.

Are you also considering protons? There is a small but significant flux of them at the Earth's surface. Way less than neutrons, yes, but I don't think you can ignore them.

We changed all mention of "neutron-induced spallation" into "nucleon-induced spallation" or just "spallation". We don't explicitly consider protons, but we use the empirically determined spallation related ^{14}C production rate from Young et al. (2014). Because it is empirically determined, then it should include a small amount of spallation-related in situ production due to protons. However, we only consider the attenuation length of neutrons (150 g cm^{-2}) to scale the spallation related production rate with depth.

Regardless, since we are only fitting data that are 6.85m deep and below, the amount of ^{14}C production from spallation is not relevant and does not change our results.

What is "present" - the LSD model uses 2010 typically, while normal radiocarbon is referenced to 1950 as "present" - clarify - should be using Hippe and Lifton (2014) to calculate in situ 14C concentrations.

We clarified that when we say “before present”, we mean before 1950CE, which is a convention in radiocarbon and paleoclimate studies. However, as noted above, our ^{14}C calculations are fully consistent with the Hippe and Lifton (2014) approach

They decrease with depth as well, just more slowly than nucleon attenuation, thus eventually overtaking nucleon production with increasing depth due to the different attenuation characteristics. This wording makes it sound like their production increases with depth. Clarify

We clarified the sentence on page 3 line 88-91:

“Nucleon-induced spallation dominates the ^{14}C production at the surface but is quickly attenuated with depth, while the relative contributions from the two muon mechanisms are lower near the surface but dominate at larger depths.”

Our Fig.1 also already clearly shows that production rates from all mechanisms (neutrons and muons) decline with depth

Balco (2017) also re-fit these data and came up with a slightly different result - should cite that as well, along with his predictions. (Balco, G. (2017). Production rate calculations for cosmic-ray-muon-produced 10Be and 26Al benchmarked against geological calibration data. Quaternary Geochronology, 39, 150–173. https://doi.org/10.1016/j.quageo.2017.02.001)

We added citation of Balco (2017) and clarified that we actually used the Balco (2017) “model 1A”.

What is the value of alpha that is being used here? See Balco 2017.

We clarified that we use alpha = 0.75 (page 5 line 147), and that we also trial alpha = 1 towards the end of the discussion section.

I'd be interested to see results with the Balco (2017) calibrated parameterization instead of the Balco et al. (2008) version.

We already use the Balco 2017 “model 1A” for ^{14}C production profile. Balco (2017) fitted Leymon High ^{14}C data using both his “model 1A” and “model 1B” (model 1B uses the latitude/longitude scaling from the Lifton et al (2014) LSD model). He tuned the f* parameter, rather than f_{tot} like Lupker et al. (2015) or this study, but found values that are close to Heisinger et al. (2002) of f* = 0.116 (model 1A) and f* = 0.114 (model 1B) while the f* in Heisinger is 0.137 and the f* from Lupker (when converted from their estimate of f_{tot}) is 0.134. As such, Balco (2017) estimated ^{14}C production rate from negative muon capture that is ~84% that of Heisinger et al.

We cannot use Balco (2017) calibrated parameterization since we are tuning the same parameters to fit our data (f_{tot} and σ_0), but we can compare the results. We estimated a production rate from negative muon capture that is ~16.6% that of Heisinger et al./Lupker et al. and ~19.8% that of Balco (2017). As such, the difference in fitting and ^{14}C production model used by Lupker et al. (2015) and Balco (2017) are small compared to the discrepancy between our study and the observed ^{14}C production rate in quartz from negative muon reaction (Lukper et a. 2015; Balco 2017; Heisinger 2002).

We note further that Greg Balco did not have any fundamental concerns about the model / parameterization we used to fit our data, and (in the first round of review) repeated our analysis using his own code, arriving at results that were similar to ours.

What value are you using here? Are you just taking it from that paper? Should be recalculated using nuclide-specific scaling from Lifton et al. (2014) (LSDn) and Hippe and Lifton (2014) - Hippe, K., & Lifton, N. A. (2014). Calculating Isotope Ratios and Nuclide Concentrations for In Situ Cosmogenic 14C Analyses. Radiocarbon, 56(3), 1167–1174. https://doi.org/10.2458/56.17917

We clarified these in page 14 and line 439-447

"The expected 14C concentration in the ice is given by the differential equation (Eq.11) where P'n is the 14C spallogenic production rate from Young et al. (2014) (12.0 ± 0.9 atoms g quartz-1 yr-1 at the surface), first scaled to SLHL production rate in ice (20.0 ± 1.5 atoms g ice-1 yr-1 at the surface) accounting for the number of 16O atoms per gram in ice vs. quartz (variable 'N', Eq. 3) (Petrenko et al. 2016), then to production rate at Scharffenbergbotnen site (1173m above sea level) using the Lifton et al. (2014) "LSDn" nuclide-specific model (P'n = 71.2 ± 3.6 atoms g ice-1 yr-1 at the surface)."

This sounds like the ice was being sublimated while adding the gases separately. Could there be some effect if the gas is contained within the ice beforehand?

Unfortunately, the ice core community has not found a way to make a "standard ice", i.e., synthetic bubbly ice that contains the standard gas of our choice in the bubbles. Performing simulated extractions with bubble free ice in presence of a standard gas is usually the accepted method for investigating procedural effects in analyses of gases in ice cores (e.g., Sperlich et al., 2013). As for the question about preservation of CO_2 and $^{14}CO_2$ in the bubbles, we know from multiple prior studies that CO_2 is well preserved in Antarctic ice. $^{14}CO_2$ is produced *in situ* from cosmic rays but to our to knowledge there is no known mechanism that removes $^{14}CO_2$ except for natural radiocarbon decay (which we account for in our calculations).

This argument seems a bit weak - the authors are first arguing that the Van der Kemp methods can produced biased results using the dry extraction method but then point to the good agreement with their results as supporting their results. What if their techniques are introducing an as-yet unrecognized bias as well? I'm not saying that's likely but seems like it should at least be addressed.

We addressed this later on in page 16 line 509-531.

The idea that mechanical extraction might bias ^{14}C measurements in ice have been around for a while (Van Roijen et al., 1994). This is discussed by Van der Kemp et al. (2002) themselves. They mentioned that the disagreement between ablation rates inferred from their measurements and ablation rates based on stake measurements could be due to incomplete release of ^{14}C from the ice grains. However, when we transferred our production rates to Van der Kemp et al. (2002) data, we get good agreement with their measurements. This seems to suggest that instead of their measurements being biased low, it is more likely that the discrepancy between expected ablation rate based on ^{14}C measurements vs. actual ablation rate from stake readings is due to the Heisinger et al. production rates not being applicable to ice. We also discussed why it seems like dry extraction is a valid method for $^{14}CO_2$ measurements – based on our experience measuring ^{14}C in both firn air and firn samples, it seems like the *in situ* produced ^{14}C migrates very quickly into the gas phase and out of the ice grains. In the firn it means that the in situ 14C ends up mainly in the open porosity, while in bubbly ice 14C ends up in the air bubbles. As such, the dry extraction method using a “cheese grater” per Van der Kemp et al. (2002) seems to be a valid method for ^{14}C measurements in bubbly ice.

Are there similar long-term data from near Scharfenbergottnen? I know the EDML core site is nearby.

We added the sentence in page 17 line 543:

“The EDML ice core drilled in the region near the Scharffenbergbotnen blue ice area also does not show large climate variability over the Holocene period (EPICA Community Members, 2010).”

YES - Heisinger uses alpha=0.75, but the authors should perhaps also consider the Balco (2017) approach in which alpha is assumed to be 1.0. What effect might that have on their predictions?

We added a sensitivity test when alpha =1 and discuss it in page 19, line 610 onwards and show the fit in Fig. S13 of the supplementary materials. In short, having alpha =1 move our best-fit parameters slightly closer to the Heisinger et al. values, but still not nearly enough.

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