

Second review, Diaz and others, Esurf

This paper is greatly improved from the first draft. I very much appreciate the authors' effort in seriously revising the paper. Most of the major problems from the first draft have been corrected. Nice job.

From the perspective of this review, though, what that means is that it is now possible to actually understand the scientific content of the paper and review it in detail. So in this review I've actually done that. The result is that, although the paper is really, massively, improved in many ways, there's still one problem, which is the discussion of the relationship between Be-10 and NO₃ concentrations. This still has some elements that are misleading because they are oversimplified and in part appear to be incorrect. So that part isn't acceptable for publication yet and needs to be fixed. I discuss this in way too much detail later.

So, going through the paper in order,

1. Introduction. This is enormously improved. It is basically fine now, although it would be helpful to better orient the reader (who perhaps has skipped over the abstract) by beginning the introduction with a sentence describing the actual research in the study, like "This study reports concentrations of atmospheric fallout constituents in soils in the southern TAM. These data can be used to understand soil age and disturbance frequency, which are biogeographically important because one of the most intriguing questions..."

2. Background section. Also much improved. Great. Minor points:

In lines 98-99, do you mean lower elevations, like water is running downhill for a significant distance, or greater depths in the soil? Clarify.

In line 111. "Considerably fewer" sounds strange to the reader here because you are saying there are fewer studies in the CTAM than in NTAM/NVL, but you cite more studies for CTAM. I would just remove the "considerably fewer" and note that there have been scattered exposure-dating studies all over the Transantarctic Mountains.

3. Methods section. This is much better. Could use minor clarification in a couple of places, as follows:

Line 151. Perhaps clearer to say "...to represent soils likely to have been covered during the LGM and exposed by more recent ice margin retreat."

Line 189. The usage of 'dz' is mathematically strange here. dz is just a generic differential in depth, it is not a parameter needed to evaluate the equation, so it is unclear what 'highly dependent on dz' means. I think what the authors are trying to say here is that the concentration gradient dn/dz depends on what D is and also varies with time, and D is unknown, so it is not really possible to calculate dn/dz. In addition, in line 187, the authors use 'concentration gradient' to describe

d^2n/dz^2 , which is confusing because 'gradient' usually means the first derivative of something (dn/dz). The second derivative (d^2n/dz^2) would typically be described as 'curvature.' In any case, both of these points give the impression of carelessness and this section needs to be carefully checked to make sure it makes mathematical sense.

Lines 211-12. This doesn't seem to make sense, because if Be-10 is supplied from the surface, the concentration has to decrease with depth at some point, no matter what. I believe the difference the authors are trying to point out is that in a normal soil one would expect a fairly smooth decrease, but in a periglacial soil one might expect a well-mixed active layer with constant concentration abruptly overlying a frozen layer with much lower concentration. However, this is not what this sentence says. In any case this sentence is oversimplified to the point of causing confusion, and it's not really very important, so I would remove it.

4. Results. The basic description of the results, up to section 3.2, is good. At this point, however, we get into the subject of the relationship between meteoric Be-10 and NO₃ concentrations, which needs work.

Basically, the approach in this paper as far as I can tell is to assume that Be-10 and NO₃ are both supplied by fallout, and both are supplied at a fairly constant rate, which in turn means that the ratio of their depositional fluxes is also fairly constant. Call this assumption 1. OK, this makes sense, and if this is true and the soil profile is a closed system (assumption 2), they can both be considered conservative tracers. So, up to now, assumptions 1 and 2 predict that total inventories of Be-10 and NO₃ in the soils should both increase monotonically with exposure time and will be highly correlated.

Correlated inventories of Be-10 and NO₃ that both increase monotonically with exposure age is, of course, what we expect, not only from first principles but due to the Graly study at Mt. Achernar, which showed these amazing correlations between exposure age and salt concentrations in sediments in a blue ice moraine. That is exactly how it is supposed to work and, like the authors, I want it to work that way in this study. Unfortunately it doesn't. I thought about this issue a lot in putting together this review – because, as noted, I would like this to work – but when you look at the actual observations in this study, the only possible conclusion is that it does not work this way. The following section of this review explains why in probably too much detail.

So, more specifically, the Be-10/NO₃ relationship should be close to linear for relatively young soil ages, but as soil age increases enough that Be-10 decay is important, the slope of the relationship will change as the Be-10 inventory asymptotically approaches an equilibrium value where deposition is balanced by radioactive decay. Stated in math, this means that the relationship between Be-10 and NO₃ inventories is given by parametric equations in t (time, yr) for the NO₃ inventory I_{NO_3} (mol/cm²):

$$I_{NO_3} = Q_{NO_3}t \quad (1)$$

where Q_{NO3} is the depositional NO3 flux (mol/cm2/yr),
and for the Be-10 inventory I_{10} (let's also use mol/cm2):

$$I_{10} = \frac{Q_{10}}{\lambda_{10}} [1 - \exp(-\lambda_{10}t)] \quad (2)$$

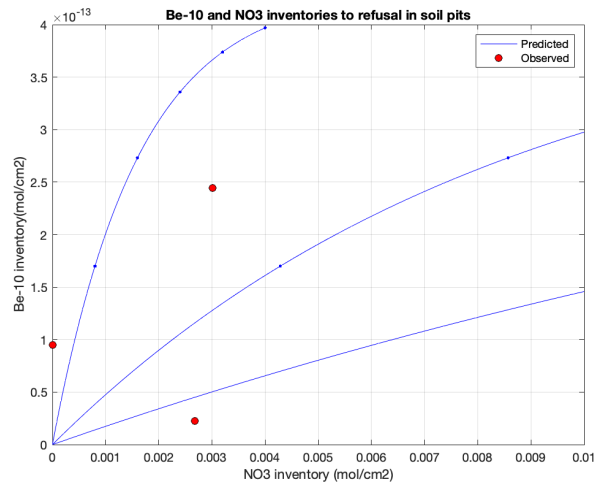
where Q_{10} is the depositional flux of Be-10 (mol/cm2/yr) and λ_{10} is the decay constant for Be-10 (4.99 e-7 /yr).

So what just happened there was we went from two simple assumptions to a quantitative prediction for how measured inventories of Be-10 and NO3 should be related. There are some additional side predictions that will be important later. One is that the Be-10/NO3 ratio should be constant and equal to the deposition flux ratio for young soils, and will be lower than the depositional ratio for old soils because of radioactive decay. The Be-10/NO3 ratio can't be higher than the depositional ratio with these assumptions.

Continuing, the authors then make a third assumption, which is that Be-10 and NO3, once deposited, are transported together. If this is true, then not only the inventories, but also the concentrations, will be highly correlated. The slope of the relationship could vary in old soils, or in parts of the soil profile that have not exchanged Be-10 with the atmosphere for a while, because of Be-10 decay, but all three assumptions together predict a positive correlation between measured concentrations. This assertion is the basis for what the authors do next, which is to further assert that if they can establish a correlation between measured Be-10 and NO3 concentrations, they can then use this correlation to estimate Be-10 concentrations and therefore inventories in samples where only NO3 was measured. They go on and do this, and many of the apparent exposure ages that are eventually presented in the paper are from estimated Be-10 concentrations.

So far, the only problem with the paper is that the authors have not actually clearly stated the assumptions that led to their assertion that they can use NO3 concentrations to predict unmeasured Be-10 concentrations. However, I am now going to point out a lot of other problems.

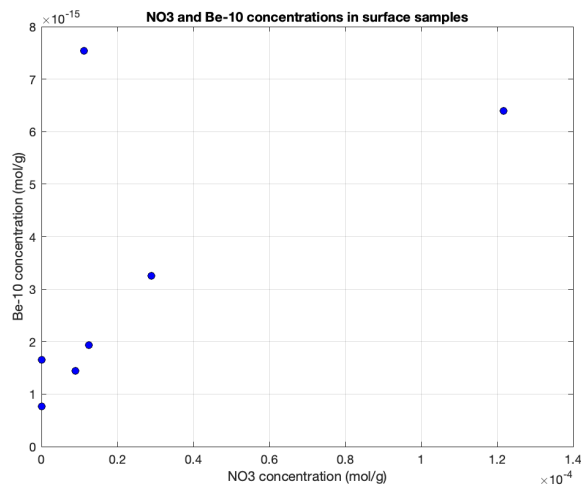
Problem 1 is that assumptions 1 and 2 predict a specific quantitative relationship between Be-10 and NO3 inventories. Both inventories were measured at three sites. The following figure compares these inventories to the predicted relationship from the equations above.



The blue lines are the predicted relationships from the equations above. The different blue lines use different values for the NO₃ deposition rate from the Graly and Adams papers. Of course these papers all use different units so I hope I got all the conversions to mol correct, although the order of magnitude variation indicates that I might have made a conversion error. Regardless, the curves show the shape of the relationship and hopefully at least the right order of magnitude.

The important thing here is that the observations look nothing like the predictions. One sample has lots of Be-10 but almost no NO₃. Two samples have about the same amount of NO₃ but wildly different amounts of Be-10. Not even any two of the observations can be fit to any one growth curve. These observations are impossible if assumptions 1 and 2 are true, which implies that, in fact, these assumptions are not true. Of course, there are some reasons that you could have both the assumptions and these results – you could have incomplete measurements of the inventories, or you could have very large and wildly different inherited concentrations of either or both...if you can have completely arbitrary inheritance you can have whatever you want. But no matter what, these results clearly show that in sharp contrast to our expectations and to the Graly study, observed Be-10 and NO₃ inventories are NOT correlated between sites, no matter whether we think they ought to be or not.

Now consider Be-10 and NO₃ concentrations. This plot shows NO₃ and Be-10 concentrations in surface samples where both were measured.



These are also not correlated. Again, it could be possible to have these results and all the above assumptions if you can have inherited concentrations be whatever you want. But if the existing set of measurements of NO₃ and Be-10 in surface samples are not correlated, then any theory that predicts that they are correlated can clearly not be used to predict Be-10 concentrations where they were not measured.

The point of all this is that the data clearly show that whatever we think ought to be happening, Be-10 and NO₃ inventories are not correlated, and Be-10 and NO₃ concentrations in surface samples are not correlated. Therefore, there is zero reason to believe that the authors' attempt to predict Be-10 concentrations or inventories in samples where they were not measured is correct. Of course it might be correct by accident, but this seems unlikely.

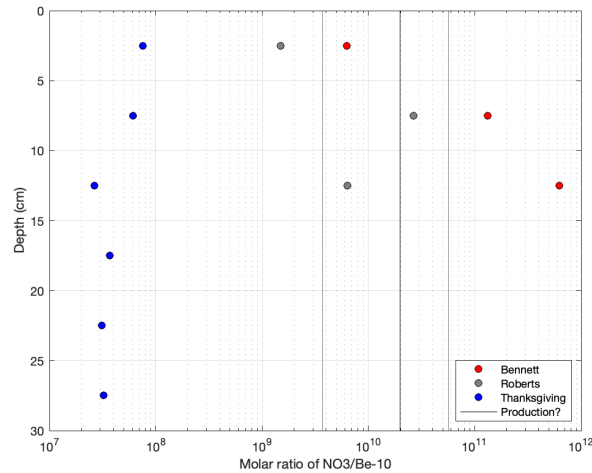
Note that the authors actually tried to do this in a more complicated way. They showed that within each of the three depth profiles where both Be-10 and NO₃ were measured, they could be related by a power-law relationship. These relationships were different for all three soils. Then they used these relationships to predict Be-10 concentrations at unmeasured sites simply by asserting which unmeasured soil was most like which measured soil.

This procedure includes several unjustified and unacceptable leaps of faith. First, there is no physical basis that would lead us to expect a power-law relationship. Thus, fitting a power law to the data makes no sense. Second, there is definitely no physical reasoning that the authors have presented that would predict both positive (as in two soils) and negative (as in one) slopes. In contrast, basic assumptions, as discussed above, predict a linear relationship in young soils and asymptotic in old soils, always with positive slopes. As far as I can tell, there is especially zero physical explanation for why there should be an inverse power law relationship. Thus, the power law fits just make no physical sense, and there is no reason to believe they have any physical significance. In fact, there are many reasons to believe that they don't have any physical significance. I conclude that they

don't have any physical significance, so predicted Be-10 concentrations based on these fits are also unlikely to be correct except by accident.

A further point is that if you just correlate the two things for each depth profile linearly without log-transforming, you get basically the same r-squared. So in addition to there being no apparent physical reason that I could discern for using a log-log fit, there is also not really any empirical reason to think that a log-log fit is any better than a linear fit.

Finally, consider the observed Be-10/NO₃ ratios in the depth profiles. Those are shown in this figure.



The vertical gray lines are various guesses at the production ratio from the Graly and Adams results. The ratios from Roberts and Bennett are somewhere in the expected range of production ratios, which is more or less consistent with what we expect for dry soils that are closed systems for both things. The increase with depth, however, does seem to indicate that even in dry soils Be-10 and NO₃ don't move together, NO₃ is more easily moved down.

On the other hand, remember in the discussion above we showed that Be-10/NO₃ ratios above the deposition ratio, which is the same as NO₃/Be-10 ratios below the deposition ratio, are impossible in a closed system without inheritance. However, we see such impossible ratios at Thanksgiving Point. There are two obvious ways to explain this. One, nearly all the Be-10 and none of the NO₃ could be inherited. This might be possible given the comparison with the in-situ Be-10 exposure ages. Two, water leaching of NO₃ has taken place in the relatively warmer and wetter soil at Thanksgiving Point. Both of these possibilities invalidate pretty much all the assumptions used to argue for a correlation between NO₃ and Be-10. If most of the Be-10 is inherited and different sites can have different inheritance, there is no expectation of a cross-site correlation between Be-10 and NO₃. If water leaching of NO₃ has taken place at some sites but not others, then, likewise, clearly it is not possible to apply any fixed relationship between NO₃ and Be-10 concentrations

across different sites. Either one of these cases highlights that the authors' approach to predicting Be-10 concentrations at unmeasured locations does not have merit and is nearly certain to be incorrect.

The summary of this rather long discussion is that there is exactly zero observational support for the authors' scheme for predicting Be-10 concentrations from NO₃ concentrations, and, in addition, zero theoretical support for some aspects of the scheme like the decision to use a power-law fit. In fact, comparison of theory to observations indicate that this should not work, except by accident. Even if the basic assumptions are correct, the expected correlation is not present due to some combination of background effects and NO₃ leaching. Thus, the predicted Be-10 concentrations for sites where only NO₃ was measured are incorrect. The authors must remove this aspect of the paper and consider only Be-10 and NO₃ concentrations that were actually measured.

Correcting this problem will involve:

- entirely removing section 5.3.2.
- Removing the results of the calculations in 5.3.2. from all tables and figures, so that only measured data are shown throughout the paper. That includes removing Table 4.
- Removing and rethinking any discussion that relied on the estimated Be-10 concentrations.
- Removing the discussion of this subject in section 6.2. Basically, as detailed at length above, the statement that "...we conclude that NO₃ appears suitable for relative age dating..." is not supported by the observations in this study. I agree that NO₃ ought to be suitable for relative dating, but unfortunately the evidence in this paper rather indicates the opposite. There are many similar remarks in this section to the effect that "the correlation between Be-10 and NO₃ is widely applicable in hyper-arid soils", etc. These might be true, but they are not proven by anything in this paper.
- Removing discussion of this subject from the abstract and conclusions.

This material could be replaced by a figure or two showing that there is no discernable correlation between NO₃ and Be-10 inventories or concentrations, and a couple of sentences speculating on why.

A couple of final notes on this aspect of the paper here. First, I want to assure the authors that the paper will be just as good, in fact, better, without this element of the paper. I get the idea that the authors have an interesting data set that is difficult to easily interpret, so they are kind of struggling to make the paper have some elements that they perceive as more significant than just a set of empirical observations. However, the authors need to keep things in perspective here. These are a set of totally new observations from a part of Antarctica and a type of setting that no one has looked at from this perspective before. Most (possibly all) of the existing meteoric Be-10 data in Antarctica are from really old dry soils that have been ice-free for a really long time. The data in this paper are from much more complex sites with a complicated history of ice cover and exposure. If this paper presents the data, points out that they are much more complicated than we expect from

simple relationships found at more simple sites, and then stops, that is a big contribution. Instead de-emphasizing the complexities and simply asserting without cause that we know what is going on with these data doesn't make the paper better, it makes it worse. A clear and comprehensive observational study is extremely valuable. It is not necessary to try to explain everything, or to add speculative material, to increase the perceived interest of the study.

Second, if you had asked me before I read this paper whether there would be a strong correlation between Be-10 and NO₃ in these samples, I definitely would have said yes. The results from the Graly paper about Mt. Achnar are extraordinarily clear in this regard, and I would have expected a similar situation here. The actual observations that show no correlation between sites are amazingly different from the Graly results. This clearly indicates that there is something that we are missing and the setting is much more complicated than we thought, and it is extremely clear from these results that the expected behaviour is wildly oversimplified. At present, this paper just asserts that the expected behaviour is true even if the observations don't agree with it. It would be much more valuable for this paper to highlight that apparently the expected behaviour is very oversimplified relative to reality and that we are missing important things, most likely having to do with variable inheritance and/or NO₃ mobility in water.

Coming back to the overall paper, the rest of the discussion having to do with the apparent exposure ages is in pretty good shape. A few comments on the discussion:

The sentence in lines 336-339 doesn't contribute anything and should be removed. Start with "The Shackleton Glacier region..." It seems like the authors had some trouble getting started in this section...they should just simplify things by starting right into the observations they want to highlight.

Area around line 364. It seems to me the easiest explanation for the greater-than-LGM apparent meteoric Be-10 ages for the lower-elevation covered-at-LGM sites is just that the inherited Be-10 inventory is large compared to the relatively short exposure time at this site. Inherited Be-10 equivalent to 10,000-100,000-ish years exposure seems unsurprising.

Area around line 425. As discussed in the first review, the authors need to be more careful not to mix up evidence for aridity with evidence for ice sheet change or lack thereof. These are not at all the same thing.

Line 436. I don't understand why a shallow active layer implies that Be-10 was able to migrate deeper into the soil in the past. How do we know that it hasn't been the opposite – the active layer was shallower in the past and has been thickening over time?

Line 448. I don't understand this argument. You can get to an increasing NO₃/Be-10 ratio lots of ways. As is evident in many otherwise dry soils in Antarctica, there is commonly a subsurface maximum in salt deposition just because of brief wetting by snow events moving salts below the surface and depositing them when the water sublimates. As NO₃ has more pathways for mobility than Be-10, it seems much easier to explain this by enhanced NO₃ transport instead of by some

complex inheritance effect.

5. Conclusions. Besides the need to remove discussion of estimating Be-10 from NO₃ in lines 489-492, I only have one comment here. As noted above, I don't understand why the presence of soil ice requires a past warmer climate. Soil ice can easily form and remain under equilibrium conditions. Explain?

6. Other items:

As discussed in my request from the editor for a revised table, it was unnecessarily difficult to get all the data from the tables that were provided, because it was hard to connect which exact samples from which depths did or did not have Be-10 and/or NO₃ measurements. The table that the authors provided in response to my request is much clearer. The authors should use that table instead of the existing Table 1, and leave the details of the Be-10 measurements in a separate table, which would be basically the current Table 3.

7. Finally. OK, that's it. As you can see from the fact that most of this review is devoted to explaining why the NO₃/Be-10 correlation approach appears to be incorrect, I thought about this for some time and tried to convince myself that this would work. Again, based on first principles and the Graly paper, I would expect a strong relationship. However, it just isn't there. Of course this is interesting because it might show that NO₃ seems to be behaving conservatively in some soils but not others, in which case these are not all dry soils and the local microclimate matters. But it makes the approach of predicting Be-10 from NO₃ untenable. It's just not right and it needs to be removed before the paper is suitable for publication.

Otherwise, I want to make sure to come back to the main point that the authors have done a great job of revising this paper. It is enormously better than the first draft, which is admirable. Most authors do not display this level of commitment to revising a paper, and I appreciate this because I would like to see this data set published. But the fact is that the paper does still have this one problem that needs to be corrected.