Dear editor, dear reviewers,

We have now completed a revision of our manuscript "U-Th and 10Be constraints on sediment recycling in proglacial settings, Lago Buenos Aires, Patagonia". We would like to acknowledge the interesting and exhaustive reviews by the two reviewers, that helped us improve the manuscript. We did our best to address all comments and suggestions.

Below we address our answer to more specific comments as well as the modifications that we made to the manuscript accordingly. We hope that it will help clarifying various addressed points.

Best regards

Antoine Cogez

Anonymous Referee #1

This is a very interesting and appropriate paper for ESD. There are enough new 10Be data and as far as I know (and the authors know) it is the 1st U-Th data and application in southern South American on sediment movement from source to sink, as they say. Hence, this is a novel contribution. I think my comments are moderate to major and not difficult to do.

Some major (or moderate?) comments.

1. One of my most significant comments is that one really needs to be an expert in U-Th series measurements and approach to really follow details of the Methods and Results (not so much Discussion). So much of it may remain largely inaccessible to the non-expert, which is a shame, given all their hard-work!!

I am actually not sure how much they can do about this, as the paper is not an ap- propriate place for a 101 lesson on the U-Th comminution and sediment approaches. On the other hand, I think there are few minor things they can do which I suggest or highlight below (e.g., see figure captions) which will at least help the reader a bit not familiar with these methods. These mainly focus around i) explain figure captions a bit better and ii) perhaos more info in supplement information section (a user friendly flow chart or Table of terms, as suggested below?) iii) some more labels on the figures (see below) and iv) a few places in text can add a few more words (see below)

I should also highlight to the Editor that I am not an expert on the U-Th series and application. I cannot really evaluate this in detail.

We recognize that U-Th series disequilibrium is conceptually difficult to grasp and it is difficult making it clear in a new paper about the topic. We added a section in suplementary materials in order to make it clearer. We hope the basic understanding can be achieved in the paper, and that this added section helps to orient the reader to find more detailed informations about U series.

Moreover we address your different detailed points below in the list of precise comments you addressed, which helped improving the clarity of those points.

2. Some more info is needed on setting and context of samples. i) Specifically some photos of the samples so reader can see the context. This includes photos of the cosmo samples. If possible, they need to provide a photo of the outwash with the moraine behind it (showing how they link); if they do not have such a photo, maybe the closest thing they have. At least one or two photos of the silts they sampled for comminution – are these from a pit? A road cut? No detailed geologic context is provided to the reader to evaluate their results. ii) I think also a more zoomed in version of the map over the sampling area. It is hard to appreciate from their short discussion. See comments below also for Figure 1. iii) Can they provide a strat section, even schematic or simplified? Minor - also, on map – show Deseado boulder location, since mentioned in the text.

Some pictures were added in the suplementary materials.

3) Also, I recall there were two telken boulders in Kaplan et al. which were used to de- rive maximum erosion rates – they are very maximum erosion ages, and very minimum exposure dates, but the authors may want to mention (?) these data because they sup- port i) need to date outwash on these old deposits as they do (and Hein et al do) and ii) the erosion rate they derive below shows these Telken bouder measurements were indeed maximum erosion rates as Kaplan et al. 2005 stated. More on this below.

It is indeed interesting that both our estimated deflation rate and the boulder erosion rates from Kaplan (2005) are comparable (maximum values at least). However can we compare boulder erosion rates and outwash surface deflation ? We prefer not mentioning it since we remain unsure both can actully be compared. It could blur the understanding about the interest of taking outwash cobbles instead of moraines boulders.

4. Apparently, Douglass had measured Deseado boulders in his PhD research that are unpublished, except for abstracts. One abstract that is particularly relevant that au- thors may want to know about, is a GSA abstract with recalculated erosion rates; these are close to what they derive on the outwash I recall. The abstract is cited in Hein et al 2017. They may want to refer to this, as it is another measurement that supports their finding for very low erosion rates in the area, much lower than the 'max rates' pre- sented in Kaplan et al. 2005. The reference (can be looked up) is GSA Northeastern Section - 42nd Annual Meeting (12-14 March 2007) Paper No. 24-2: CONSTRAIN- ING BOULDER EROSION RATES AND AGES OF MID-PLEISTOCENE MORAINES, LAGO BUENOS AIRES, ARGENTINA (again see Hein paper for references).

Same comment than above, we might be wrong but we prefer not mentioning it since we are not sure the comparison is appropriate.

5. Can they say if there are any changes in the U-series data over the time of the moraines and sediments listed in Table 3? They sampled a nice profile or transect from old to young moraines, spanning almost 1 million years; this is unique aspect of their study (has this been done even anywhere?). They do not really discuss this at all (unless I missed it). Is there a lot more information here they can highlight or speculate about? For example, are the U-Th results

different for moraines from stage 8 and 2, compared with their data on the older telken and deseado sediments Or vice versa? A point is that I am wondering if they can elucidate changes as Southern Andes are eroding down (they discuss Kaplan et al. 2009 mechanism, for example, in this context).E.g., glacial erosive products produced and recycled more and more. . .?

One assumption of the model we use is that the recycling time is the same for all samples, so we cannot discuss a trend in the evolution of the recycling time over the last million year. First, given the number of uncertainties in the problem, we need to solve a single recycling time. Second we can argue that this hypothesis of a constant recycling time over the last million year remains reasonable : see our response to the third main point of reviewer 2 for a more detailed discussion on these explanations. The main point is that the glacial valleys and the overdeepening (i.e. the main sediment sinks, responsible for increasing the sediment transport time) were most probably already carved by 1 Ma. However it appears that this was unclear, so we tried to make clearer in the revised version of the manuscript by adding a paragraph in section 2.3.5.

Other, detailed comments corresponding to pages (and providing more info on above comments):

page (p.) 2 line 1 – what do they mean by 'remobilization of moraines' - this sounds catastrophic ! Please rephrase to be more precise in the wording. Do they mean remobilization of fine sediments in matrix? Material they sampled/measured? Moraine matrix?

We changed to 'remobilization of sediments from moraines…'

p. 3. For the part of the study dealing with sediment measurement and findings, it may be of interest to mention that ice sheet glaciations started around 7 to 5 Ma (Mercer and Sutter (1982 i recall)) with tills and supposedly moraines and ice sheet glacial erosion, for several million years, even before the Telken stuff deposited (are there implications for history of fines, glacial rock flour etc?)

We added « Mercer and Sutter (1982) showed that the oldest glaciogenic sediments we find in the area are 6 Ma, and consist of tills interbedded with lava flows that preserved them from erosion », in the section settings

line 23 "finally we are able. . ...sediment history is likely. . ." I would clarify what kind of sediment – fine sediment? For example, some would include boulders in this context, as the statement is written. I think they mean the type of materials they are working on – if not, their data only apply to the type of sediment sizes they are working on (e.g., rock flour).

Ok, added « fine » to make it clearer

p. 4. There is another moraine, the Menucos moraine (Singer et al., 2004; Douglass et al. 2006). Kaplan et al. (2011) recalculated these ages.; they are albeit buried in the last table of the 2011 supplement. The key point is the recalculated ages are around 17 ish (18 to 17 ka), I recall, ignoring obvious outliers.

These moraines have their importance in the area, however our objective is to constrain the U-series measurements with the 10Be data, so we feel it was outside the scope of the study.

Line 18 to 20. A clear example in which some photos (2 for example) would of immense help for evaluating what they sampled. A pit? A road cut? Can they provide a strat section, even schematic or simplified?

Some pictures were added in the supplementary materials.

p.5 see comment about photos

Changed

p.6 lines 14+ 1) say what the 'global rate' of Borchers et al rate is at SLHL. 2) how does global rate of Borchers et al compares with the local rate in Kaplan et al. 2011 and whether the difference in rates will mean for their older results. For younger features, should not matter too much, but once you get to Deseado and Telken age, even small differences between Borchers et al and Kaplan et al. rates could be significant given non-linear age equation.

I bring this up – in part – because I am actually wondering if they will get slightly better agreement with Ar/Ar and/or marine oxygen isotope comparisons; I cannot recall exact #s, but I recall the Borchers et al. rate is higher than that in Kaplan et al. which is for Patagonia. It is easy for the authors to estimate the difference – on the online cronus calculator Balco provides 'alternative versions for 2.2. Although still v.2.2 it is good enough to see the effect of using a slightly lower PR (Kaplan et al. 2011), if correct.

Thanks for making this point. We mention l. 21 p. 6 that changing the site production rate of +/- 0.3 at/g/yr does not significatively change the results. This range of site productions rates corresponds to changing the SLHL production rate from 3.6 to 4 at/g/yr. 3.6 is the minimum production rate at SLHL in Kaplan et al. (2011) and 4 is the production rate at SLHL in Borchers et al (2016), both using the Lm scaling scheme. We already tested what is mentioned by the reviewer. However, it was not clear in the paper. So we added two sentences in paragraph 2.2.3 to precise this point.

p. 7 to 11 – see comment above about U-Th analyses.

p. 11 section 2.3.4. if they can give one sentence on why this matters, to the non- expert, that would be helpful.

The recoil loss factor is a key parameter for the determination of the comminution age. This parameter and the equations are presented in the section 2.3.1. In section 2.3.4, we present the method we used to estimate it. We added a few sentence in section 2.3.1.

p. 13. As mentioned above, it is relevant to mention the Douglass et al study, albeit just in GSA abstract form, which has low erosion rates on par with their values. It is important for them to point out that this is much lower (and more realistic) than that boulder erosion rate presented in Kaplan et al 2005, which presented maximum rates. Albeit Kaplan et al. 2005 and douglass et al were for moraine boulders (may be slightly different than their data given possibly more exhumation on the moraines).

See our response above

p. 14 line 8 and 9 (last sentence before new section). Regarding the sensitivity tests, I suggest they may want to mention that the global rate in Borchers et al vs local rate in Kaplan et al., although very small, does not make a difference given a non-linear age equation. If the “global” rate in Borchers et al. is slightly higher (I recall), which is what is listed on the Cronus 2.3 site. The importance is that with a higher rate they get slightly lower ages, especially for Deseado and Telken time.

Given your previous comment, this point is addressed in section 2.2.3.

p. 15. Line 13 Section 3.3 title For non-experts, what do they mean by ‘: a 3- dimensional problem”

This is another example how a little bit more info may make these aspects of the paper a little more accessible to the general ESD audience.

This expression ‘3 dimensional problem‘ is misleading, we removed it from the title of the section.

Line 16. What do they mean by youngest moraine sample – which sample (refer to which sample in table)?

The youngest moraine is the sample F1. We modified the text accordingly.

p. 16. glacial periods are much dustier – with likely much of the dust coming from Patagonia (lots of literature on this). Perhaps 1 or 2 sentences on what such dust inputs may be bringing in, in the context of their results. Is this another implication they can highlight of their U-Th results and findings?

Your point is indeed very interesting. We discuss it at the beginning of section 4.2 (first paragraph).

p. 17. Line 18. Add something along the lines of “slight changes in scaling factors (see methods) do not affect this finding.”

Ok

Line 18 – the 476 ka – is this recalculated? I assume it is from the 2005 paper, but please state so.

This value is the value published in Kaplan et al. 2005, see table 2.

Line 24 before "Smedley" a word or two missing or open parentheses.

OK

p. 18. Top line 1 – spelling after the word Telken Line 27 grammar problem p. 19 Line 14-15. First two sentences - awkward writing.

Done

p. 20. per comment above, sorry, perhaps my non-expertise, but, can the authors say if there is a change in the U-Th data between Telken and Fenix moraines. A change in time of weathering rates? The relevance of the issue is testing the idea they discuss, in Kaplan et al. 2009. Is there a change in results and thus sediment transfer times/comminution age over the 1 Ma as valleys are excavated and Andes erode down?

There are changes in U-Th data as written in section 3.3. For 234U/238U these changes are small between Fenix and Telken compared to the total variability, however when combined to the variations of the f_alpha parameter, we observe a clear trend from Fenix to Telken in terms of age. The recycling time and weathering rate that we estimate from the Monte Carlo simulation is common to every samples. We just estimate that the weathering rate was higher before the final deposition (dated by 10Be exposure age) of the moraine. We cannot say whether there is a change in the transport time between 1 and 0 Ma, because this is a prerequisite from the Monte Carlo problem as we built it, that the recycling time is the same for every sample. This is an assumption that we make. The result that we obtain is an average. We clarified it in the paper.

Lines 17-19. Starting with "this is exacerbated. . ." I do not understand this sentence – it is not clear. The authors need to clarify.

This sentence has been removed since it was not really relevant and apparently not clear.

Line 23. What do they mean by 'moraine sediment.' That is very broad. Does this also apply to boulders? Or just find matrix in moraines? Please clarify (a similar question was raised earlier about the whole moraine being remobilized).

Ok, this is right, it remained unclear this question of size fraction of the moraine. Here we added '(the fine silty fraction)'.

p. 21. Line 24. I think I know what the authors mean, but as written, not recorded or not preserved is same thing. Rephrase to, not recorded because not preserved, or do they mean not preserved or ice did not reach this far? Something similar?

Yes, not recorded mean that ice did no reach this far (moraine not deposited in the area, but farther upstream). This is explianed in the following sentence. To make it clearer we linked both sentence with 'i.e. respectively'.

Table 1. It is becoming customary to also provide AMS ratios and carrier added (9Be). Given that (I think) it is standard enough to do this now in cosmo literature, that I have to insist these two columns be added to the table. Also, in caption, please remind reader of AMS standard used, given it is not common (except at ETH).

Added.

FIGURES Figure 1. It is difficult to evaluate fully the morphostratigraphic relations that are essential for interpretation of data from this figure: 1) need some photos associated with these maps – see comments above. This is especially so for the outwash linked to the moraine – cannot appreciate this from the map. Otherwise need to say perhaps not directly related to the specific moraine – does it actually really matter for their main findings if they establish this? that is, deseado outwash is deseado outwash whether specifically linked to a specific crest in Singer et al, 2004. 2) I suggest one more panel really zooming in on the key area where the crests, outwash, and samples are. A panel D. This could be in the SOM if there is not enough room in the paper. The photos could also be in the SOM – just add one figure of several photos.. Any strat sections, with where the samples are from, that they can provide in SOM? I think it is always best to have the photos in the main text if there is space. However, if there is not enough space, at least to have them in the supplement info as a figure. 3) explain CDI and SPI in caption. SPI is obvious, but not CDI ? there are lots of glaciers in C. Darwin– what is specifically being highlighted and why for CD? the biggest ice mass in CD? 4) there is an error in the caption – the transect is in panel B (it says panel C, but this is in B)?. 5) after "[c]": add "[c] Inset shows a west-east transect. . ..." 6) For cosmo ages, it would better to make the symbols the same colors as those on figure 2. Easier for the reader not familiar with the area. I would also make the 2 outwash symbols as squares on Figure 1, just like on figure 2. 7) plot Deseado boulder that you refer to, from the literature (is from Kaplan et al. 2005 ?)

We improved the figure as per your recommendations. We also included pictures in supplementary materials. It is however difficult to see something, gven the fact that this area is rather flat. And it is hard to figure out the relief on those pictures. The boulders locations would add too much informations, and the figure would become unreadible.

Figure 2 1) maybe also plot here the Deseado boulder age (without erosion) that refer to? If do plot it, say it is a minimum age. Up to authors though whether they think it is best to plot the boulder cosmo age on figure 2, or it would be too messy. It would highlight again the moraine oulder ages too young. 2) Could all Deseado 2 ages be minima? Not just the 293? Deseado 1 has such a nice tight cluster, hard to envision being too minimum, but Deseado 2 is more scattered and only 2 ages. 3) I suggest showing the full LR 2005 curve. It is relatively easy to do, it does not take up much room and would be more informative than showing positive Stages. It would convey more info such as stage 8 not being a big global glacial maxima (cf., Hein et al.) 4) in caption, say what the effect is of using local PR versus global PR of Borchers et al, if significant ? see above comment?. Although not much for younger moraines, given the age equation is non-linear, it can add up for older Deseado moraines?

We imroved the figure as per your recommendations. Idem than for figure 1 with boulders data, we think the figure would become hardly readible.

Figure 3. 1) This plot is difficult to digest for the broader readership of ESD. Perhaps add some labels. For example, in the bottom 3 quadrants, can they add labels to all or some of the quadrants to summarize what we are looking at? In the top panels maybe also some more labels would help – what is the blue line? What are the red lines? It is explained in the caption, but, per my comment at top of review I am just trying to think of ways to make this figure more accessible to all readers. 2) Also, it is not clear – are these all hypothetical – or based on the monte carlo simulations – it is implied so ('results' mentioned in caption), but it is not specifically stated, and obvious to non-expert. 3) 4th line from bottom – 10Be ages – please explain which 10Be ages – reader cannot tell simply from the figure. 4) third line from bot- tom – "Weathering intensities are lower after moraine deposition" – how do you know this? That is, please explain from the plot how this is known. Or show visually with suggestion above to add labels.

The legend of this figure has been rewritten and we added a legend, and deleted a curve. Obviously this was unclear, we hope we made it clearer in the new legend.

Figure 6. Needed to explain a little better for non-experts – see comments at start of review. For non-experts, what is likelihood signify? Instead of Trecycle – spell out? Maybe a table in the SOM of all of these terms with a brief explanation, for non-experts? In panel A for example, they may add some info to the plot itself – some labels of what we are looking at. For example, what does the bend and dark vertical-aligned purple dots mean? Maybe a few labels on this plot and the bottom plot would make this more accessible to nonexperts in U-Th approach.

Lot of the informations are contained in the text (section 2.3.5). We tried to make clearer what the figure is. The 'bend and dark vertical-aligned purple dots' do not mean anything particular. What is really important is the likelihood of the dot. We rewrote the meaning of this term in the caption.

Supplement information.

Section 2. Given the prominent role the profiles and monte carlo simulation play in the paper - this section needs to be expanded to explain: 1) what the monte carlo optimization is and how it is done? Some of this is in the main text, but more details in the context of the Supplement are appropriate here in my opinion. 2) a paragraph (at least) explaining what we are looking at on Figures 1 and 2. What are these panels? The caption should refer to each panel. Or the text. Please explain each panel in more depth and/or in caption. The right panel – this is a key figure. Explain more – what is reader looking at? Not much is needed, perhaps just a longer caption, but more is warranted in explaining Figures 1 and 2 in the context of supplementary information and simply to describe what the reader is looking at, which are some of the key results discussed in the main text.

We added some sentences in the text to explain more about it.

Figure 3 – in caption, for non-expert, can they add one more sentence saying what f230 = 0 means? The authors also may want to add some labels along the lines of the comment above for main text figures.

Ok

Figure 5. Explain specifically where time comes from (e.g., U-Th dates associated with moraine/outwash dating. . ..and so on, see table 3)

Done

Per comments above, add photos and any strat sections. I would put the photos in the main text if there is room.

Anonymous Referee #2

Received and published: 5 October 2017

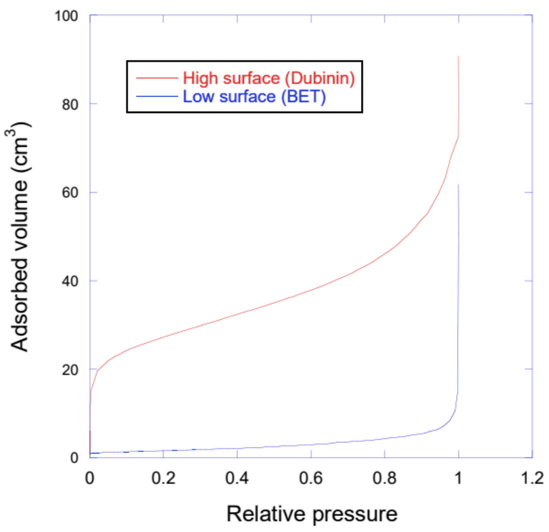
This manuscript present some interesting cosmogenic and U-series isotope data. While the combination is interesting, it seems that isotopic systems are used inde- pendently with little overlap. Cosmogenic isotopes were used to date the moraines on the one hand, and U-series to infer information about sediment transport on the other hand. It almost reads like 2 independent manuscripts.

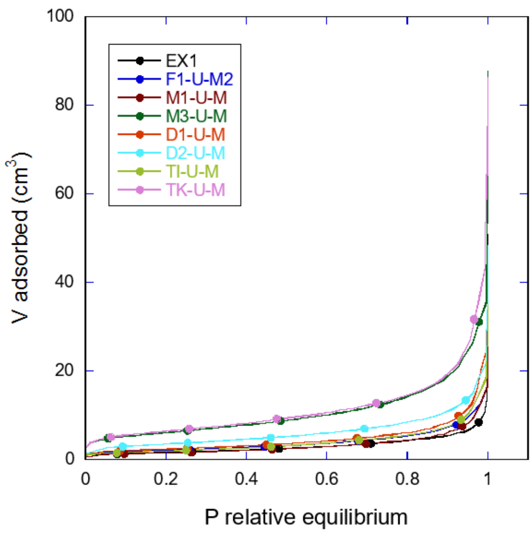
We feel the conmbined 10Be/U-Th data is key for our study. The 10Be data are used to date the moraines and U series are used to infer information on sediment transport. Both are not independant, because the ages determined using 10Be are needed to determine the sediment recycling time. Without these ages, the U-Th data do not give as much informations as they give with the 10Be data. Moreover, both the sequence of exposure ages and the recyling time determined from U-Th data, tell us that there is sediment reworked and at which timescale. Both set of data (10Be and U-Th) highlight the same timescale of 100 kyrs (recurrence of glacial cycles). So we think that it is appropriate to keep the two sets of data together.

While I command the authors' efforts, and this work should of course eventually be published, there are many aspects that the authors may want to consider, and which significantly affect how the U-series data are interpreted. I provide detailed comments in the PDF attached but the main points are that:

- the fractal correction is possibly not needed and result in an underestimation of recoil fractions. As a result, the authors explore a more complex (but less constrained) model (previously proposed by Scott et al. in the early 1990's, but also more recently re-introduced). A simple way to test this is to look at the type of isotherms obtained during BET analysis. Only a mesoporous would require the fractal correction to be invoked. I think Lee investigated this but as far as I know this is unpublished, but the authors may want to consider this point.

It is difficult to adress this comment since it is based on unpublished work. However, the isotherms we obtain are of type 2, following the classification of Brunauer (1945). Two extreme cases can be distinguished (see figure below), depending on their specific surfaces : Dubinin type (high surface → microporous), and BET type (lower surface → mesoporous). We report a plot showing the isotherms of our samples. One can see that they are all of the BET type (mesoporous). So if we follow the reasoning of the reviewer, a fractal correction must be applied to ours amples.





The comment of the reviewer made us realize there was a mistake in the section 1 of supplementary material. It is mentioned we applied the Dubinin-Radushkevich equation to the samples. A larger amount of samples (designed for ther purposes) were analyzed at the same time as those published in the paper. The section 1 of supplementary material was written by Thierry Reuschlé, who performed the analysis, for the whole group of samples, whereas only a few samples were concerned by the paper. The samples concerned by the Dubinin Radushkevitch equations (microporous) are actually not published in this paper. So we corrected this section, which now addresses the reviewer's concern.

- while the weathering model is presented, there is a lack of details on how it is solved, and what justifies the assumption made in the model (e.g. same 'recycling time' and weathering intensity for all samples? why? this record cover a very variable period of Earth's history). I only recommend rejection because I believe the extent of revisions to be undertaken is large, but a new manuscript should be submitted, taking into consideration the comments above and in the document attached.

We are aware that the assumptions made (same recycling time and weathering intensity for all samples) can be debated because the period covered by the record is the last several hundreds of throusands of years. Parameters that could drive variation of the weathering intensities are the climatic parameters mainly, and the freshness of the sediment. We consider a particular size fraction, so this latter parameter should not vary significanty. Climate varied, however the minima and maxima of the glacial interglacial variations remain almost similar from a cycle to another by 700-800 ka (Mid Pleistocene Transition). We can then assume that the processes occuring during a glacial cycle are comparable in intensity from a cycle to another.

Regarding the recycling time, the main other parameter (apart from climate) influencing it is the morphology (mainly valleys and overdeepening). The Great Patagonian Glaciation occured before 1 Ma, these glaciations are most probably responsible for carving the U shaped valleys and the overdeepening in which the lake is nowadays. Moreover the results of the numerical experiments of Kaplan (2009) about the extent of ice related to the elevation of the accumulation area and the depth of the overdeepening suggest that this overdeepening was most probably already carved before 1 Ma. Recently Christeleit (2017) published a paper in which she argues that much of the observed valley relief was carved between 10 and 5 Ma. So it seems reasonable to assume that the recycling time remained constant over the period covered by the samples. Fianlly, the 10Be data suggest the same timescale than U-Th suggest : apparently every second glacial cycle is preserved n the morainic sequence, suggesting a timescale of 100 to 200 ka of sediment reworking. Of course this must have changed on longer timescale, but over the period covered by the moraine sequence it seems those variations are not detectable.

We recognize those points were not clear, or absent in the paper, we improved it by adding a paragraph in section 2.3.5.

Please also note the supplement to this comment: https://www.earth-surf-dynam-discuss.net/esurf-2017-45/esurf-2017-45-RC2- supplement.pdf

Page 8 : in theory this is correct. but this ignores that 234U will be in damaged sites, so f230 is likely to be greater than the value given in equation 1.

This is right, it remains however impossible to quantify this. We added a sentence to make ths point.

not new. already given in Scott et al. 1992, and Dosseto and Schaller 2016.

We did not find Scott et al. 1992 : is it « Natural decay series studies of the redox front system in the Pocos de Caldas uranium mineralization » by MacKenzie and Scott et al. (1992) ? We added the reference Dosseto and Schaller.

Page 9 : problem is that Lee shows that ignition affects surface area. also, Martin et al. 2015 show that ignition is not optimal for (234U/238U)

Lee showed slight differences on 234U/238U between the ignition method and the H2O2 leach for removing organic matter. However other studies showed that H2O2 leaches were not fully selective (Tessier 1979). This explained in point #2 of section 2.3.2.

why introduce a bias by removing the <4um fraction? Handley shows that it is not recommended

If clays are directly precipitated onto the mineral surface after weathering then they should be preserved and analyzed. In such a case the problem and equation do not need to take weathering into account. But how can we be sure that clays present in the sample are kept on mineral surface? Moreover our understanding of Handley's work is that she points out that some clays should be conserved and some should be removed, but it is impossible to isolate both. If clay fraction is made by primary minerals smaller than 4 μm then they should be conserved. However our MEB images show that clays (secondary minerals) are present.

Page 10 : sodium citrate should be added at each step to avoid re-adsorption. while not affecting the (234U/238U) ratio (see Martin et al. 2015) this could be critical for Th and could explain differences in Th concentration (although I acknowledge it doesn't seem to affect the 230Th/238U ratio)

We acknowledge this would have been better to add this sodium citrate. As mentionned the problem is for Th. That’s why we tested the sensitivity of the Monte Carlo to changing values of fa230 (recoil loss fraction of 230Th) : a lower fa230 is equivalent to readsorption of the ejected 230Th. See supplementary material. Also, increasing the number of chemicals added to the samples increases the chance of disolving or corroding the surface of the grains.

Page 11 : results on rock standards need to be reported.

Page 14 : no evidence there was anything to remove in the first place

Page 15 : clays are not a problem: the are the product of incongruent weathering so unless they are completely precipitated from soluion (which is only common in marine environments; see textbooks on clays), they are not a concern and shouldn't be removed,

In the lake clays can be precipitated. So we cannot be sure that the clays in the samples are directly originated from the weathering of the silts that we analyze.

Page 16 : or the recoil fraction is under-estimated because applying a fractal correction when it's not needed. Depending on the shape of adsorption-desorption isotherms, you can identify the type of surface porosity; and the fractal correction should allow be applied when dealing with a mesoporous material, because only then the size of surface pores is comparable to the length of recoil and including these pores would result in overestimation of the surface area and recoil fraction (and thus requires the fractal correction). if isotherms suggest a microporous or macroporous material (as it is often the case), no fractal correction is needed. In this case, you find that you can obtain numerical values for the comminution age. For instance, D1-U-M yields 400 ka; D2-U-M gives 320 ka.

See our response above to your main comments.

does this assume that all samples have the same 'recycling time'? what would justify such assumption? so you have 8 samples x 2 ratios = 16 inputs to the moded; and 5 outputs (again assuming the recycling time is the same for all samples?)? in this case, the number of unknowns could be increased since they're so many inputs. for instance, samples from different moraines could have different recycling times (thus, 13 outputs). I think this needs a bit more details (even if the model is described above) before jumping into the results.

See our response above to your main comments.

U-Th and ^{10}Be constraints on sediment recycling in proglacial settings, Lago Buenos Aires, Patagonia

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Abstract. The estimation of sediment transfer times remains a challenge to our understanding of sediment budgets and the relationships between erosion and climate. Uranium (U) and Thorium (Th) isotope disequilibria offer a means of more robustly constraining sediment transfer times. Here, we present new Uranium and Thorium disequilibrium data for a series of nested moraines around Lago Buenos Aires in Argentine Patagonia. The glacial chronology for the area is constrained using *in situ* cosmogenic ^{10}Be analysis of glacial outwash. Sediment transfer times within the periglacial domain were estimated by comparing the deposition ages of moraines to the theoretical age of sediment production, i.e. the comminution age inferred from U disequilibrium data and recoil loss factor estimates. Our data show first that the classical comminution age approach must include weathering processes, accounted for by measuring Th disequilibrium. Second, our combined data suggest that the pre-deposition history of the moraine sediments is not negligible, as evidenced by the large disequilibrium of the youngest moraines despite the equilibrium of corresponding glacial flour. Monte Carlo simulations suggest that weathering was more intense before the deposition of the moraines and that the transfer time of the fine sediments to the moraines was on the order of 100-200 ka. Long transfer times could result from a combination of long sediment residence times in the proglacial

lake (recurrence time of a glacial cyle) and the remobilization of sediments from moraines deposited during previous glacial cycles. ^{10}Be data suggest that some glacial cycles are absent from the preserved moraine record (seemingly every second cycle), supporting a model of reworking moraines and/or fluctuations in the extent of glacial advances. The chronological pattern is consistent with the U-Th disequilibrium data and the 100-200 ka transfer time. This long transfer time raises the question of the proportion of freshly eroded sediments that escape or not the proglacial environments during glacial periods.

1 Introduction

The sedimentary cycle incorporates the erosion of rocks followed by the transport and deposition of sediments. While rates of erosion and deposition can be accurately documented, tracing the history of sediments between production and deposition remains challenging. Importantly, mechanisms of transfer and alteration of sediments during transport play a key role in the evolution of basins and feedbacks between erosion and climate, especially because the age of sediment strongly controls its susceptibility to weathering (e.g. White & Brantley, 2003; Vance et al., 2009). This is particularly the case in glacial settings because glaciers are highly efficient at eroding landscapes (e.g. Hallet et al., 1996; Koppes & Montgomery, 2009) and produce highly-reactive and easily-weathered sediments (Anderson et al., 1997; White & Brantley, 2003; Anderson, 2005). Moreover, glaciers can create large overdeepenings that are subsequently filled with sediments isolated from interactions with surface processes. The efficiency of evacuating those sediments post-deposition is poorly known.

Silicate weathering is an important surface parameter for cooling climate over geologic and glacial-interglacial timescales because it consumes CO_2 (Ebelmen; Walker et al., 1981). But its role in controlling CO_2 concentrations and climate variations over glacial-interglacial cycles is contentious (Foster & Vance, 2006; Vance et al., 2009; Lupker et al., 2013; von Blanckenburg et al., 2015; Cogez et al., 2015). Understanding whether weathering varied over these cycles requires robust determinations of sediment transfer times.

For example, long transport times can cause a lag and/or a damping of the response of weathering to climate or erosion forcing and bias reconstructions of erosion and weathering intensity and variations through time.

Geochemical tools offer a means of measuring sediment transport times. Uranium series isotopes are particularly useful because the diversity of chemical elements in the radioactive decay chain (U, Th and Ra) leads to disequilibrium during surface process fractionation. Moreover, the half-lives of these isotopes range from 1,500 to 250,000 years, corresponding to sediment transport process times. The timescales of weathering processes have been particularly well documented using U-Th-Ra disequilibria in soil sediments and river waters (e.g. Ackerer et al., 2016; Chabaux et al., 2008, 2012, 2013; Dosseto & Schaller, 2016; Dosseto et al., 2010, 2012, 2014; Granet et al., 2010; Keech et al., 2013; Ma et al., 2013). An alternative approach uses the fine fraction of silicates to date the time since physical erosion of sediments (DePaolo et al., 2006, 2012). The method is based on the α-recoil of uranium during radioactive decay, triggering the loss of a fraction of the daughter isotopes compared to the parent in small ($\leq 50 \mu$m) grains. The time since comminution can theoretically be estimated, despite difficulties and limitations discussed in previous studies (Maher et al., 2006; Lee et al., 2010; Handley et al., 2013a, b; Dosseto et al., 2010). However, this method only involves the ^{234}U/^{238}U ratio and so neglects the effect of chemical weathering and further reduction of grain size after comminution.

In this study, we sampled sequences of nested moraines around Lago Buenos Aires in Argentine Patagonia, which range in age from 0 to 1 Ma (Singer et al., 2004; Kaplan et al., 2005, e.g.). The aim was to constrain the pre-deposition history of sediments within the moraines. First we refined the deposition age of five of the moraines using *in situ* cosmogenic ^{10}Be exposure and depth profile dating. To constrain the sediment transport times, we used the comminution ages approach combined with ^{230}Th disequilibrium measurements. We show that taking weathering into account can help to resolve previous issues with the comminution ages methodology. Finally, we are able to estimate that the pre-deposition history of the fine sediment (4-50 μm) is likely on the order of 100-200 ka, including sediment recycling and chemical weathering in the proglacial system. Our findings have implications for the understanding of the couplings between erosion and climate.

2 Settings and methods

2.1 The Lago Buenos Aires moraines

Lago Buenos Aires in Argentina (Lago General Carreras in Chile) is a large proglacial lake, around 100 km long and 5-20 km wide, oriented east-west, at 46°S (Figure 5). The present climate of the area is temperate, with an average annual precipitation of 100 mm/yr and temperatures between 4 and 14 °C, producing open steppe vegetation. On the eastern edge of the lake, a series of frontal moraines are nested from the youngest close to the lake to the oldest 50 km further east. The chronology of these moraines has been studied: the five innermost moraines (Fenix 1-5) are Last Glacial Maximum (LGM) in age (Kaplan et al., 2004; Douglass et al., 2006). Singer et al. (2004) dated lava flows interbedded with six of the intermediary moraines (Moreno 1-3 and Deseado 1-3) and showed that they range in age between 109 ka and 760 ka. Finally, six moraines (Telken 1-6) in the outermost part of the system were likely deposited between 760 ka and 1016 ka. Mercer & Sutter (1982) showed that the oldest glaciogenic sediments we find in the area are 6 Ma, and consist of tills interbedded with lava flows that preserved them from erosion. Cosmogenic nuclide exposure dating of boulders supports the assertion that erosion and degradation of older moraines in this region yields erroneously young ages (e.g. Kaplan et al., 2005; Hein et al., 2009). Recently, Hein et al. (2017) used exposure dating of cobbles on outwash related to the Moreno moraines to show that they were constructed at ca. 260-270 ka, during Marine Isotope Stage (MIS) 8. For this study, we further refined the chronology by providing direct age constraints for the Deseado 1, Deseado 2 and Telken 5 moraines (see section 2.2).

U and Th samples were taken from silty beds inside the moraines, several meters below the surface in order to avoid potential post depositional modifications, such as weathering or dusts inputs (see picture in supplementary materials). In addition to moraine samples, a glacial flour sample was taken at the front of the Los Exploradores Glacier in Laguna San Rafael National Park to estimate the initial U-Th composition of silts found in the moraines (Figure 5). This glacier covers part of the total catchment draining into Lago Buenos Aires. Consequently, it is unlikely to be representative of the entire basin but provides an estimate of the initial composition of silts after comminution.

2.2 ^{10}Be methodology

2.2.1 Sampling

The cosmogenic nuclide exposure dating methodology followed the sampling recommendations of Hein et al. (2009) and Darvill et al. (2015). We sampled quartzite cobbles for ^{10}Be analysis from the surfaces of outwash grading to the Deseado 1, Deseado 2 and Telken 5 moraines. Similar samples were taken for Moreno 1 and 3 moraines and are presented in Hein et al. (2009). Using outwash cobbles has been shown to overcome issues of post-depositional erosion that affects moraine boulders in this area (Hein et al., 2009), but relies on finding locations where there is a clear geomorphic relationship between the glacial outwash and moraines. At suitable locations, cobbles were targeted based on quartz composition, size (6-15 cm and 300-1,000 g to contain sufficient ^{10}Be), and preservation (sub-rounded shape, little aeolian erosion, partially buried suggesting they were *in situ*). Surface lowering or up-freezing of cobbles can result in vertical movement through the outwash unit. In this region, it is more likely that cobbles will underestimate the true age of the unit (Hein et al., 2009; 2017). To circumvent this issue three or four cobbles were analysed at each sampling location, and the oldest taken as the best estimate of moraine deposition age.

As well as cobbles from the outwash surfaces, ^{10}Be depth profiles through exposures within the outwash units were used to estimate surface erosion rates and nuclide inheritance. We sampled through two outwash profiles relating to the Deseado 2 and Telken 5 moraines. Each of the samples making up the depth profiles consisted of 50 to 100 quartz-rich gravel clasts taken from horizontal lines, 2-4 cm wide at various depths below the surface. Five depths were sampled per profile between 50 cm and 240 cm deep; sufficient depth to account for nuclide attenuation from the surface and reliably estimate any cosmogenic ^{10}Be inheritance. The top 50 cm of the profiles were carbonated due to post depositional processes but the rest of the profiles were of relatively consistent density. See supplementary materials for pictures.

2.2.2 Analytical methods

Surface cobbles were analysed individually as independent estimates of exposure age. The gravels at each depth in the profiles were amalgamated to produce an average nuclide concentration for that depth. Samples were crushed and sieved to obtain the 125-250 μm and the 250-500 μm fractions, which were then purified using a Frantz magnetic separator to isolate non magnetic fractions. Following dissolution in concentrated

HF, the residual sample material was leached with 10mL H_2O to extract $Be(OH)_2$ (Stone, 1998). A combination of anion and cation resins and precipitation was used to purify Be (von Blanckenburg et al., 2004; Norton et al., 2008). The oxidized BeO was mixed with Nb powder and pressed into cathodes and measured on the 0.5 MeV Tandy accelerator at ETH Zürich (Müller et al., 2010). The resulting $^{10}Be/^{9}Be$ ratios were normalized to S2007N ($^{10}Be/^{9}Be = 28.1$ x $10-12$; (Christl et al., 2013; Kubik & Christl, 2010)), with a ^{10}Be half-life of 1.387 ± 0.012 Ma (Chmeleff et al., 2010; Korschinek et al., 2010). Measured $^{10}Be/^{9}Be$ ratios were 30 - 500 times higher than the procedure blank (9.60 x $10^{-15} \pm 4.77$ x 10^{-15}). Blank corrected ^{10}Be concentrations decrease exponentially with depth from greater than 36 x 10^5 for near surface samples to less than 2 x 10^5 at 2 meters depth.

2.2.3 Deposition ages calculations

Exposure ages for the outwash surface cobbles were calculated from ^{10}Be concentrations using version 2.3 of the CRONUS online calculator (Balco et al., 2008). Shielding was measured in the field (>0.999999 for every sample), a density of 2.65 g.cm^{-3} was used for every sample, and no erosion rate was applied since there were no visible signs of surface erosion. We used the global ^{10}Be production rate of Borchers et al. (2016) (4 at/g/yr) and the 'Lm' scaling scheme (as described in Balco et al. (2008), Lal (1991) and Stone (2000) with paleomagnetic corrections from Nishiizumi et al. (1989)). However the scatter between the different scaling schemes is comparable to the external uncertainties. The production rate from Kaplan et al. (2011), calculated from a site very close to LBA, gives 3.6 at/g/yr (instead of 4 for Borchers et al. (2016)). The difference in calculated exposure ages between those two productions rates remains within uncertainties.

We modeled the most probable deposition age, surface erosion rate, and nuclide inheritance for each depth profile using the Monte Carlo simulation approach of Hidy et al. (2010). A site production rate of 6.2 atoms/g/yr was used, taken from an average of the scaling schemes in the CRONUS calculator. Changing this production rate by ±0.3 atoms/g/yr does not significantly alter the results. This range of site production rates (5.9-6.5 atoms/g/yr) corresponds to SLHL production rates 3.6-4 at/gr/yr, using the Lm scaling scheme. We used an average profile density of 2.5 g.cm^{-3} (based on field observations), and a shielding factor of 1. The *a priori* values assigned to the Bayesian Monte Carlo simulation were: 0 to 1 cm/ka erosion rate with a maximum erosion threshold of 1000 cm and uniform distribution (based on erosion rates in Hein et al. (2009)

for the Hatcher profile in nearby Lago Pueyrredon and Darvill et al. (2015)); 0 to 10^5 atoms/g inheritance with uniform distribution (based on concentrations in the lower levels of the profiles); 200 to 800 ka age range for Deseado 2 and 700 to 1200 ka age range for Telken 5, with uniform distribution. These ages were chosen based on the argon ages of Singer et al. (2004) and the Moreno age refinement of Hein et al. (2017).

2.3 U-Th methodology

2.3.1 Principles and theory

The comminution age theory introduced by DePaolo et al. (2006) is a promising approach to estimating the age of sediment production. ^{238}U decays into ^{234}Th with a half life of 4.5 Ga. During this α emission, the daughter isotope recoils over a distance of several tens of nanometers depending on the type of mineral (e.g. 30 nm for a feldspar). ^{234}Th quasi instantaneously produces ^{234}U, with a longer half life of 245 ka: long enough to be measurable and usable on geologic timescales. If the ^{238}U decay occurs on the edge of a sediment grain, the daughter isotope is ejected from the grain and a disequilibrium in the decay chain starts as the ^{234}U/^{238}U ratio of the grain starts to decrease. If the grain is small enough, with a large surface area compared to its volume, the disequilibrium can be measured. Steady state is reached after a time characterized by the half life of ^{234}U. During this transient state, the magnitude of the disequilibrium in a grain equates to the time since it was produced. The steady state disequilibrium value depends on the fraction of ^{234}U ejected out of the grain due to α recoil. The process can be modeled using the following two equations describing the evolution of ^{238}U and ^{234}U:

$$\frac{dN_{238}}{dt} = -\lambda_{238} N_{238}$$

$$\frac{dN_{234}}{dt} = -\lambda_{234} N_{234} + (1 - f_{\alpha}^{234})\lambda_{238} N_{238}$$

λ_{234} and λ_{238} are the decay constants of ^{234}U and ^{238}U respectively. N_{234} and N_{238} are the number of nuclides for each isotope, and f_{α}^{234} is the recoil loss fraction of ^{234}U (the proportion of ^{234}U in the grain ejected during α decays). This parameter is critical, since it represents the process whose existence determine the possibility of tracing the comminution age. When the effect of this process starts to be measurable, the clock starts. We discuss in more detail how this parameter can be evaluated in section 2.3.4. The evolution of the U/Th ratio over time is shown in Figure 3 (blue curves). The steady state disequilibrium model is based on the assumption that the sediment grain has not been weathered over time, or at least that the loss of nuclide due to weathering was negligible compared to loss by α-recoil. The assumption may be valid for very arid areas, but is otherwise unlikely to hold true. However, the influence of weathering on the comminution age model has not been investigated.

In order to decouple the respective influence of α-recoil and weathering, we measured ^{230}Th concentration. The solubility of thorium is much lower than uranium. Therefore the ratio of ^{230}Th/^{234}U can inform weathering rates. ^{230}Th is produced by ^{234}U decay with a half life of 75 ka. It can also be ejected out of the grain due to α-recoil. The recoil loss fraction of ^{230}Th, $^{230}_{\alpha}$ is related to $^{234}_{\alpha}$ (Hashimoto et al., 1985; Neymark, 2011)

$$f_{\alpha}^{230} = f_{\alpha}^{234} \times \frac{\alpha_{230} M_{234}}{\alpha_{234} M_{230}} = 1.176 \times f_{\alpha}^{234} \quad (1)$$

α_{230} and α_{234} are the decay energy of ^{238}U to ^{234}U and of ^{234}U to ^{230}Th, respectively (71.79 and 82.96 keV respectively). M_{234} and M_{230} are the atomic mass of ^{234}U and ^{230}Th. This gives $f_{\alpha}^{230} = 1.176 \times f_{\alpha}^{234}$. (In practice f_{α}^{230} is probably larger since ^{234}U is on damaged sites, so it increases the probability for ^{230}Th to be ejected from the grain during a decay). The evolution of the number of nuclides ^{230}Th is then given by

$$\frac{dN_{230}}{dt} = -\lambda_{230} N_{230} + (1 - f_{\alpha}^{230}) \lambda_{234} N_{234} \quad (2)$$

Since $f_{\alpha}^{230} > f_{\alpha}^{234}$ and $\lambda_{230} > \lambda_{234}$, (^{230}Th/^{238}U) < (^{230}Th/^{234}U) < (^{234}U/^{238}U) is always verified. However, weathering would increase the ratio of ^{230}Th/^{234}U and ^{230}Th/^{238}U because Th is less soluble than U, and likely decrease the ratio of ^{234}U/^{238}U because radioactive decay places ^{234}U on a more fragile site compared to ^{238}U so that it could be more easily weathered (DePaolo et al., 2012; Handley et al., 2013a). Consequently, Th isotopes can help to identify and quantify the effects of α-recoil and weathering, respectively. New equations can be derived for the evolution of the different nuclide contents following Chabaux et al. (2003, 2008), and Dosseto et al. (2008); Dosseto & Schaller (2016) :

$$\frac{dN_{238}}{dt} = -k_{238} N_{238} - \lambda_{238} N_{238}$$

$$\frac{dN_{234}}{dt} = -k_{234} N_{234} - \lambda_{234} N_{234} + (1 - f_{\alpha}^{234}) \lambda_{238} N_{238}$$

$$\frac{dN_{230}}{dt} = -k_{230}N_{230} - \lambda_{230}N_{230} + (1 - f_{\alpha}^{230})\lambda_{234}N_{234}$$

where k_{238}, k_{234} and k_{230} are the leaching coefficient of ^{238}U, ^{234}U and ^{230}Th respectively. The solution is shown in figure 3.

The steady state activity ratios can also be derived:

$$\left(^{234}U/^{238}U\right)_{steady} = \frac{\lambda_{234}\left(1 - f_{\alpha}^{234}\right)}{\lambda_{234} + k_{234} - k_{238}}$$

$$\left(^{230}Th/^{234}U\right)_{steady} = \frac{\lambda_{230}\left(1 - f_{\alpha}^{230}\right)}{\lambda_{230} + k_{230} - k_{238}}$$

$$\left(^{230}Th/^{238}U\right)_{steady} = \frac{\lambda_{230}\lambda_{234}\left(1 - f_{\alpha}^{230}\right)\left(1 - f_{\alpha}^{234}\right)}{\left(\lambda_{230} + k_{230} - k_{238}\right)\left(\lambda_{234} + k_{234} - k_{238}\right)}$$

Here, parentheses are used to represent activity ratios, corresponding to the isotopic ratio multiplied by the ratio of the respective decay constants of each isotope.

2.3.2 Sample preparation

Silt samples were collected from moraines. Water has likely percolated through these silts since moraine deposition, triggering the precipitation of secondary carbonates, oxides, organic matter and clays. These secondary fractions have different U and Th disequilibria than the primary silicate and it is therefore important to remove them before analysis without altering the primary silicate fraction. We used the protocol established by Lee (2009), with slight modifications to help overcome the low solubility of Th that can make it difficult to remove during successive leaching, and allow Th to adsorb onto mineral surfaces after ejection from the grain. Our protocol is as follows:

1) The samples were sieved at 50 μm in order to collect the $\leq$ 50 μm fraction.

2) Approximately 10 g of this fraction was then weighed in a quartz crucible and heated for 4 hours at 550°C to burn organic matter. Lee (2009)'s optimal protocol uses H_2O_2 to dissolve organic matter, but other studies have shown that this reagent is not always fully selective (Tessier et al., 1979). The ashing of organic matter yields similar results, but is normally conducted at the end of the procedure (Lee, 2009). By ashing first, we were able to process larger amounts of silt, limiting furnace contamination and aiding the process

of dissolving of precipitates (mainly oxides formed during heating) in the following steps. Oxides turn the samples a brown to reddish color.

3) Sample grains were dispersed in 0.1 N sodium oxalate and the ≤ 4 μm fraction was removed using Stokes settling. The ≤ 4 μm fraction consists of clays and also primary silicates with larger U-Th disequilibria.

4) For each sample, 4 g of sediment was weighed in a 50 mL centrifuge tube and leached twice with 32 mL 1M magnesium nitrate to remove the exchangeable fraction and any residues of the ashing process.

5) 32 mL of acetic acid buffered to pH5 with 1M sodium acetate was then added and agitated at room temperature during 5 hours to dissolve the carbonates.

6) 40 mL of 0.04M hydroxilamine hydrochlorid in 25% (v/v) acetic acid was added and heated at 95°C for 6 hours twice over to dissolve oxides. After this procedure, the samples still had a brown to reddish color from oxides formed in the furnace. These amorphous oxides can be resistant to the hydroxylamine hydrochloride treatment, as shown by Gontier (2014). These authors recommended using oxalic acid in ammonium oxalate (agitated in the dark at room temperature for 4 hours) (Leleyter & Probst, 1999). This procedure preserves silicate from dissolution (Gontier, 2014). Note that for the last three steps, the leaching residues were centrifuged and rinsed twice with mQ water.

At this stage the samples should contain only primary silicates between 4 and 50 μm. To evaluate whether sample preparation fully removed secondary phases (e.g. carbonates, oxides, clays) without affecting primary silicates, we made Scanning Electron Microscopy (SEM) images at the University of Strasbourg, shown in section 3.2. Our observations showed the method to be effective (see the Supplementary Information for further discussion on the efficacy of Th removal in this protocol) and the samples were deemed ready for disequilibrium analysis.

Two duplicates of this pretreatment protocol were measured (see Table 3). They are remarkably consistent within errors for (^{230}Th/^{238}U) and (^{230}Th/^{234}U) and only slightly different for (^{234}U/^{238}U) (only twice the external uncertainty). Since the surface area data are also slightly different, this small difference could be associated with heterogeneities in the powder, or fractionation of the grain size during the sampling of the powder, or biases in the leaching process.

2.3.3 U-Th disequilibrium analysis

A first aliquot of the samples was dedicated to U-Th analysis. This aliquot was powdered in an agate ball mill to optimize sample homogeneity and acid dissolution. Approximately 100 mg of this powder was spiked with ^{233}U-^{229}Th tracer and digested with concentrated HF-HNO_3-$HClO_4$ acids following Pelt et al. (2013). Separation and purification of U and Th followed standard anionic resin chromatography (Dequincey et al., 2002; Granet et al., 2007; Pelt et al., 2008). U and Th isotopic ratios and concentrations were determined by standard-sample bracketing (SSB) using an MC-ICPMS Neptune, following an optimised procedure upgraded from previously published protocols (Pelt et al., 2008; Ma et al., 2012; Bosia et al., 2016). The ^{233}U-^{229}Th spike was calibrated against the gravimetric NIST SRM 3164 and 3159 U and Th pure solutions. IRMM-184 and IRMM-035 isotopic standard solutions were spiked and used as bracketing solutions for U and Th SSB analysis, with the ^{233}U/^{234}U and ^{229}Th/^{230}Th ratios of these mixtures calibrated by TIMS Triton. We used the consensus value from Sims et al. (2008) for the ^{232}Th/^{230}Th ratio of the IRMM-035 standard and our own absolute TIMS Triton measurements for the ^{234}U/^{238}U ratio of the IRMM-184 standard instead of the certified values. The peak-tailing of the ^{232}Th over the ^{230}Th was corrected using an exponential law fitted by signals measured in 229.6 and 230.6 masses. The precision and accuracy of the data were estimated from regular analysis of international pure solutions (HU1 for (^{234}U/^{238}U): 1.0008$\pm$0.0005 (2σ, N=5), IRMM-036 for ^{232}Th/^{230}Th: 329064$\pm$1313 (2σ, N=5)) and rock basalts (BCR-2, AThO and BE-N). Results are consistent with published data and the ^{238}U-^{234}U-^{230}Th secular equilibrium assumed for old (several Ma) BCR-2 and BE-N 'unweathered' basalts, within errors. Based on these pure solutions and homogeneous basalt powders we estimate an uncertainty (2SD) of 0.2% for (^{234}U/^{238}U), 0.5-1% for (^{230}Th/^{232}Th) and 1-1.5% for (^{238}U/^{232}Th), (^{230}Th/^{238}U) and (^{230}Th/^{234}U). A duplicate of the protocol was measured and agree within the internal measurement uncertainties for (^{234}U/^{238}U), (^{230}Th/^{238}U), (^{230}Th/^{234}U) (see Table3). Usual blanks in the lab are in the range of 20-50 pg for U and 100-200 pg for Th and therefore negligible here.

2.3.4 Estimation of the recoil loss factor

The recoil loss parameter (f_α) is a critical parameter in comminution age theory as presented in section 2.3.1. Different methods have been proposed to estimate this parameter, summarized in Maher et al. (2006) and

Lee et al. (2010), but discrepancies up to an order of magnitude can be found between the different methods (Handley et al., 2013a, b). Here we used the N_2 gas absorption technique, which consists of surface area and fractal dimension measurements. Other methods are more subjective in that they involve visual estimation and assumptions for the aspect ratio and surface roughness coefficient. We measured the specific surface area for all samples and assessed the efficiency of this method to determine f_α and the relation between f_α and U-Th disequilibria.

A second aliquot of the samples (at least 1 g per sample) was prepared for these analyses, conducted at the Institut de Physique du Globe de Strasbourg on a Carlo Erba Sorptomatic 1990 machine. Details of the procedure are given in the Supplementary Information. Fractal dimension were also measured for each sample and the recoil loss factor f_α calculated following Bourdon et al. (2009), after Semkow (1991):

$$f_\alpha = \frac{1}{4}\left[\frac{2^{D-1}}{4-D}\left[\frac{a}{R}\right]^{D-2}\right] R.S.\rho_s \tag{3}$$

Where S is the specific surface area, D is the fractal dimension, a is the diameter of the adsorbate molecule (0.35 nm), R is the recoil lenghth (30 nm), and ρ_s is the density of the solid (2650 kg.m^{-3}).

2.3.5 Determination of the recycling time and weathering intensities : Monte Carlo analysis

A Monte Carlo approach was used to determine weathering intensities during both the pre- and post-deposition histories of the different moraine silts, and the duration of this pre-deposition history (called 'recycling time' from this point forward).

^{10}Be exposure ages inform on the deposition age of the moraines. The age extracted from U-Th data inform on the time since the sediment has been produced. So it is equal to the sum of sediment recycling time (time elapsed between commminution and deposition of the moraine) and the deposition age. The problem as formulated in section 2.3.1 has a large number of unknowns compared to the number of data in each equation. So the problem is underdetermined. However if geomorphology and climate did not vary too much between each glacial period, we can assume that recycling times and weathering intensities are comparable between the different sample. In facts, the glacial maxima observed since the Mid Pleistocene Transition are quite comparable. We can then assume that the processes (especially weathering) occuring during a glacial cycle are comparable in intensity from a cycle to another. Regarding the recycling time, the main other parameter (apart from climate) influencing it is the morphology (mainly valleys and overdeepening). The Great Patagonian Glaciation oocured before 1 Ma (Mercer & Sutter, 1982), these glaciations are most probably responsible for carving the U shaped valleys and the overdeepening in which the lake is nowadays. Moreover the results of the numerical experiments of Kaplan et al. (2009), about the extent of ice related to the elevation of the accumulation area and the depth of the overdeepening, suggest that this overdeepening was most probably already carved before 1 Ma. Moreover, recently Christeleit et al. (2017) argued using thermochronometric data that the relief of Patagonia at the latitudes of our study was probably achieved between 10 and 5 Ma. On that basis, one can assume that the recycling time probably remained constant over the period covered by the samples. Hence the recycling time and weathering coefficients are considered the same between the different samples.

We used the theoretical scheme described in section 2.3.1 to estimate these weathering intensities and recycling time. Known or informed parameters in the Monte Carlo simulations were the activity ratios (^{234}U/^{238}U) and (^{230}Th/^{238}U), initial activity ratio (from the glacial flour sample), recoil loss factor f_α and deposition ages of the moraines (the post-depositional duration). Unknown parameters were the weathering intensities of ^{238}U, ^{234}U and ^{230}Th for the pre- and the post-deposition history. The relative weathering intensity of ^{234}U compared to ^{238}U, and ^{230}Th compared to ^{238}U, were assumed to be the same before and after moraine deposition. Consequently, we needed to determine five unknowns: the respective weathering coefficients k_{238}^{pre}, k_{238}^{post}, k_{234}/k_{238}, k_{230}/k_{238} and the recycling time t_{recycl} using Monte Carlo simulations. We chose a large, arbitrary number of values for the unknowns and calculated the activity ratios associated with these set of values. Then, we calculated a misfit M for the data as the difference between measured and modeled values, weighted by the uncertainty. The lower the misfit, the larger the probability of this set of values (we show the likelihood L for better visibility):

M=(m-o)$^T\sigma^{-1}(m-o)$

$L = \exp(-M/2)$

Where m is the vector of modeled parameters (the five unknowns above), o is the observed values (the activity ratios (^{234}U/^{238}U) and (^{230}Th/^{238}U)), and σ is the diagonal matrix of uncertainties on the data. In theory, an optimal solution can be determined, corresponding to the lower misfit (larger likelihood). However, no solutions were found because the number of unknowns was too great. We also know the relative weathering intensities of ^{238}U, ^{234}U and ^{230}Th : since Th is less soluble than U and ^{234}U is on more fragile mineralogic sites. Hence, k_{230} is lower than k_{238}, and k_{234} is slightly larger than k_{238}. This information was used to further constrain the Monte Carlo simulations. For different couples of k_{234}/k_{238} and k_{230}/k_{238}, we ran a Monte Carlo optimization to find an optimal solution (lower misfit). All optimal solutions for k_{238}^{pre}, k_{238}^{post} and the recycling time are shown in Figure 6, and we use the optimal solutions to characterize the pre-depositional duration of these sediments.

To test whether exchangeable Th might not be efficiently leached by our chemical protocol, we also made a simulation with $f_\alpha^{230} = 0$ and $f_\alpha^{230} = \dfrac{1.176 \times f_\alpha^{234}}{2}$ (half of the theoretical value described in section 2.3.1). If Th was totally readsorbed onto the mineral surfaces after ejection from the grain (and not subsequently removed during leaching), then it is equivalent to having no ejection ($f_\alpha^{230} = 0$). The case with $f_\alpha^{230} = \dfrac{1.176 \times f_\alpha^{234}}{2}$ is an intermediary between total removal of the adsorbed Th and no removal at all, discussed further in the Supplementary Information.

3 Results

3.1 Cosmogenic ^{10}Be and deposition ages

The results of ^{10}Be analyses are shown in Figure 2 and Tables 2 and 1. Refer to supplementary information for more details on the profiles ages results.

Eight surface cobbles from outwash relating to the Moreno 1 and 3 moraines yielded ^{10}Be exposure ages ranging from 168-269 ka (published in Hein et al. (2017)). To these we add six more surface exposure ages and two depth profile ages from outwash related to the Deseado and Telken moraines. Three cobbles from Deseado 1 produced relatively tightly-clustered exposure ages of 430-468 ka. A cobble from the ice-distal Deseado 2 outwash yielded a much younger, stratigraphically-inconsistent age of 293 ka (sample D2-T12), similar to the Moreno ages. Two further cobbles from the Deseado 2 outwash yielded ages of 520 ka and 618 ka, the oldest of which agrees, within errors, with a modelled depth profile age of 600 ka (+70/-35) from the same outwash unit. The agreement between older ages from surface cobbles and the Deseado 2 depth profile is unsurprising given that modelling suggests surface erosion (<0.2 cm.ka^{-1}) and inheritance within the outwash sediments (<2.10^4 atomg.g^{-1}) was relatively low (see Supplementary Information). There are a number of factors that can result in some age scatter within outwash surface cobbles, including surface deflation and up-freezing of clasts (Darvill et al. (2015); Hein et al. (2017)). Moreover, Hein et al. (2009,

2017) suggested that the oldest surface exposure ages from outwash in this region will most closely relate to deposition age. The D2-T12 sample is >200 ka younger than the other Deseado 2 cobbles, and it is likely that this cobble was either deposited at a much later time (perhaps during Moreno-stage glaciation, although the difference in altitude makes this unlikely) or was not in situ. The close agreement between other Deseado 2 ages supports the removal of D2-T12 as an outlier. Finally, a modelled depth profile through outwash relating to the Telken 5 moraine produced an age of 780 ka (+170/-80, 1σ confidence interval), with relatively low surface erosion (<0.1 cm ka-1) and inheritance (<6.10^4 atomg.g^{-1}). This age is within the lower range of 760-1016 ka known from radiometric ages (Singer et al., 2004). Sensitivity tests showed that modifications of the a priori values of erosion rates and inheritance lead to insignificant changes in the resulting age.

3.2 Evaluation of the U-Th chemical procedure with SEM images

For three samples taken randomly, we looked at SEM images after 50μm sieving and preparation (Figure 4) to assess the effects of our chemical and mechanical treatment. We were particularly interested in whether secondary phases could remain after processing, and if the primary silicates could have been altered by this treatment. The minerals observed are mainly quartz, feldspars and micas, corresponding to the lithology of the Andes in these latitudes. We list below the main observations :

- No trace of carbonate, organic matter or oxides were observed in samples that underwent the entire protocol. Prior to treatment, we only note the presence of oxides (Figure 4b). This does not, however, preclude the presence of carbonate or organic matter since a difference of mass before and after steps 2 and 5 of the protocol were observed. Therefore, the protocol seems efficient at eliminating carbonates, oxides, and organic matter.

- Samples were generally cleaner after preparation. This is because ≤ 4 μm fractions, containing mainly clays, were removed by Stokes settling. However, clays are still observed in the samples after preparation (see Figure 4b and c), mainly agglomerated around primary grains as clay pellets. In one sample, up to one third of the grains have clay pellets. These clays could have been precipitated directly at the surface of the grains during weathering or agglomerated subsequently. It is difficult to fully eliminate the clays and a potential bias could be introduced if too many clays remain in the sample. This potentially limits the application of comminution ages to samples with very low amount of clays, since their complete removal is currently not possible.

- Most micas show surfaces having experienced weathering (Figure 4d). The shapes observed are typical of weathering both before and after the protocol. This shows that our silts samples have experienced weathering and that it could potentially have affected U-Th disequilibrium.

- Following the preparation protocol, two feldspar grains in one sample displayed evidence of corrosion (Figure 4e and 4f). While this damage could be associated with alteration of the primary silicates during leaching, these are the only two corroded grains visible in the samples we tested. Since the corrosion pits are smaller than 4μm, and all the grains are larger than 4μm in our samples, ubiquitous damage would be observable on other grains. As such, we assume that primary mineral corrosion during the protocol is minimal.

3.3 U-Th - f_α - age

U-Th results are shown on Figure 5 and Table 3. (^{234}U/^{238}U) from moraines display little variation, ranging from 0.95 to 0.97, while glacial flour has a composition of 1.007, close to equilibrium. For the moraine samples, a general decrease of (^{234}U/^{238}U) is observed with time, though with significant noise. (^{230}Th/^{238}U) mostly vary between 0.94 and 0.97 with the youngest moraine sample (F1-U-M2, Fenix moraine) having a composition around 1. Similarly (^{230}Th/^{234}U) mostly range from 0.99 and 1.01, with the youngest moraine around 1.03. Again, an overall decrease in (^{230}Th/^{234}U) is observed with time. The glacial flour is close to equilibrium compositions ((^{230}Th/^{238}U)=1.02 and (^{230}Th/^{234}U)=1.01). Specific surface areas vary from 1.5 to 7 m^2/g, which is typical for this kind of samples, and the fractal dimensions range from 2.45 to 2.6. The recoil loss factors f_α calculated using equations 3 are between 0.005 and 0.025, with no observable trend with time.

Handley et al. (2013a) analyzed only a few samples with the gas adsorption technique to characterize the specific surface area and f_α. We analyzed every sample with this technique, highlighting the consistency of the noise in (^{234}U/^{238}U) and the dispersion in f_α derived from specific surface area (Figure 5). The relationship between (^{234}U/^{238}U), f_α and age is described by a surface in three dimensions. In other words, the noise observed in the (^{234}U/^{238}U)-age diagram, around the general decreasing pattern is in a large part associated with the dispersion of f_α. This suggests that specific surface area data are consistent and reliable, or that, if a bias exist in these data, it is systematic. Such a systematic bias would arise from a rather random event due to the sample processing, so this latter explanation seems unreasonable. We conclude that the specific surface area determination based on the gas adsorption technique is valid to evaluate the recoil loss factor in the U-Th comminution age theory.

We observe that (^{230}Th/^{234}U) is larger than (^{234}U/^{238}U) and (^{230}Th/^{238}U) has comparable compositions to (^{234}U/^{238}U). As mentioned in section 2.3.1, this observation is incompatible with a simple comminution age model (only α-recoil). An enrichment of ^{230}Th compared to ^{234}U could be the imprint of weathering on silts found in the moraines. Similarly, the incompatibilities of (^{234}U/^{238}U) and f_α in the framework of the simple comminution age model suggest that weathering must be considered. In the comminution age theory as described by DePaolo et al. (2006), $1 - f_\alpha$ must always be lower than (^{234}U/^{238}U), and for a sample old enough to have reached steady state, (^{234}U/^{238}U)=$1 - f_\alpha$. This is not what we observe, similarly to Handley et al. (2013a). However, as described in section 2.3.1 such an observation can be explained if weathering is considered and $k_{234} \geq k_{238}$.

Given the large disequilibrium of the youngest samples in (^{234}U/^{238}U) and in (^{230}Th/^{234}U) (Table 3 and Figure 5), and the fact that the glacial flour (the most probable inital composition for the moraine silts) is close to equilibrium, we cannot ignore the pre-depositional history (i.e. before deposition in the moraines). Based on a starting value close to equilibrium, the disequilibrium values measured here cannot be reached in only 20, or even 200 ka. Since our estimations of the f_α seem consistent and reliable, this requires consideration of a pre-depositional history involving weathering with a different intensity than during the post-deposition history.

3.4 Predeposition history and Monte Carlo analysis

We attempt to constrain the pre-depositional history using a Monte Carlo analysis. The results show an inverse relationship, with the greatest probability for $k_{238}^{pre}/k_{238}^{post}$ between 2 and 3 and a recycling time between 100 and 200 kyrs. The optimal values of k_{234}/k_{238} and k_{230}/k_{238} associated with this solution describe an anti-correlation with $1.2 \leq k_{234}/k_{238} \leq 2$ and $0.01 \leq k_{230}/k_{238} \leq 0.8$ (figure 6). These parameters are, however, poorly constrained by the inversion process.

Despite the numerous parameters to be constrained, the model converges for key results as shown above. We show in particular that, in the framework of the modified comminution age theory', incorporating weathering, described in section 2.3.1, we can say that the silts from the LBA moraines were likely eroded 100 to 200 kyrs before being deposited in the moraines and that, on average, they experienced around 2-3 times more intense weathering during this interval than after deposition (Figure 6). The best fit, corresponding to a recycling time of 180 kyrs, a ratio $k_{238}^{pre}/k_{238}^{post} = 2.35$, $k_{234}/k_{238} = 1.4$, and $k_{230}/k_{238} = 0.6$, is shown on supplementary Figure 5.

4 Discussion

4.1 Glacial chronology and perspectives on the control of ice extent in the Patagonian Andes

The new ^{10}Be exposure ages in this study help clarify the timing of deposition of several of the LBA moraines. The oldest outwash cobbles are taken as closest to the age of deposition because depth profiles show that average nuclide inheritance is relatively low and outwash surfaces show evidence for deflation (exposing younger cobbles; Hein et al. (2011, 2017)). It is possible that even the oldest cobbles underestimate the age of deposition if they have also been exhumed, and we apply no erosion correction to our exposure ages. As discussed in Hein et al. (2017), outwash surface cobbles imply that the Moreno moraines were deposited at ca. 260-270 ka, during Marine Isotope Stage 8.

Unlike the Moreno system, surface cobbles from the Deseado moraines appear to date from two different glacial cycles. Exposure ages from Deseado 1 outwash yield relatively tightly clustered ages of 430-470 ka, suggesting that the limit was deposited during MIS 12. The older of two published moraine boulder exposure ages from the Deseado 1 moraine (erosion-corrected to 476 ka; Kaplan et al. (2005)) is consistent with the cobble ages in this study. In contrast, surface cobbles from Deseado 2 outwash yield ages of 520 ka and 618 ka (excluding the anomalously young D2-T12 age of 293 ka). The dating would be less conclusive without the accompanying depth profile age of 600 +70/-35 ka. Slight changes in scaling factors (see section 2.2.3) do not affect this finding. Taken together, the oldest surface cobble and depth profile suggest that the Deseado 2 limit was deposited at ca. 600-620 ka. Within errors, the limit may relate to MIS 16. The scatter in ages is interesting given that luminescence dating yielded an even younger age of 123 ± 18 ka for the Deseado 2 limit (Smedley et al., 2016). This scatter may imply greater sediment reworking or a more complex relationship between moraines and outwash in this sequence. The Telken 5 depth profile gave an age of 780 +170/-80 ka that is stratigraphically consistent within our chronological dataset, but less helpful in determining when the limit was deposited. The error range spans MIS 18-24 (and radiometric datings gave a lower age of 760 ka for the Telken serie), with larger probability around MIS 20, so it is possible that the moraine was deposited during MIS 20. Since the entire Telken moraine system must be older than 760 ka (Singer et al., 2004), this implies that Telken 1 to 4 are also that age, and that the Telken moraines represent different glacial advances during the same glacial cycle.

In summary, there is a clear pattern in the timing of moraine deposition in which alternate glacial cycles are represented in our chronology: Telken 5 during MIS 20; Deseado 2 during MIS 16; Deseado 1 during MIS 12; and the Moreno moraines during MIS 8. Previous work has shown that the innermost Fenix moraines relate to the Last Glacial Maximum during MIS 2 (Kaplan et al. (2004); Douglass et al. (2006); Smedley et al. (2016)). We strongly caution that scatter in surface cobble ages and error ranges in depth profiles may complicate this pattern, particularly for the older limits. Moreover, we did not analyse the enigmatic Deseado 3 moraine and so cannot say whether this relates to the counterparts dated here or intervening glacial stages. However, dating of the Moreno and Deseado 1 and 2 moraines does imply that the intervening glacial cycle is absent from the record.

The pattern in timing of moraine deposition around Lago Buenos Aires implies that there are a few glacial cycles that are either not recorded in this area, or have been erased or removed. Either the ice lobe did not advance during alternate glacial cycles, or advanced to similar or less extensive positions so that moraines and outwash were then removed by the following advance. A similar pattern was observed by Hein et al. (2009, 2011) for the Lago Pueyrredon ice lobe, 100 km to the south, and so there could be a regional driver of alternate glacial advances. One possibility is that erosion over successive glacial cycles caused entrenchment of ice lobes within large basins on the eastern side of the Patagonian Ice Sheet (Kaplan et al., 2009). This erosion model has been linked to the pattern of nested limits seen across the former ice sheet (Kaplan et al., 2009; Anderson et al., 2012) but may be more complex if only alternate cycles are represented in the moraine record. Alternatively, a purely climatic forcing could have caused alternate strong advances, although there is no simple relationship between available climate records and alternate glacial advances at Lago Buenos Aires. A complex erosion-climate feedback mechanism may determine when or how far glacial advances occurred in this region, but more detailed glacial models are required to further test such a model.

The chronology presented here suggests that moraines reworking has probably occured in the area, and/or that glacial erosion in the Andes may feedback into the glacial lobe advance, to produce this observed pattern of absent intervening glacial cycles. Regardless, this chronology allows us to broadly constrain deposition ages of moraines targeted for U-Th analysis, which may in turn inform sediment recycling times between preserved moraines.

4.2 Implications for sediment recycling, chemical weathering, climate and their interactions, in proglacial systems

Following the conclusions of Handley et al. (2013a), this residence time of 100-200 kyrs could be an artifact due to an addition of old dust. In our area, when a new moraine is being formed, the older sediments are on the east, whereas the dominant winds comes from the west. So it is unlikely that these winds could add older material to the one being deposited.

The long time estimated using the whole set of data and the Monte Carlo simulations, along with the absence of inheritance in the ^{10}Be data, suggest that the sediment was not exposed at the surface in the proglacial system before being deposited in its moraine. This implies that the sediment must have been buried, either in the proglacial lake, or within a sedimentary pile (moraine, till, channel, etc. at least a few meters below the surface) before final deposition.

The dated sediment was most probably eroded during glacial periods, as erosion rates are likely much larger than during interglacial. Likewise, deposition in an end moraine occurs during a glacial period. This means that the sediment deposited in the moraine may spend on average as long as 100-200 kyrs in the proglacial system (till, lakes sediments, etc.) before being deposited in the moraines. Hence the sediment eroded during a glacial period is, on average, deposited in the frontal moraine during the next glacial cycle. Using postglacial sediment budget and reservoir theory, Hoffmann & Hillebrand (2016) modelled the residence time of sediments in a periglacial system in the Canadian Rocky Mountains, and also found a time of 100 kyrs. This average residence time implies that a fraction of the sediment deposited in a moraine may have been produced during the same glacial cycle and with an other fraction being much older.

This time appears to be long. However, our measurements of cosmogenic exposure ages using ^{10}Be nuclide concentrations in outwash cobbles and profiles suggest that the preserved moraines represent every other glacial cycle, indicating an advance over the proglacial sediment of one or two previous advances.

Moreover, it has been shown that proglacial lakes occupying overdeepenings are filled following deglaciation (Eyles et al., 1991; Houbolt & Jonker, 1968). The lakes are filled with sediments during interglacial periods, and emptied during the subsequent glacial periods. These processes imply sediment erosion and deposition over a full glacial-interglacial cycle which is consistent with both ^{10}Be and U-Th disequilibrium data.

We also obtain weathering rate that is 2-3 times higher during the 100-200 kyrs of the predepositional phase than after deposition. Based on the argument above, these sediments are exposed to water in the proglacial system before deposition in the moraine. Authors such as Anderson (2005) have shown that proglacial environments favor weathering. Since weathering rate is a time dependent parameter (e.g. White & Brantley (2003)), this recycling time of 100-200 kyrs has a likely non-negligible impact on weathering fluxes.

Understanding how sediments are evacuated and transported to the oceans, and when they experience weathering would help to quantify the relationships between erosion and climate. In particular, if 100 to 200 kyrs are needed to escape the periglacial area, important lags could be observed between the erosion forcing, and the weathering and climate response. This suggests that measured weathering variations could have occured over the last glacial-interglacial cycles, or have been smoothed/damped because of a lag in the sediment transport. Estimating these lag times would help understand the relationships between erosion, climate, and marine biogeochemical cycles. Erosion rates can be determined using thermochronology or cosmogenic isotopes. Sedimentation rates in the ocean can be estimated as well using marine core dating methods. The link between erosion and sedimentation is poorly known (Sadler, 1981) especially because the transport times are poorly constrained. The transport time of 100 to 200 kyrs presented here suggests that the pathway between initial erosion and deposition is potentially complex. An effort to constrain these transport times appears to be potentially fruitful to reveal the actual link between erosion/sedimentation rates and climate.

4.3 Perspectives on comminution age method and its applications to characterize sediment transfer

Our data let us estimate only a mean recycling time for moraine sediment (the fine silty fraction). Here we have discussed it in terms of recycling of the previous glacial cycle moraine, but it could also be interpreted as a mixing of much older, deeper, reworked sediment with new freshly eroded sediment. In other words, we do not quantify the amount of sediment escaping the proglacial system, or the amount of sediment which is deposited in the moraine rapidly after erosion. It would be beneficial to think about ages distributions, and not only mean ages. However, this necessitates being able to measure U-Th disequilibrium on single grains, which remains an analytical challenge (Bosia et al., submitted).

Quantifying or reducing the effects of weathering remains a major challenge. Pure primary minerals with no clays would minimise these secondary processes. To this end, working on pure zircons grains could be an option. This would require improved mineral separation methods for fractions smaller than 50 μm. One would also have to assume that comminution effectively occured on such a heavy mineral, and that there is no initial disequilibrium (a problem being that zircons are much enriched in U than there surrounding minerals).

5 Conclusions

Our study is a first attept to quantify long term sediment transfer times in proglacial area. We take advantage of the particularly well preserved series of nested moraines of the Lago Buenos Aires in Patagonia. Our approach involves ^{10}Be exposure dating of these moraines combined with U-Th disequilibrium measurement on the fine fraction of moraine sediment, within a 'modified' comminution theoretical framework, to characterize the pre-deposition and post-deposition histories of the sediments.

We show that weathering cannot be neglected when determining the comminution age. Measuring Th isotopes helps constrain the weathering process. Even with this complication, it remains possible to calculate comminution ages if the studied samples have well constrained deposition ages and have experienced the same predepositional history. In this case, the weathering intensities and the predepositional history duration are the same for all the samples. We also show that specific surface area measurements based on the gas adsorption technique for estimation of the recoil loss factor is a reliable method, and should be applied to all samples. One caveat is that clay minerals may not be removed from the samples, especially clay pellets agglomerated at some mineral surfaces, which could be a strong limitation of the use of this method to calculate transport times.

Our new chronology shows that some glacial cycles are either not preserved or not recorded in the LBA area, i.e. respectively the associated moraines may have been removed by following glacial advances or may have been deposited farther upstream. Hein et al. (2011) found a similar pattern in Lago Pueyrredon area, suggesting that the mechanisms driving glacial advance may have a regional extent. Future work may reveal wether erosion feedbacks in the Andes could be responsible for this pattern.

The absence of moraine records for a few glacial cycles in the LBA area can be explained by our U-Th data showing that there has been reworking/recycling of the sediments. Using a Monte Carlo approach we estimate that the silts from the Lago Buenos Aires frontal moraines system have 100 kyr residence time in the proglacial system (transported in the lake sediments, remobilized from the previous moraines or stored in intermediate reservoirs), and experienced three times more intense weathering before deposition than in the moraine. Our data represent a step toward the effort of constraining the pathways and timescales over which sediments are transported from source (erosion in mountain belts) to sink (ocean sediments). Although considered as a daunting challenge by Sadler & Jerolmack (2015) and requiring additional development, this approach can contribute to our understanding of basin-scale sediment budgets and how erosion, weathering, and sedimentation evolve through time.

Code availability. TEXT

Data availability. TEXT

Code and data availability. TEXT

Author contributions.

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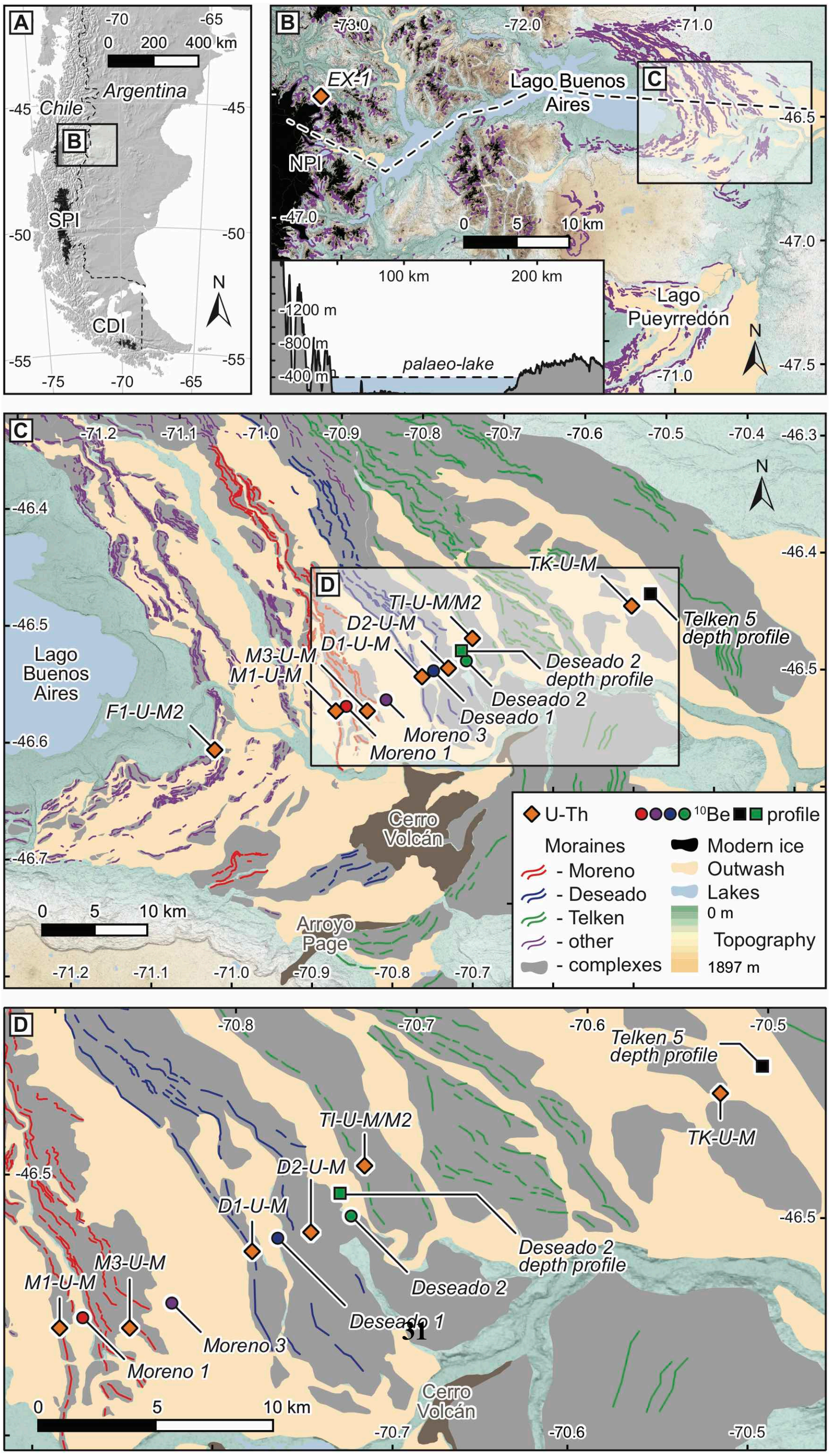

Figure 1. Map of the study area, adapted from Bendle et al. (2017). [A] Southern South America showing the location of Lago Buenos Aires. The three main contemporary ice masses in Patagonia are also shown: The Northern (NPI) and Southern Patagonian Icefields (SPI) and the Cordillera Darwin Icefield (CDI). [B] Inset shows a west-east transect through the study area, from the Patagonian Andes to the frontal moraine systems of the proglacial lake (adapted from Kaplan et al. (2009)); sample locations as per [a]. [C] and [D] The series of frontal moraines sampled in this study. Silt samples are shown by red circles and cobbles and profiles for cosmogenic analysis are shown in blue; the glacial flour sample at the mouth of Los Exploradores Glacier is shown; and the extent of the present day ice field is shown in black. Successive glaciations have formed the large over-deepening in which the proglacial lake has formed.

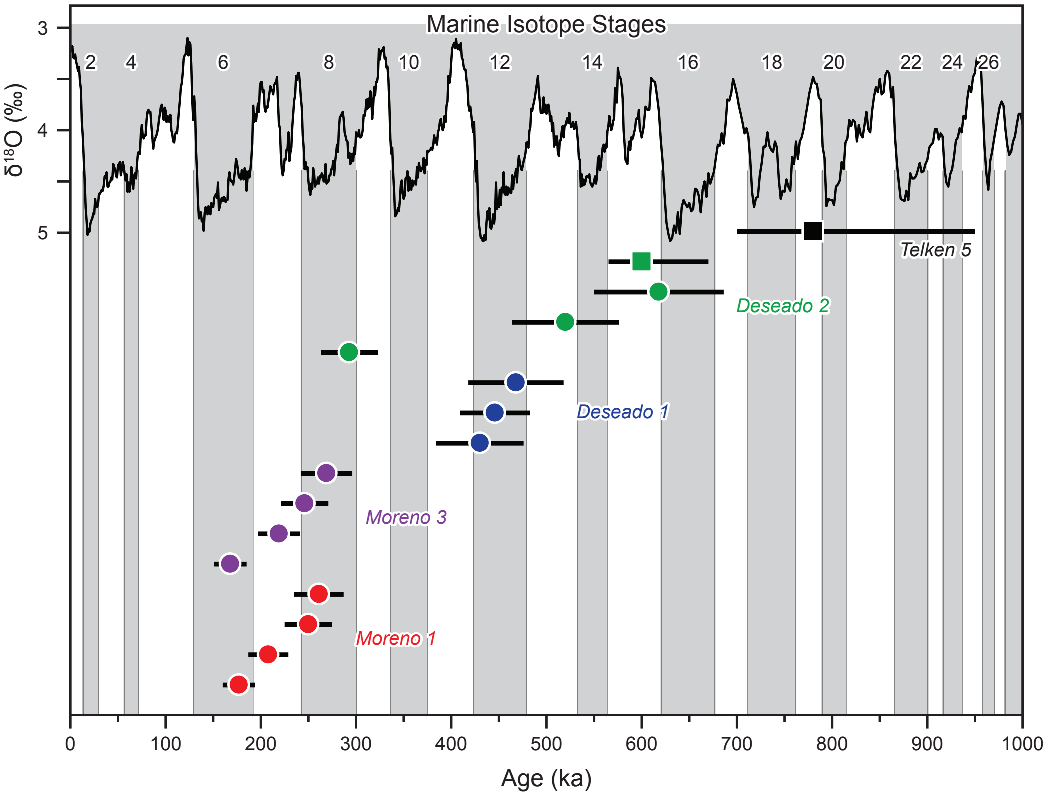

Figure 2. ^{10}Be exposure ages from outwash surface cobbles (circles) and modeled depth profiles (squares) for five of the Lago Buenos Aires moraines. Depth profile uncertainties are 1σ. Symbols and colors are the same as in figure 5. The $\delta^{18}O$ benthic stack record from Lisiecki Raymo (2005) is also plotted, and grey shading shows glacial stages.

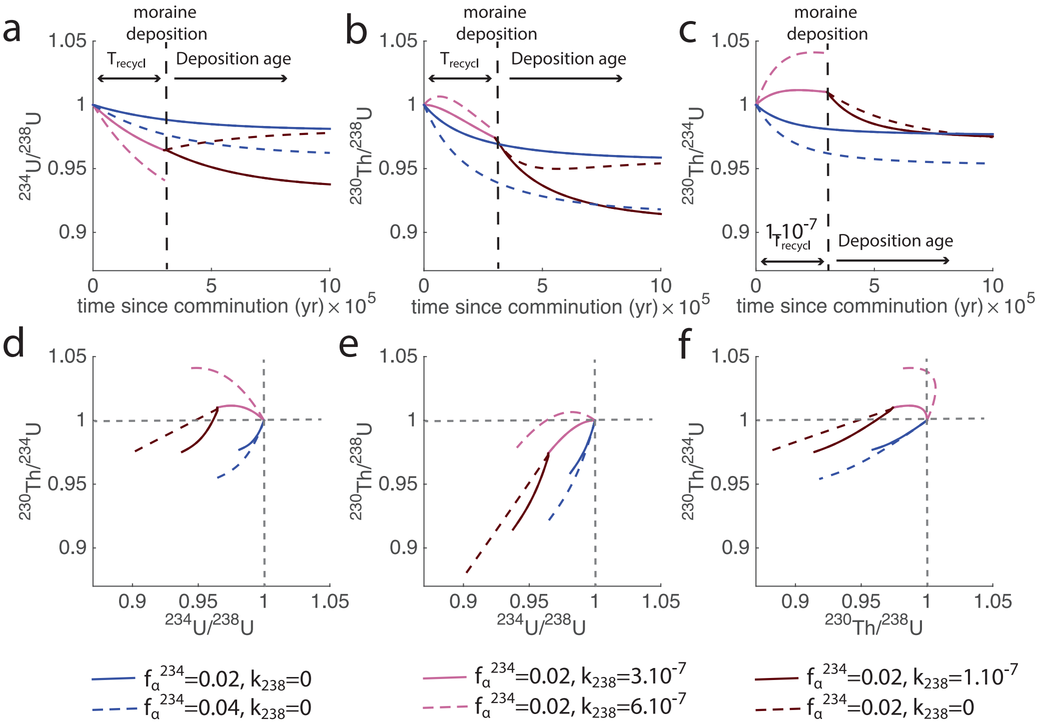

Figure 3. Solutions to the equations 2.3.1 that we use as the theoretical framework to analyze our data, for different values of f_α and k_{238} (We consider k_{234}/k_{238}=1.4 and k_{230}/k_{238}=0.5). (upper panels: a, b and c) the time evolution of $^{234}U/^{238}U$ (a), $^{230}Th/^{238}U$ (b) and $^{230}Th/^{234}U$ (c) activity ratios. The vertical dashed line represent an event through which the weathering intensity vary: this corresponds in our case to the time of sediment remobilization. Our ^{10}Be exposure ages give the time elapsed since this event (materialized by this dashed vertical line) whereas the U-Th data give the dureation of the first phase between comminution and remobilization, i.e. between Y axis and this dashed vetical line. (lower panels: d, e and f) the co-evolution of these ratios presented above. In order to make sense of the complexity of the different possible patterns in the framework of the comminution model incorporating weathering (see section 2.3.1. Different cases are plotted, corresponding to different f_α values, and different weathering intensities (see legend). Observing these figures helps to understand the range of variations of the different activity ratios, and the effect of each parameter (α-recoil and weathering) on these ratios, before and after remobilization of sediments.

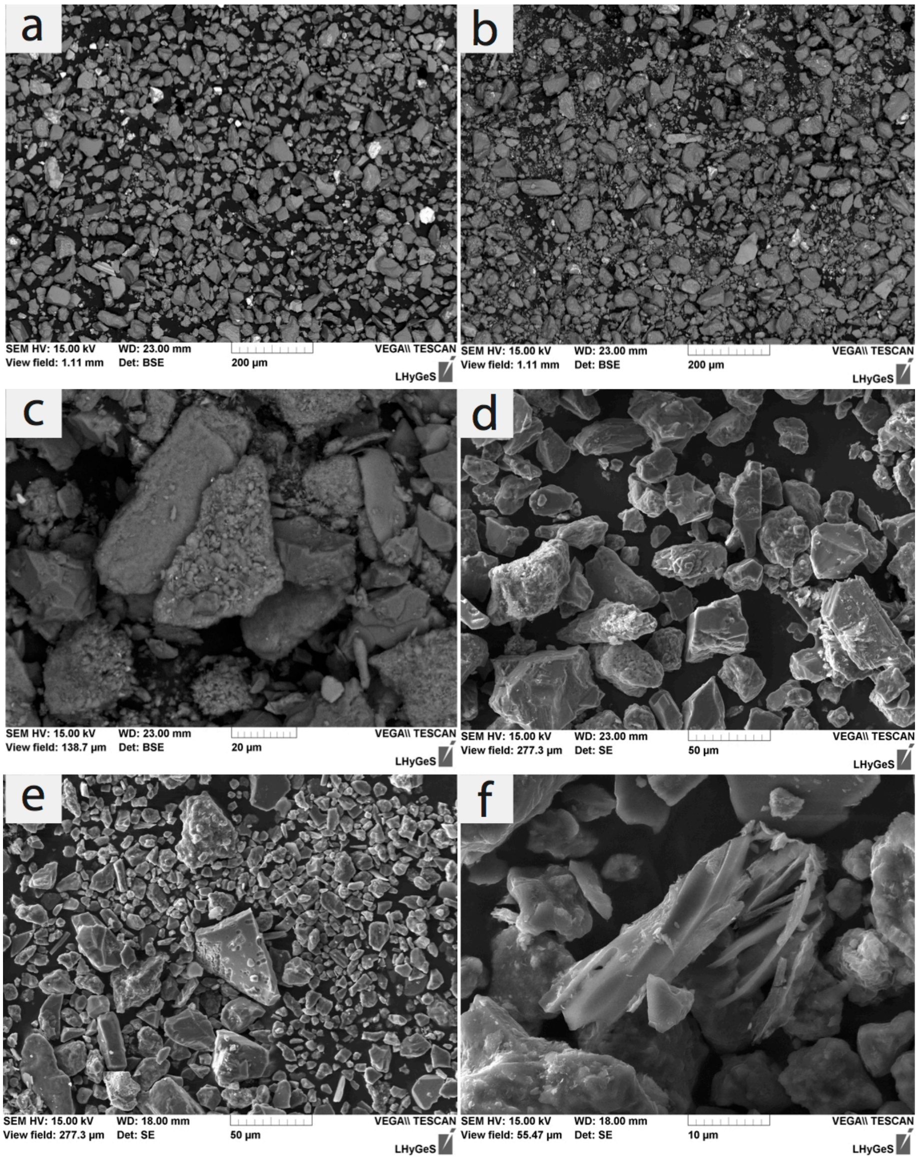

Figure 4. Scanning Electron Microscopy (SEM) images of selected samples. [a] Overview of a sample after the preparation process described in section 2.3.2. Grains vary in size and shape between 4 and 50μm and angular to subangular. White grains are heavy minerals such as zircon or oxides. [b] The same sample before 50μm sieving. The only notable difference is the presence of clays. [c] Zoomed image of a mineral grain covered in clays pellets that were not removed by the preparation process. [d] Mica grains with cauliflower faces, characteristic of weathering. [e] and [f] Feldspar grains with corroded surfaces. It is unclear whether these corrosion features result from geological processes or the sampling procedure, but they were only observed twice.

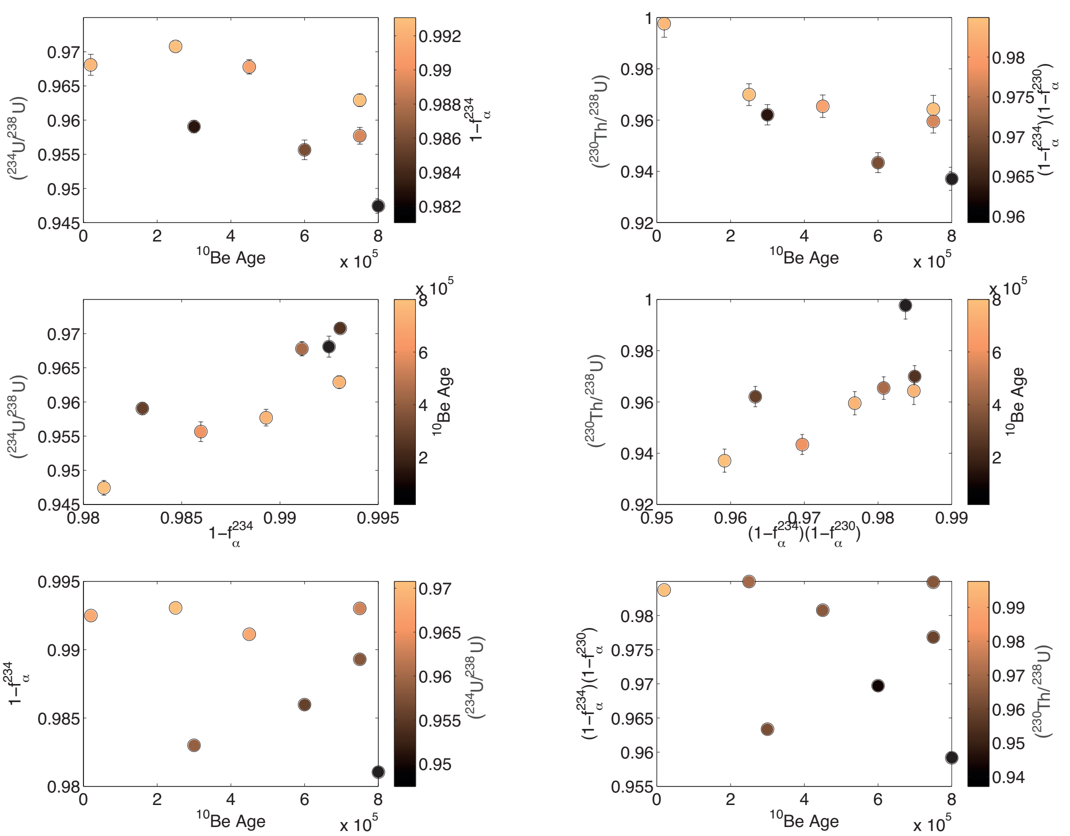

Figure 5. 2D cross plots of ^{234}U/^{238}U and ^{230}Th/^{238}U activity ratios; associated recoil loss factors $(1 - f_{\alpha}^{234})$ and $(1 - f_{\alpha}^{234}) \times (1 - f_{\alpha}^{230})$); and ^{10}Be exposure ages. Symbol colours represent the third parameter in each plot. Sorting observed in the symbol colours helps to explain the noise associated with the other two parameters. In particular, the scatter in the decrease of the activity ratios over time can be largely explained by the variability in the recoil loss factor (top left plot). The plots also let us assume that the variability in the weathering coefficients could be low, otherwise a residual dispersion would be observed. In all cases, error bars are smaller than the size of the dots.

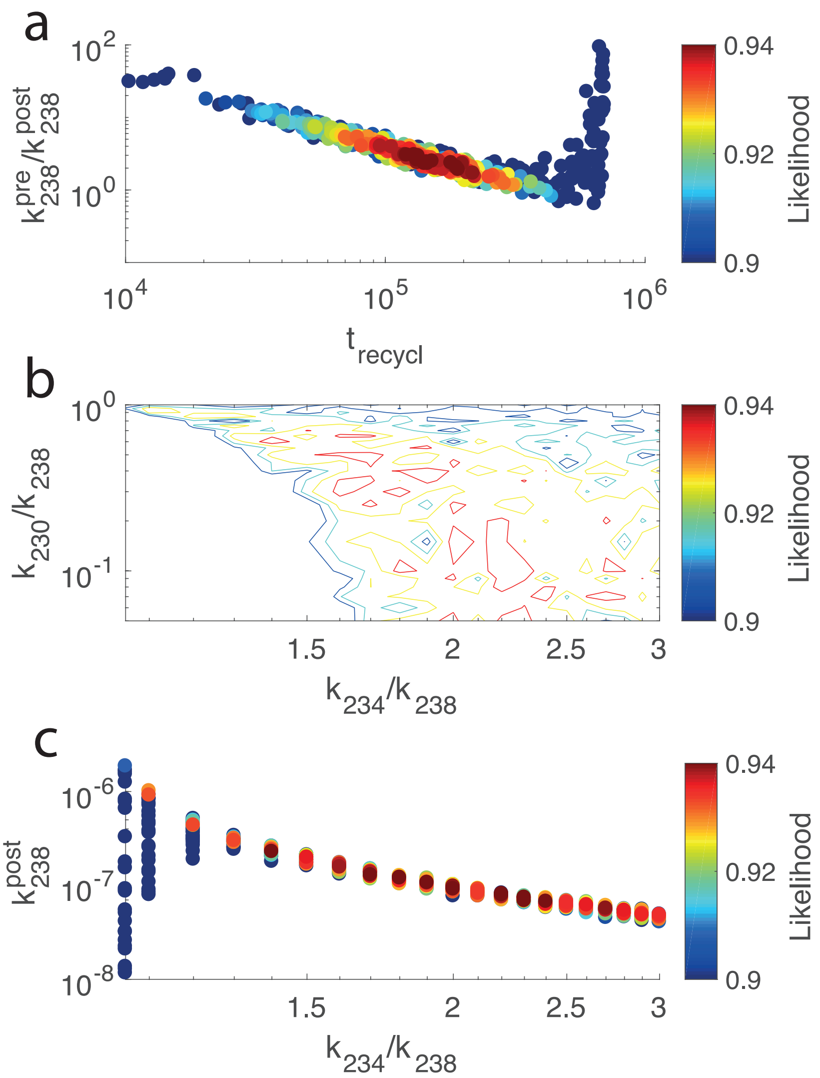

Figure 6. Results of Monte Carlo simulations for the U-Th data. The same Monte Carlo simulations were conducted for different fixed values of k_{234}/k_{238} and k_{230}/k_{238} (see section 2.3.5). For each simulation we estimated the optimal solution for k_{238}^{pre}, k_{238}^{post} and recycling time (pre-deposition duration, after comminution), i.e. the solution with the lowest misfit between measured and calculated activity ratio data. We used this misfit to calculate a likelihood between 0 and 1 (see equation 2.3.5), shown by the colourmap (red is lower misfit, blue is higher misfit, stretched to fit our results). The larger the likelihood, the closer to the data the solution is, so the more likely the set of parameters values is. [a] The ratio $k_{238}^{pre}/k_{238}^{post}$ as a function of the recycling time. Each dot is one solution to the Monte Carlo. The optimal solution (red dots) is for three times stronger weathering before deposition and a recycling time of 100 ka. [b] Relative weathering intensities of ^{234}U, ^{238}U and ^{230}Th. The relative weathering intensities are poorly costrained, but a relationship can be extracted between their ratios : the larger k_{234}/k_{238}, the smaller k_{230}/k_{238} must be. [c] A relation between k_{238}^{post} and k_{234}/k_{238} can be extracted, even if both parameters are poorly constrained. So the estimation of sediment transfer times using U-Th disequilibria series could be improved with better constraints on the relative mobility ^{234}U, ^{238}U.

Table 1. Cosmogenic ^{10}Be nuclide data for outwash surface cobbles and depth profile samples. Moreno 1 and 3 data are already published in Hein et al. (2017).

Sample ID	Latitude (DD)	Longitude (DD)	Elev. (m asl)	Depth (cm)	Shielding correction	Thickness (cm)	^{10}Be/^{9}Be ($\times10^{-12}$)	Sample weight (g)	^{9}Be added by carrier (10^{-4} g)	^{10}Be $\pm 1\sigma$ (10^{5} at.g^{-1})
Moreno 1 outwash surface cobbles										
M1-T5	-46.5544	-70.8758	496	-	1	7	2.097 ±0.040	39.5464	3.046	10.75 ± 0.21
M1-T9	-46.5544	-70.8758	496	-	1	6.5	3.438 ±0.062	51.9881	2.867	12.64 ± 0.23
M1-T12	-46.5544	-70.8758	496	-	1	7	3.912 ±0.070	48.1040	2.875	15.59 ± 0.28
M1-T13	-46.5544	-70.8758	496	-	1	6	4.513 ±0.081	57.1220	2.864	15.09 ± 0.27
Moreno 3 outwash surface cobbles										
M3-T6	-46.547	-70.8268	482	-	1	7	1.939 ±0.058	38.8251	3.053	10.14 ± 0.31
M3-T11	-46.547	-70.8268	482	-	1	7	3.282 ±0.059	45.9404	3.052	14.53 ± 0.26
M3-T14	-46.547	-70.8268	482	-	1	4	4.299 ±0.116	61.5663	2.869	13.36 ± 0.36
M3-T16	-46.547	-70.8268	482	-	1	5	4.965 ±0.089	58.8835	2.857	16.07 ± 0.29
Deseado 1 outwash surface cobbles										
D1-T1	-46.5219	-70.7732	522	-	1	4	1.667 ±0.034	12.3329	2.866	25.75 ± 0.54
D1-T3	-46.5219	-70.7732	522	-	1	7	2.190 ±0.042	15.3978	2.863	27.10 ± 0.53
D1-T12	-46.5219	-70.7732	522	-	1	6.5	4.182 ±0.075	30.7913	2.882	26.11 ± 0.47
Deseado 2 outwash surface cobbles										
D2-T5	-46.5140	-70.7364	535	-	1	3	2.609 ±0.047	16.0937	2.879	31.09 ± 0.57
D2-T6	-46.5140	-70.7364	535	-	1	3	4.787 ±0.086	25.3782	2.874	36.16 ± 0.65
D2-T12	-46.5140	-70.7364	535	-	1	3.5	2.364 ±0.042	24.5217	2.882	18.50 ± 0.34
Deseado 2 outwash depth profile										
D2-50	-46.5015	-70.7368	544	50	1	4	2.889 ±0.052	37.9697	3.005	15.23 ± 0.28
D2-60	-46.5015	-70.7368	544	60	1	4	2.189 ±0.045	37.0362	3.044	11.97 ± 0.25
D2-70	-46.5015	-70.7368	544	70	1	4	1.905 ±0.037	37.1552	3.037	10.36 ± 0.21
D2-130	-46.5015	-70.7368	544	130	1	4	0.858 ±0.032	37.1738	3.050	4.65 ± 0.18
D2-230	-46.5015	-70.7368	544	230	1	4	0.307 ±0.013	43.5459	3.056	1.40 ± 0.07
Telken 5 outwash depth profile										
TK-50	-46.4439	-70.5069	552	50	1	4	2.402 ±0.053	27.4445	2.856	16.65 ± 0.37
TK-60	-46.4439	-70.5069	552	60	1	4	2.826 ±0.050	36.0118	3.045	15.92 ± 0.29
TK-70	-46.4439	-70.5069	552	70	1	4	2.516 ±0.049	35.8816	3.042	14.20 ± 0.28
TK-130	-46.4439	-70.5069	552	130	1	4	0.903 ±0.052	35.4972	3.048	5.13 ± 0.30
TK-240	-46.4439	-70.5069	552	240	1	4	0.417 ±0.024	42.5856	3.039	1.94 ± 0.12

Table 2. Modeled exposure ages for outwash surface cobbles and depth profile samples (see main text and Supplementary Information for details of profile modeling). Moreno 1 and 3 ages have been already publised in Hein et al. (2017).

Sample ID	Age (ka)	External error (ka)	Profile age (ka)	Upper error (ka)	Lower error (ka)
Moreno 1 outwash surface cobbles					
M1-T5	177	17			
M1-T9	208	21			
M1-T12	**261**	**26**			
M1-T13	250	25			
Moreno 3 outwash surface cobbles					
M3-T6	168	17			
M3-T11	246	25			
M3-T14	219	22			
M3-T16	**269**	**27**			
Deseado 1 outwash surface cobbles					
D1-T1	430	46			
D1-T3	**468**	**50**			
D1-T12	446	47			
Deseado 2 outwash surface cobbles					
D2-T5	520	56			
D2-T6	**618**	**68**			
D2-T12	*293*	30			
Deseado 2 outwash depth profile					
			600	70	35
Telken 5 outwash depth profile					
			780	170	80

Table 3. U-Th concentrations, activity ratios and BET data (specific surface areas and fractal dimensions, and calculated recoil loss factors). Duplicates are also shown, two for the whole process (including sieving, leachings, HF digestion, chromatography), and one for HF digestion and chromatography. The samples are sorted by deposition age (F1=Fenix, M1=Moreno 1, M3=Moreno3, D1=Deseado 1, D2 = Deseado 2, TI=Telken 1, TK=Telken 5).

sample	moraine	Latitude	Longitude	[U] (ppm)	2SE	[Th] (ppm)	2SE	Specific surface area (m2.g-1)	Fractal dimension	f_α^{234}
EX-1	glacial flour	-46.5026	-73.1642	4.186	0.010	22.70	0.06	1.051	2.49	0.0047
F1-U-M2	Fenix 1	-46.5974	-71.0359	1.680	0.002	6.36	0.01	1.831	2.52	0.0075
M1-U-M	Moreno 1	-46.5587	-70.8883	1.855	0.003	7.63	0.01	1.392	2.46	0.0069
M3-U-M	Moreno 3	-46.5574	-70.8494	2.148	0.003	8.67	0.02	4.719	2.56	0.0170
D1-U-M	Deseado 1	-46.5255	-70.7837	1.928	0.004	8.00	0.02	2.238	2.53	0.0089
D2-U-M	Deseado 2	-46.5170	-70.7518	1.503	0.002	5.94	0.01	3.535	2.53	0.0140
TI-U-M	Telken 1	-46.4904	-70.7240	1.633	0.003	6.56	0.01	2.081	2.45	0.0107
TK-U-M	Telken 5	-46.4552	-70.5292	1.593	0.003	6.96	0.02	5.435	2.57	0.0190
Duplicates		***Observation***								
M3-U-M 2	duplicate of the whole process			2.067	0.004	9.38	0.02	7.034	2.59	0.022
M3-U-M 2 bis	duplicate from the HF digestion step			1.990	0.003	8.52	0.02			
TI-U-M 2	duplicate of the whole process			1.776	0.003	6.92	0.01	1.818	2.54	0.0070

sample	moraine	(^{238}U/^{232}Th)	2SE	(^{234}U/^{238}U)	2SE	(^{230}Th/^{238}U)	2SE	(^{230}Th/^{234}U)	2SE
EX-1	glacial flour	0.561	0.002	1.007	0.001	1.022	0.005	1.015	0.005
F1-U-M2	Fenix 1	0.803	0.002	0.968	0.001	0.998	0.005	1.031	0.006
M1-U-M	Moreno 1	0.739	0.001	0.971	0.001	0.970	0.004	0.999	0.004
M3-U-M	Moreno 3	0.753	0.002	0.959	0.001	0.962	0.004	1.003	0.004
D1-U-M	Deseado 1	0.733	0.002	0.968	0.001	0.965	0.004	0.998	0.005
D2-U-M	Deseado 2	0.769	0.001	0.956	0.001	0.943	0.004	0.987	0.004
TI-U-M	Telken 1	0.757	0.002	0.958	0.001	0.960	0.005	1.002	0.005
TK-U-M	Telken 5	0.696	0.002	0.947	0.001	0.937	0.004	0.989	0.005
Duplicates									
M3-U-M 2		0.671	0.002	0.955	0.001	0.964	0.006	1.009	0.006
M3-U-M 2 bis		0.711	0.002	0.955	0.001	0.962	0.004	1.006	0.005
TI-U-M 2		0.780	0.001	0.963	0.001	0.964	0.005	1.001	0.006