Dear Mark,

On behalf of my co-authors I'm resubmitting to you here our revised manuscript entitled "Regional Pliocene Exhumation of the Lesser Himalaya in the Indus Drainage" for publication in Solid Earth. In this letter we describe the changes that we have made to the manuscript in response to the detailed reviews. We only focus on the negative issues and where we disagree with the reviewers we explained that.

Best regards Peter

Response to anonymous reviewer #1

....The authors use the percentage of total zircon age population to determine which source is eroding most rapidly at different points in time. Based on this they argue for a previously unrecognized Quaternary exhumation event within the inner Lesser Himalayan Sequence (iLHS). The authors acknowledge that this interpretation of their data can only be made consistent with published bedrock zircon (U-Th)/He data from same region if iLHS rapid exhumation was not accompanied by its exposure at the surface. This would require rapid erosion of the overlying outer LHS and/or Greater Himalayan Sequence depending on the geometry of the underlying thrust or duplex. There is no indication of this rapid erosion of overlying material present in their data.

Response: We agree that this would require rapid erosion of the Greater and/or Tethyan Himalaya, although not necessarily the Outer Himalaya, which were also largely buried at this point of time (Webb, 2013). Contrary to what is stated, the record provided in this study does provide evidence for this erosion because there is a steady increase in erosion of Greater/Tethyan Himalayan material since the Miocene, accelerating after 5.7 Ma, as shown by the drift to more negative ε_{Nd} bulk sediment values. The zircon U-Pb ages are dominated by Greater/Tethyan Himalayan grains throughout the record, also indicative of erosion from these sources.

Furthermore their interpretation of a Quaternary iLHS exhumation event is inconsistent with data from Najman et al. 2009 (who the authors cite). Najman et al. (2009) show that the LHS in this region was exposed at the surface and contributing sediment to the foreland basin by 9 Ma. The results of Najman et al. (2009) are consistent with the widely recognized 10 Ma thrust wedge reorganization during which the Main Boundary Thrust (MBT), the fault underlying the LHS, became active across most of the Himalaya.

Response: Yes, this new record is not the same as the earlier work by Najman et al., but then they are not measuring the same thing. The deep sea fan record is compatible with the record of Najman et al. (2009), in that the initial pulse of Lesser Himalayan zircon appears after 8.3 Ma, not long after the erosion shown by this earlier work after 9 Ma. The Siwalik record considered by Najman et al. (2009) reflects the erosion history in that particular part of the foreland basin, likely a paleo-Beas River, but does not constrain other regions of the Indus catchment. The Jhellum, Chenab, Ravi and most notably trunk Indus Rivers would never have flowed through this part of the basin as the regional topography forces them to flow to the SW away from the

range front. This means that the Lesser Himalaya could have been exposed in the paleo-Beas valley but not to much extent in other parts of the basin. Just because the Lesser Himalayas were exposed in the area studied by Najman does not require them to be exposed everywhere else in the catchment.

Neither the submarine fan or the Siwalik records directly relate to fault motion, only to exposure of units to erosion. In this respect the data are "plausibly consistent" with the bedrock thermochronology data as the new study does not deny cooling of the Lesser Himalaya in the Late Miocene, 6–10 Ma (Caddick et al., 2007), only the lack of regional exposure and erosion.

It should also be noted that LHS age consistent zircons are well known components of the granites of Nanga Parbat. The increase in LHS age consistent zircons at 1.9 Ma may reflect the well documented post 3 Ma rapid exhumation of Nanga Parbat e.g. Koons et al. 2002 (and many other papers).

Response: Regarding Nanga Parbat as a possible source of the Lesser Himalayan zircon grains seen in the submarine fan we argue that this is unlikely because only $\sim 16\%$ of the zircons in the modern trunk river downstream of Nanga Parbat are >1500 Ma and of Lesser Himalayan affiliation. We do not ignore the erosion of Nanga Parbat and our results are compatible with the recent cooling noted by Koons et al. (2002). As noted in the manuscript the shift in bulk Nd isotope since the Miocene appears to be 35% caused by Nanga Parbat and 65% by the rest of the Lesser Himalaya. Nanga Parbat is too small and its erosion is insufficient to drive all the observed changes by itself.

Alternatively the increase in LHS age consistent grains at 1.9 may be driven by rapid exhumation of the proximal foreland basin that accompanied the Quaternary initiation of faulting along the Main Frontal Thrust in the area. Since the LHS is the nearest bedrock to the foreland basin it follows that its detritus makes up a significant proportion of the immediate foreland basin. The onset of MFT faulting and accompanying exhumation of the foreland basin should be considered an alternative driver of the shift in percentage of zircon ages at 1.9 Ma.

Response: Regarding the role of recycling of older sediments from the Siwalik Group as a source of the Lesser Himalayan material, we do not agree that the "Lesser Himalaya Series is the nearest bedrock to the foreland basin it follows that its detritus makes up a significant proportion of the immediate foreland basin". Tectonic juxtaposition does not transfer material into the sedimentary record, only unroofing and erosion can do that. Unfortunately, there is no zircon U-Pb dating of the Siwalik Group within the Indus catchment so any estimate of its influence is based on data from Nepal where the stronger monsoon rains may have driven faster erosion. There are indeed >1500 Ma zircons in the Nepalese Siwaliks, but a mass balance of these drainages by Lavé and Avouac (2001) indicated that only ~15% of the net erosion could be from Siwalik sources. Furthermore, an isotope-based mass balance for the Ganges catchment placed the influence of the Siwaliks at <10% (Wasson, 2003). Similar modest values would seem appropriate in the western Himalaya too, as the zircon U-Pb populations of the major modern rivers are quite diverse and are largely consistent with the bedrock geology of their mountain catchment, especially in the wetter southern parts (Alizai et al., 2011), implying that the Siwalik Group is not a major buffer on the zircon population. In any case, if the Lesser Himalaya had

been exposed earlier and had been supplying >1500 Ma zircon to the foreland basin then these materials should also have been seen in the fan at that time, which they are not.

The detrital zircon analysis in this paper would benefit from a more rigorous statistical treatment. I recommend something along the lines of DZ Mix (Sundell and Saylor, 2017). At the very least the authors need to ensure that their interpretations of the DZ data are plausibly consistent with published bedrock thermochronology from the area.

Response: We are reluctant to include the Sundell and Saylor analysis in this paper as that is the subject of another paper about zircons now in review elsewhere (G-Cubed) and is not essential for the objectives of this paper. The statistical treatment of the zircon data is considered in the MDS diagram which compares the detrital spectra with the bedrock data. This was part of the supplement but recognizing this importance we now add this to the main paper text to address this concern.

Regarding the detailed comments

Detailed Notes: Page 1 Line 13 – authors state that a decrease in "Nd values correlates with increasing abundance of >300 Ma zircon grains. Since no correlation coefficients are calculated I suggest replacing "correlates with" to "corresponds with" or "coincides with".

Response: We rephrase Page 1, line 13 as suggested.

Page 1 Line 14 – authors state that the increase in >300 Ma zircon grains precludes large-scale drainage capture as the cause of a long-term decrease in "Nd values. I realize that this is an abstract and space is limited but it is not clear why an increase in zircon grains whose age is consistent with Himalayan bedrocks warrants such strong language. Without further explanation it seems more correct to say that the increase in >300 Ma grains suggests increasing Himalayan contribution, or suggests that large scale drainage capture is not the cause of the "Nd decrease. Based on my evaluation of the reported zircon ages the increase in >300 Ma grains precludes drainage capture or reorganization of syntaxial or Karakorum draining rivers. However it could be explained by drainage capture or reorganization of rivers draining southern Tibet and the High Himalaya, specifically changes in the upper reaches of the Indus, Sutlej, or Chenab rivers where they flow orogen parallel across Tethyan rocks in southern Tibet.

Response: Our argument was badly phrased. It is not the increase in >300 Ma zircons that disproves drainage capture but rather the fact that there are large volumes of such zircons throughout the section, which in turn requires continuous sediment supply from the Himalaya, thus precluding any former drainage pattern that excluded much of the Himalaya-draining rivers. The abstract is edited to reflect this.

Page 1 Lines 16-19 – Authors use the increasing percentage of >1500 Ma grains in the post 1.9 Ma sediment samples to suggest a previously unrecognized episode of LHS exhumation in the western Himalaya. This is an exciting prospect. Maybe I'm missing something, but can't the shift in relative abundance of >1500 Ma grains also reflect decreasing exhumation rates in the Karakorum and Kohistan regions as well? Also, I think a discussion of lag times between

exhumation and deposition in the fans is need as well. Especially when we are discussing rivers with headwaters on the Tibetan plateau, these rivers flow through multiple sub-basins where long term (>100 ky) sediment storage has been predicted (Blothe and Korup, 2013). I realize that 100 ky is not long compared with million year timescales but it at least warrantsdiscussion.

Response: Yes, the change in the provenance we see is relative and could be caused by slowing of Karakoram erosion in the other sources, including the Greater Himalaya. However, the sediment budget of the Indus Fan implies increased total net erosion rates in the Pleistocene compared to the Pliocene making this scenario extremely unlikely. This point is now made in the revised manuscript.

Regarding lag times between exhumation and deposition we now add a note concerning the work of Blöthe and Korup, (2013) and provide details about recently published information concerning buffering in the upper Indus catchment from Jonell et al. (2017b). We also note that zircon travel times in the Indus River are estimated to be 7–14 ky since the Last Glacial Maximum (Clift and Giosan, 2014), thus precluding these processes from influencing the interpretation of the erosion record presented here.

Page 1 Line 25-27 – Authors state that foreland basin sediment provides an incomplete sedimentary record of orogenic unroofing since it is dominated by more local sediment sources. While this may be true of the most proximal coarse grained deposits more distal fine grained foreland basin deposits should reflect a more complete picture of the fluvial sediment load. Maybe it a failure of imagination on my part, but I don't see why distal foreland basins deposits should differ greatly from the submarine fan deposits analyzed here. The only additional sediment source the fan is sampling is the foreland basin deposit itself.

Response: The sedimentary section at Jawalamukhi analyzed by Najman et al. (2009) lies in the east of the Indus Basin and would represent sediments deposited in the foreland in front of the range in this area that were then accreted into the thrust stack. In the present day this means sediment from the Beas and Sutlej Rivers, and possibly the Yamuna in the past when this used to flow west into the Indus (Clift et al., 2012). The geometry of the foreland basin means that the Himalayan tributaries must have flowed SW in the geologic past too (Burbank et al., 1996). The trunk Indus River is essentially pinned on the western edge of the foreland, as also shown by studies of the Siwalik Group in that area (Chirouze et al., 2015). The Indus mainstream has never flowed eastwards along the foreland basin to join the Ganges as far as anyone is aware. Thus, the section at Jawalamukhi and similar locations would never preserve sediment derived from the trunk Indus and so only be able to reconstruct the flux in the Beas/Sutlej \pm Yamuna. The Siwalik section is not equivalent to the fan at all.

Page 1 Line 28-32 – The Authors argue that basin deposits are the only complete record of the long term exhumation of mountain ranges, which is true. However the events with which they are most concerned happened in recent geologic time (Miocene and younger) these tectonic changes in the thrust wedge are well documented in studies of bedrock exhumation. Any claim of rapid Quaternary exhumation of the LHS should be evident in bedrock studies as well, but to my knowledge no such study has found evidence for this.

Response: We agree that our sedimentary record of Lesser Himalaya erosion needs to be compatible with the bedrock thermochronology. We emphasize that we are looking at erosion, which is the final stage of exhumation, not cooling. Most bedrock thermochronology does a poor job of constrain final unroofing. Lesser Himalayan thermochronology in the Indus catchment suggests rapid cooling at 6–10 Ma based on high temperature U-Pb monazite dating (Caddick et al., 2007), but this still predicts that the Inner Lesser Himalaya now at the surface were at ~550°C, 4.5 kbar depth (~15 km) at 6 Ma. Apatite fission track dating of the Lesser Himalaya in the Sutlej Valley shows much younger ages of 1–2 Ma (Vannay et al., 2004), implying rapid uplift of 3–5 km since that time. However, these methods can only constrain the cooling of the rocks now exposed. The thermochronology of the bedrock alone does not inform how much Lesser Himalaya erosion has taken place in the past, or what used to cover the presently exposed units. The bedrock thermochronology does not preclude final unroofing after 1.9 Ma and indeed structural reconstructions are consistent with late stage unroofing in the Indus catchment (Webb, 2013). Our erosion data needs to be consistent with the thermochronology but likewise cooling models need to be consistent with the erosion record and if there was little Lesser Himalayan erosion before 1.9 Ma then its likely that those sources were not yet exposed.

Page 2 Lines 10-15 – The authors acknowledge that zircon U-Pb dating is only possible when sediment is sufficiently coarse grained. In their samples this means they could only analyze post 8.3 Ma sediment using this technique. This is problematic. They would like to use detrital zircon (U-Pb) dating to discern changes in sediment source, but are restricted to a very short window of time over which to do so. For example, prior studies have shown that a major foreland propagation event occurred around 10 Ma when the MBT became the locus of active thrusting across the orogen. This event led to widespread exhumation of the LHS. If the authors want to argue for a Quaternary period of rapid LHS exhumation it would be nice to see what the well known _10 Ma event looks like in their data for comparison. As it stands now we are left to wonder how the 1.9 Ma increase in LHS age consistent zircon grains compares with the well documented earlier event.

Response: The reviewer is concerned about the short duration of the zircon erosion record (largely <8.3 Ma) compared to thrusting and cooling of the Lesser Himalaya after 10 Ma. Clearly we can't say anything about erosion at or before 10 Ma without sediment of that age but the data we present are not incompatible with this model. Exhumation of a given unit does not require its unroofing and exposure, as explained above. The erosion record does not preclude the cooling and thrusting summarized here. Erosion of bedrock into sediment is almost the only way to track that final stage in the exhumation history. The rocks eroded in the pulse we see after 1.9 Ma probably did start to exhume after 10 Ma. The data we present doesn't constrain when the source started to exhume, so we can't say anything about that process.

Page 2 Lines 25-35 – This section is titled "Provenance Methods" but these lines are a discussion of prior studies more appropriate in a background section than a methods section.

Response: As suggested we change the title of the "Provenance Methods" section to "Background and Prior Studies", but these lines are a discussion of prior studies more appropriate in a background section than a methods section.

Page 5 Line 5 – Authors state that the "Nd values increase from 17 Ma to 9.5 Ma. This trend is barely discernable in their plot. The long term running average hovers between -9 and -10 over this interval.

Response: The reviewer was concerned that we overemphasize the drift to positive ε_{Nd} values from 17 to 9.5 Ma. We now change the text to note that the change is modest. Nonetheless, the total change is greater than the ± 1 uncertainty, so we consider the slight drift to be real.

Page 5 Lines 10-30 – There is a great deal of discussion of the potential effects of paleo-rivers on the submarine fan compositions. It would be helpful to see a figure showing these rivers. This entire discussion could benefit greatly from information on longshore currents in the region. Without this the reader is left to wonder what effect longshore drift, or storm events may have had on submarine sediment distribution.

Response: The reviewer wants to see a figure showing the rivers discussed in this section, but their modern courses are shown in Figure 1, which is called out in this section too. We further add a comment concerning longshore currents in the region as requested.

Page 5 Lines 38-42 – The authors state that young zircon grains (< 25 Ma) are restricted to the Nanga Parbat massif in the Indus catchment and then go on to acknowledge that zircon of 1850 Ma and 400-500 Ma age are known to be common in the Massif as well. This has been recognized as a LHS contribution to the granites of Nanga Parbat. This seems to be an alternative explanation for their proposed young LHS exhumation event. Especially since Nanga Parbat has been recognized as one the most rapidly exhuming place on Earth.

Response: The reviewer was concerned about the Lesser Himalayan contribution from the granites of Nanga Parbat. As detailed above, and in the manuscript, we discuss this possibility and suggest that Nanga Parbat is responsible for 35% of the provenance change but cannot be the only cause of the change in bulk isotope composition. The Indus downstream of Nanga Parbat is very poor in grains of these ages and does not explain the trends seen in the fan.

Page 6 Line 30-35 – The authors state that the increase in LHS age consistent zircons can only be achieved by preferential erosion of the LHS. However such an increase could also be achieved by rapid exhumation of the most proximal foreland basin deposits. The strongest increase the authors report is at 1.9 Ma, right around the time when the Main Frontal Thrust began to rapidly deform and uplift the foreland basin. The authors state previously that foreland basin deposits are dominated by proximal sources: : : in this case the LHS is the most proximal bedrock unit. The rise in LHS age consistent grains may actually reflect the onset of MFT deformation in the region.

Response: The reviewer was concerned about rapid exhumation of the most proximal foreland basin deposits rather than preferential erosion of the LHS to explain the provenance change in the fan. The arguments against this are provided above, namely that the estimated flux is small in volume (10-15% of the total) and that there is no evidence that the Siwaliks in the NW Himalaya have many of the right aged zircons, as all existing data are from Nepal.

Page 7 Line 26-30 – The authors note that despite a well documented exhumation event in the K2 region of the Karakorum they do not see a signal of this enhanced erosion in their samples. This is a common theme of this manuscript. The same is true of the 5 Ma to recent rapid erosion of the Nanga Parbat massif which does not show up in their samples (at least not in their interpretation). These examples should be viewed as red flags that the dataset presented here does capture the complete exhumation history of the region. Whether it is due to sediment residence times in upstream sub-basins, some sample bias introduced by hydrologic sorting, or some other unknown cause it seems clear that the dataset cannot be interpreted as a complete record of western Himalaya exhumation.

Response: The reviewer is concerned at the lack of evidence for an exhumation event in the K2 region of the Karakorum, as well as at Nanga Parbat, and thinks that the fan dataset cannot be interpreted as a complete record of western Himalaya exhumation, although as the final depocenter its hard to imagine what would be if not there. We argue that the erosional record in the submarine fan is providing an averaged record of western Himalayan exhumation because it derives sediment from across the entire catchment. No matter how dramatic a tectonic event might be locally if it does not result in a large erosional flux then this will not be noticed in the fan record. As far as K2 is concerned, it is far from clear how much of the Karakoram this represents because it is largely governed by the Karakoram Fault, which does not influence most of these ranges. Moreover, around half the sediment from K2 is transported north into the Tarim Basin. The lack of impact from K2 and Nanga Parbat is simply a reflection of their small size relative to all the other sources.

Page 8 Line 30-33 – Authors argue that their data shows the iLHS was not significantly exposed at the surface until 3-4 Ma. However, published zircon (U-Th/He) data cited in the manuscript shows that the iLHS was being rapidly exhumed by 11 Ma (Colleps et al. 2018), and that it was exposed the surface and contributing grains to the foreland basin by 9 Ma (Najman et al. 2009). The authors simply state the exposure of the LHS happened later that recognized in previous studies but do nothing to reconcile their dataset with those prior publications.

Response: The reviewer is again concerned about reconciling bedrock thermochronology with the erosional data, especially the zircon (U-Th/He) data of Colleps et al. (2018). As explained above, and in the manuscript, these data describe exhumation but not unroofing and these data are not inconsistent. The rocks studied by Colleps et al. (2018) do not lie in the Indus drainage, but even if they were representative of equivalent rocks in that catchment, rapid cooling after 11 Ma in the Lesser Himalayan rocks now exposed does not mean that the same unit was exposed at that time. Cooling precedes final exposure.

Response to anonymous referee #2

1. The role of Indian monsoon evolution on the sediments sources need to be better integrated. The authors argue that the summer monsoon does not reach the Karakoram (Karim and Veizer, 2002), therefore, changes in the erosion of those mountains is unlikely to be linked to changes in the summer rains. However, monsoon precipitation could impact the LHCS, and therefore, influence the erosion and sediment transport to the basin. Shubham Tripathi et al., (2017,

scientific reports) could be a good reference to discuss the evolution of monsoon since 18 Ma which is also from IODP 355 results.

Response: We agree completely with the reviewer that monsoon rains impact the Lesser Himalaya, including the Lesser Himalaya Crystallines, and indeed all the ranges on the southern side of the Himalaya rain shadow. The monsoon does affect erosion north of the Greater Himalaya too (Jonell et al., 2017a), but the volumes of sediment derived are somewhat less. Water supplied to the Karakoram today is more dominated by the Westerlies rather than the monsoon because of the high topography blocking the SW Monsoon. We note that over the last glacial cycle, erosion patterns within the Himalaya are strongly controlled by monsoon intensity, mostly act to cyclically strengthen and weaken the erosion of the Lesser Himalaya. Comparison of the changing erosion patterns with the foreland basin carbon isotope record was intended to show the possible linkages between erosion and climate. Carbon isotopes reflect the balance between C3 to C4 plants, which in turn is dependent on the aridity of the Indus basin. In contrast, the study of nitrogen isotopes published by Tripathi et al. (2017) does not make a suitable comparison to the erosion because it tracks marine productivity offshore western India and not rainfall in the Himalaya. The fact that this record is at odds with other paleoceanographic monsoon records in the Arabian Sea (Betzler et al., 2016; Gupta et al., 2015) also suggests a need for caution in its application. Nonetheless, we now note the correlation between increased Lesser Himalaya erosion and the change in the nitrogen isotope record after 3 Ma in the revised manuscript.

2.More information from Karakoram need to be present. What is its e Nd range (this will not be only a value, to be serious, we need to use a range)? What is the history of its precipitation and sediment transportation method? This could be a difficult question but with the eNd curve in fig 7, one could say Karakoram contribute a lot sediment to Laxmi Basin if calculated with a sample two-end member mixing. However, if monsoon could not influence Karakoram, how could the sediment be denuded and transported to the sea? with glacier melt water or West wind precipitation?

Response: As far as constraining the influence of the Karakoram is concerned, a range of Nd and Sr isotope compositions are considered and are plotted as a field in Figure 4. While a single value for the Karakoram isotopes could be misleading, we note that the average ϵ Nd measured in Karakoram bedrocks is -9.3 compared to -14.7 for the Greater Himalaya. Although the Karakoram bedrocks vary greatly in ϵ Nd values, from -1.5 to -22.8, measurements from rivers draining wide areas of the mountains also imply relatively positive ϵ Nd values for the erosional flux, e.g., ϵ Nd = -9.0 for the Karakoram in Nubra Valley, India, and -11.3 for the Braldu River in Pakistan (Clift et al., 2002). However, the influence of the Karakoram to the erosion flux is mostly clearly defined by the zircon U-Pb ages. A simple two-part Nd isotope mixing model of the type suggested may be applicable before the strong influence of the Lesser Himalaya, but also does not account for the influence of Kohistan and other parts of the Transhimalaya. Using the average ϵ Nd values of -14.7 for the Himalaya and -8 for the combined Karakoram and Kohistan, yields a range from only 15.4% Karakoram contribution at 1.13 Ma up to 94.8% at 6.74 Ma. Unfortunately, there is no separate climate record for the Karakoram extending over these tectonic timescales. Erosion in the modern Karakoram is driven by glaciers, with generated

sediments then being remobilized in rivers fed by snow melt delivering material largely to the Indus, but also north to the Tarim Basin.

3. Any further implications for earth climate evolution from tectonic activity on the Asia? Your e'Nd curve looks very similar to global deep-sea oxygen curve (James Zachos et al., 2001 Science) since 17 Ma. Recently, Francis A. Macdonald et al., (2019 Science) found a strong correlation between the extent of glaciation and arc-continent collisions in the tropics through a comparison of latitudinal distribution of ice-sheets and paleogeographic position of major Phanerozoic arc-continent collisions. Do you think the tectonic activities, which control the changes of sediment sources in Arabian Sea, could be related to set the earth climate or not ? why ?

Response: Whether erosion in the Indus basin has played a role in controlling global climate is not clear from the data presented in this study, which only tracks the original sources, not the volumes or chemical weathering state of the sediment. Falling ɛNd values through time imply less erosion of primitive magmatic rocks of the type favored by Macdonald et al. (2019) and would not be supportive of their enhanced weathering playing a role in driving cooling over the same period. A change in source would not otherwise imply more or less weathering flux. Regional sediment budgets from earlier work show falling sedimentation rates from the Late Miocene to the Pliocene at a time when chemical weathering intensity was also declining (Clift et al., 2008), which argues against surface processes in the Indus Basin playing a role in forcing global climate.

4. Is the changes of sediment sources be related to swing of turbidity channel, (therefore the deep sea fan) ? Laxmi basin is located in the eastern Arabian Sea and could only receive part of the sediment from the channel system now. But from recently study of Yu Zhaojie et al., 2019 QSR of the same cruise, it seems that the channel system could vary significantly through glacial-interglacial cycles due to sea-level variations. Hence, is that possible the swing of channel impact your sediment sources ? Therefore, it would very helpful to see a high resolution sedimentation rate in the Arabian Sea, particularly in the Laxmi basin. Did you also consider the contribution of eolian supply as many colleagues highlights eolian is important in this area?

Response: Avulsion of the active depositional lobes of the Indus Fan does not explain the changing sediment provenance at the drill site. Sedimentation rates at the drill sites in the Laxmi Basin are controlled by the location of the active lobe and are reported to be just 10 cm/k.y. in the Late Miocene but increasing to 45 cm/k.y. in the Early Pleistocene at IODP Site U1456 (Pandey et al., 2016a). When the active lobe is not directly over the sites then this would allow sediment flux from other non-Indus sources to be seen and not diluted. This may be the explanation for the modest amount of sediment with high ϵ Nd possibly derived from peninsular India. The zircon U-Pb data, however, are conclusive in requiring all sands and silt analyzed to be Indus derived as evidenced by the abundance of zircon grains dated <200 Ma. Eolian sediment is not considered to be important, even in those parts of the section where sedimentation was slow. When sedimentation rates were 10 cm/k.y. then using a dry density of ca. 1.6 g/cm³, equates to a dry mass accumulation rate (MAR) 160 g/cm²/k.y. total sediment. In contrast, the nearest sediment traps from the Arabian Sea showed MARs of 1.05–0.29 g/cm²/k.y.

and with a clear reduction away from Arabia (Honjo et al., 1999). It thus seems unlikely that wind-blown material would have contributed much of the sediment in Laxmi Basin.

Detailed comments:

Page 2 line 7: You probably need more information in the age-model

Response. It's not clear what extra information the reviewer would like to see in the age model beyond the nannofossil, foraminifers and magnetostratigraphy employed in the existing scheme (Pandey et al., 2016a; Pandey et al., 2016b). Although the age model continues to be refined the overall structure will not change greatly, especially with regard to long-term trends of the type we focus on in this study.

Page 2 line 17: I think HCL is better as acetic acid could not destroy all thecarbonate.

Response. Use of HCl rather than acetic acid is not favored for the decarbonation of the sediment prior to Nd and Sr isotopic analysis because it has been demonstrated that strong HCl can affect the Nd isotope composition of the analysis (Hein et al., 2017). Because Nd was the more critical isotope for provenance work we chose to use acetic acid for this work, which has a good track record of removing the carbonate.

Page 2 line 39: How many samples are used to do all those analysis ?

Response. Regarding grain size analysis, we measured grain size for the 11 coarser sediments for which zircon U-Pb data were obtained, not for the Nd-Sr isotope samples. This is now clarified.

Page 5 line 11: Is that possible those Sr-Nd isotopic end-members change through time ?

Response. The isotopic compositions of the end members that we compare to our sediments have to be assumed to remain constant through time. This is likely to be mostly correct for the bedrock sources in the mountains and in any case because the older rocks have now been eroded there is no way to know if any given source was the same in the past as it is now. As far as the Tapti, Narmada and Mahi Rivers are concerned, these have likely changed as erosion has changed the geology exposed in each catchment. Reconstructing this would require a sediment section from close to the mouths of each river to look at their changing discharge. Most likely ongoing erosion has stripped away Deccan Flood Basalts from over the older Precambrian peninsula rocks. The Tapti is still dominated by basaltic sources, while the Mahi may have been most affected. The effect would have been to have made each peninsula river more ENd negative through time, i.e., more similar to the Indus River. Sediment supply from these rivers would thus have been easier to see in the past then it is now. However, the major element data from the sands and silts do not favor significant sediment supply from the peninsula at any time because they overlap with compositions from the Quaternary delta and canyon of the Indus on the CN-A-K triangular diagram but are dissimilar to sediments from the modern continental shelf of peninsular India.

Page 5 line 1-11: It is better to integrate the Sr-Nd results from Yu Zhaojie et al., 2019 QSR even they present in a short time scale.

Response. As requested, we add the data from the recent Yu et al. (2019) study to the temporal evolution diagram together with some suitable accompanying text.

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Regional Pliocene Exhumation of the Lesser Himalaya in the Indus Drainage

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Abstract. New bulk sediment Sr and Nd isotope data, coupled with U-Pb dating of detrital zircon grains from sediment cored by International Ocean Discovery Program in the Arabian Sea, allow reconstruction of erosion in the Indus catchment since ~17 Ma. Increasing ε_{Nd} values from 17 to 9.5 Ma imply relatively more erosion from the Karakoram/Kohistan, likely linked to slip on the Karakoram Fault and compression in the Southern and Eastern Karakoram. After a period of relative stability from 9.5 to 5.7 Ma there is a long-term decrease in ε_{Nd} values that <u>corresponds</u> with increasing relative abundance of >300 Ma zircon grains that are most common in Himalayan bedrocks<u></u>. The continuous presence of abundant Himalayan zircons

- 15 precludes large-scale drainage capture as the cause of decreasing ENd values in the submarine fan. Although the initial increase in Lesser Himalaya-derived 1500–2300 Ma zircons after 8.3 Ma is consistent with earlier records from the foreland basin the much greater rise after 1.9 Ma, has not previously been recognized and suggests that widespread unroofing of the Crystalline Lesser Himalaya and to a lesser extent Nanga Parbat did not occur until after 1.9 Ma. Because regional erosion increased in the Pleistocene compared to the Pliocene the relative increase in erosion from the Lesser Himalaya does not reflect slowing
- 20 <u>erosion in the Karakoram and Greater Himalaya.</u> No simple links can be made between erosion and the development of the South Asian Monsoon, implying a largely tectonic control to Lesser Himalayan unroofing.

1 Introduction

The Western Himalaya region represents a classic example of an orogen formed by the collision of two continental plates. Sediment eroded from across this area records the development and unroofing of the mountains and has been accumulating in
the Arabian Sea since the start of the collision, likely in the Eocene (DeCelles et al., 2014; Najman et al., 2010). Because erosion removes rocks from the mountains the older history of the ranges is best reconstructed from the sedimentary record. Although some of this record is preserved onshore in the foreland basin (Najman, 2006), these sequences represent a relatively poorly dated and incomplete archive, with the erosion history of any particular section of the foreland basin being dominated by the immediately adjacent ranges rather than providing a complete regional record (Burbank et al., 1996). This makes it
difficult to use the foreland sequences <u>alone</u> to address the ongoing debate regarding the competing roles of climatically modulated surface processes compared to solid Earth tectonic forces in controlling the structural evolution of the Himalaya (Beaumont et al., 2001; Robinson et al., 2006; Webb et al., 2011). Without a detailed erosional history, it is impossible to fully reconstruct how the mountain belt has evolved over long periods of geologic time and what role erosion has played in focusing exhumation and controlling the location of major structures (Beaumont et al., 2001; Wobus et al., 2003).

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2 Marine Erosion Records

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Scientific drilling conducted in 2015 by International Ocean Discovery Program (IODP) Expedition 355 now provides the opportunity to examine how Himalayan erosion has changed since ~<u>10.8</u> Ma (Pandey et al., 2015). Although drilling in the Laxmi Basin offshore western India was able to reach the Cretaceous basement at Site U1457 (Fig. 1) the recovered submarine fan sequences are mostly limited to the past 10.8 Ma as a result of a large mass transport complex (MTC), which removed much of the older record (Calvès et al., 2015). At Site U1456 a single sample of siliciclastic sand dated ~15.5 Ma was recovered

- below the carbonate breccias of the MTC, but otherwise the sediments span the same 10.8 Ma seen at Site U1457. Sediments from within the MTC are not included in this study. Nonetheless, the new core provides a much better dated and more complete basin-wide history of erosion than has been previously available for the western Himalaya covering the period since the Middle
- 10 Miocene. Here we present new provenance data from the IODP drill sites, as well as additional data from industrial borehole Indus Marine A1 drilled by Wintershall Holding (Germany), located near the river mouth (Fig. 1), in order to reconstruct the erosion of the area in detail since ~17 Ma, the age of the oldest material from Indus Marine A1.

3. Background and Prior Studies

- Age control at Sites U1456 and U1457 is provided by a combination of biostratigraphy and magnetic stratigraphy (Pandev et al., 2016b), while at Indus Marine A1 age control is accomplished through biostratigraphy alone (Shuaib, 1982). We used bulk decarbonated sediment Nd and Sr isotope compositions coupled with single grain detrital U-Pb zircon ages in order to constrain how the source of sediment changed over long periods of time. Zircon U-Pb dating is only possible when the siliciclastic sediment is sufficiently coarse grained, restricting application of this method to the last ~8.3 Ma. Nd isotopes have a history as robust provenance indicators of siliciclastic sediment and are especially effective in the Western Himalaya where there is a
- 20 wide range of isotopic values known from the different bedrock source terranes (Clift et al., 2002). This isotope system is particularly effective <u>because</u> it is not believed to be affected by sediment transport or chemical weathering processes (Goldstein et al., 1984), although it is moderately influenced by grainsize in the Indus catchment (Jonell et al., 2018). Although it has been noted that HCl leaching during carbonate removal prior to analysis can affect the measured ¹⁴³Nd/¹⁴⁴Nd values (Hein et al., 2017), in this study we treated all analyses with acetic acid so that no systematic bias was introduced. Sr isotopes
- 25 are also employed because they provide an additional dimension for resolving sources, although the sediment has to be carefully decarbonated prior to analysis. Sr isotopic values are also influenced by chemical weathering and grain size variations (Derry and France-Lanord, 1996; Jonell et al., 2018). Earlier work from the Arabian Sea, including samples from Indus Marine A1, used only Nd isotopes to demonstrate a substantial change in source since ~6 Ma, a trend that was attributed to large-scale drainage capture of the eastern Himalaya-draining tributaries of the Indus away from the Ganges basin towards the East (Clift and Blusztajn, 2005; Zhuang et al., 2015).
- More recently this Late Miocene-Recent Nd isotopic trend was attributed in part to the onset of uplift of the Nanga Parbat Massif within the Western Syntaxis (Chirouze et al., 2015) <u>since</u> this source yields extremely radiogenic Nd (Whittington et al., 1999). Accelerated erosion elsewhere in the Himalaya was also inferred to have a played a part in causing the evolution in Nd isotopes, <u>Comparison</u> of cosmogenic and high temperature thermochronometers (e.g., Ar/Ar muscovite) <u>implies</u> that <u>faster</u>
- 35 erosion has occurred during the last few million years in the western Himalaya (Vance et al., 2003). We supplement bulk sediment Nd analyses with U-Pb dating of single zircon sand grains that are also recognized as effective provenance proxies within the western Himalaya (Alizai et al., 2011; Gehrels et al., 2011) but which has never before been used at the basin-wide scale over million year times scales due to lack of appropriate sections. Zircon grains are physically robust during transport and may be recycled many times from their original sources, but the U-Pb age is not easily reset because of the high temperatures required, ~950°C (Gehrels, 2014). Sources rarely have single distinctive ages, but rather a characteristic spectrum

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of zircons ages and these are transferred to the sediments during erosion. Comparing sediment and bedrock age spectra allow the source of the sediment to be constrained.

4 Analytical Techniques

5 4.1 Grain size Analysis

Samples were prepared using standard procedures, with material being taken from each of the 11 samples that were the subject of zircon U-Pb dating. We put a small amount of sample into a cleaned 50 ml plastic centrifuge tube and added 5–7 ml of sodium phosphate solution. The tube was capped and vortexed to defloculate clay-sized sediment and separate organic particles. The sample was poured through an 850 µm sieve and funneled into a 15 ml glass test tube. After centrifuging and

- 10 removing the clear supernatant, 2–3 ml of sodium phosphate and 5 ml of 30% H₂O₂ were added. Tubes were vortexed again and then put into a hot bath that was heated to 70°C. This step requires persistent monitoring to prevent loss of reactant by spraying it with acetone until the reaction is stabilized. Reactants then sat overnight to completely oxidize organic matter. Reacted supernatant was removed and 5 ml of sodium phosphate was added. These treated samples were then rinsed with deionized water, transferred into clean 50 ml plastic centrifuge tubes, and topped with sodium phosphate into a sample solution
- 15 of up to 40 ml. Samples were vortexed again prior to grain size analysis. Grain size analysis was conducted on a Beckmann Coulter LS13 320 laser diffraction particle size analyser at Louisiana State University (LSU). The obscuration of all running samples in the aqueous liquid module (ALM) was between 8–12 %. Results are provided in Supplement Table 1.

4.2 Major Element Analyses

- 20 Bulk_y samples of the sands targeted for zircon U-Pb dating were analyzed for their major element contents by Inductively Coupled Plasma Emission Spectrometry (ICP-ES) at Boston University, USA. Sediment samples were decarbonated with acetic acid, washed with distilled and deionized water with a purity of 9–12 megaohms, and hand powdered before total fusion preparation. Glass beads for each sample were made in a muffle furnace under 1050°C by fusing 100 ± 0.5 mg of sample mixed with 400 ± 0.5 mg lithium metaborate (LiBO₂). The melted mixture was then dissolved in 5% HNO₃, sonicated,
- 25 manually shaken until no visible grains were observed, and further diluted for analysis (Dunlea et al., 2015). Precision for all elements was better than 1% of the measured value, and accuracy was confirmed by repeated analyses of International Standard Reference Materials (Basalt, Hawaiian Volcano Observatory, BHVO-2)(Wilson, 1997). Results of the geochemical measurements are shown in Supplement Table 2.

30 4.3 Isotope analysis

Nd and Sr isotopes were measured from powdered whole sediment samples. After decarbonation with 10% acetic acid and dissolution, Sr and Nd were concentrated using standard column extraction techniques, and isotopic compositions were determined by Thermo "Neptune" multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS) at Woods Hole Oceanographic Institution. Sr results were corrected against NBS 987 standard ⁸⁷Sr/⁸⁶Sr=0.710240 and Nd data were

35 corrected against JNdi-1 standard ¹⁴³Nd/¹⁴⁴Nd = 0.512104. We calculate the parameter ENd (DePaolo and Wasserburg, 1976) using a depleted-mantle model age and ¹⁴³Nd/¹⁴⁴Nd value of 0.512630 for the Chondritic Uniform Reservoir (CHUR (Bouvier et al., 2008)). Results of the geochemical measurements are shown in Supplement Table 3.

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4.4 Zircon U-Pb dating

After standard mineral separation <u>zircon grains</u> were sprinkle-mounted onto double-sided tape on 1" acrylic discs and <u>analyzed</u> at random using depth-profiling LA-ICP-MS U-Pb geochronology. For each sample at least 120 zircons were <u>analyzed</u> to obtain provenance datasets that resolve any <u>component comprising</u> >5% of the total population (Vermeesch, 2004). The
analyses were completed using a PhotonMachine Analyte G.2 Excimer laser (30 µm laser spot size) with a large-volume Helex sample cell and a Thermo Element2 ICP-MS at the UTChron facilities at the Jackson School of Geosciences at the University of Texas at Austin using procedures described in Hart et al. (2016). GJ1 was used as the primary reference standard (Jackson et al., 2004) and a secondary in-house zircon standard (Pak1 with a TIMS ²⁰⁶Pb/²³⁹U age of 43.0 Ma). The data from the analyses were then reduced using the Iolite data reduction software and VizualAge (Paton et al., 2011; Petrus and Kamber, 2012). For <u>analyzed</u> detrital zircons, the ²⁰⁶Pb/²³⁸U age was used for grains younger than 850 Ma and the ²⁰⁷Pb/²⁰⁶Pb age was

used for grains older than 850 Ma<u>(Gehrels et al., 2008)</u>. All ages reported use 2σ absolute propagated uncertainties, ²⁰⁷Pb/²⁰⁶Pb ages are less than 30% discordant, and ²⁰⁶Pb/²³⁸U ages are less than 10% discordant (Gehrels et al., 2011). The discordance reported is calculated with the ²⁰⁶Pb/²³⁸U and ²⁰⁷Pb/²³⁵U ages if <850 M.y. and the ²⁰⁶Pb/²³⁸U and ²⁰⁷Pb/²⁰⁶Pb ages if >850 Ma. Results of the geochemical measurements are shown in Supplement Table 4.

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5 Results

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Sediment grain size can be assessed using the classification scheme of Folk (1974) (Fig. 2A). Sediments are dominantly silty sand and sandy silt, with one sample defined as a silt (U1456A-11H-6W). If we consider the range of grain sizes in any given sample (Fig. 2B) we see a generally good sorting (positive kurtosis) and a positive skew, i.e., a dominance of the finer grain sizes and a tail of coarser grains comprising a diminishing proportion of the sediment. Very little material of coarse sand size

is seen in any of the material.

The general geochemical character of the sediments can be seen on CN-A-K ternary diagram (Fedo et al., 1995)(Fig. 3A). The data plot in an array close to a Chemical Index of Alteration (CIA) of ~70 (Nesbitt et al., 1980). They form a roughly linear array trending towards the illite end member and suggestive of its progressive involvement as the primary <u>mineral product of</u>

- 25 breakdown. Here we compare the Laxmi Basin samples with sediments from the Indus delta (Clift et al., 2010) and Indus Canyon (Li et al., 2018), as well as sediments from the western Indian shelf and slope between Saurashtra peninsula and Bhatikal (Kurian et al., 2013)(Fig 1). The samples from the Indian shelf largely lie offshore from extensive exposures of the Deccan Trap, flood basalt sequences. We note that the Laxmi Basin sands have very similar bulk compositions compared to the Quaternary Indus Canyon and delta but plot below the array of the Indian shelf sediments. Only the sample taken near
- 30 Bhatikal plots lower than the Laxmi Basin sediments. This plot confirms that the analyzed sands have little in common with material eroded from peninsular India and <u>is</u> consistent with an Indus River origin. Likewise, the <u>Laxmi Basin</u> sediments plot <u>overlapping</u> with the Quaternary Indus sediments on the discrimination diagram of Herron (1988)(Fig. 3B). These sediments form an array defined as wackes and litharenites, while the western Indian shelf sediments fall into the Fe shale and Fe sand fields.
- 35 Nd and Sr isotope compositions of silty and muddy sediments from the boreholes are shown in Figure 4 and are compared with source regions in the Greater Himalaya, Karakoram sand Transhimalaya, as well as the Deccan Plateau of neighboring Peninsular India. The Lesser Himalayan Series have very low ε_{Nd} and high ⁸⁷Sr/⁸⁶Sr values and lie outside this plotted area, but still contribute material. The Lesser Himalayan influence on the fan sediment is not resolvable from the Greater or Tethyan Himalaya using the Nd and Sr isotopic systems. Many of the analyses overlap the range of bedrocks in the Karakoram, allowing
- 40 for these ranges also to be an important source. However, on the basis of these isotopic data alone the relative influence of basement sources cannot be quantified. <u>Interestingly, several IODP samples <10.8 Ma old, as well as those from the high</u>

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resolution study by Yu et al. (2019) spanning the past 0.6 Ma, often have higher ε_{Nd} values than samples from Indus Marine A1 or sedimentary rocks of the Kirthar and Sulaiman Ranges of southern Pakistan (Fig. 5). Both of these latter sequences are generally considered to be ancient deposits of the lower reaches of the Indus River and <u>are</u> largely free from other influences (Zhuang et al., 2015). The IODP samples also have higher ε_{Nd} values and lower 87 Sr/ 86 Sr values than many Quaternary sediments from the Indus Delta (Clift et al., 2010)(Fig. 4).

U-Pb zircon ages in all samples span a wide range but all show a significant detrital age component <120 Ma, comparable to bedrock ages from the Karakoram and Kohistan<u>, as shown in the kernel density estimate diagram of Vermeesch (2012)</u> (Fig. 6). In general, the abundance of grains with U-Pb ages of 750–1250 and 1500–2300 Ma increases up section. The 300–750 Ma component is present throughout, mostly at 20–30% of the total, peaking briefly at 39% at 5.9 Ma and falling from 30%

at 3.4 Ma to 18% in the present day. The 1500–2300 Ma detrital age component shows an initial increase from 5 to 17% after
 8.3 Ma and shows another jump from 20 to 35% of the total load after 1.9 Ma (Fig. 6).

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6 Discussion

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6.1 Shifting Erosion Patterns

- 15 The long-term temporal evolution in Nd isotope character can be used to reconstruct the erosion of source rocks and exhumation patterns. Long-term trends in Nd isotopic composition are highlighted by a five-point running average that defines a trend to slightly increasing ε_{Nd} values from 17 Ma to ~9.5 Ma, followed by a period of approximate stability until 5.7 Ma and declining values after that time. Phases of steep decline were identified, most notably after 5.7 Ma and after 3 Ma (Fig. 5). The tendency of some IODP samples to have higher ε_{Nd} values compared to previously analyzed Indus-sourced sediment could
- 20 reflect a non-Indus contribution to the IODP sites from sediment eroded from the Deccan Plateau and delivered by the Tapti River (Fig. 1). <u>Rivers draining the peninsula and located further south than the Tapti River are not considered to be likely suppliers to the area of the drilling sites because of the southward directed longshore currents (Shetye et al., 1994), as well as the contrasting bulk sand samples geochemistry character (Fig. 3). Alternatively, this difference might reflect relatively greater erosional flux from the Transhimalaya or Karakoram during some of the time since 10.8 Ma in a way not seen in earlier</u>
- 25 analyses (Zhuang et al., 2015). Significant flux from a paleo-Mahi River (Fig. 1) can be ruled out due to its low ε_{Nd} and high ⁸⁷Sr/⁸⁶Sr values, <u>although this river could have had higher ε_{Nd} values in the past when much of this catchment was covered by</u> Deccan volcanic rocks that have now been removed by erosion. Because no independent record of Mahi River discharge exists this possibility remains speculative. The Narmada River is isotopically indistinguishable from the post-15 ka Indus (Fig. 4). Indus Marine A-1, located close to the river mouth, and the Kirthar/Sulaiman ranges that have preserved the lower reaches of
- 30 the Indus River these may be considered to represent a relative pure Indus signature (Zhuang et al., 2015). This allows for a better assessment of whether the more distal sediments in the Laxmi Basin were truly derived from this river or may be influenced by significant flux from peninsular India. Differences between the IODP sample compositions and the isotope record from closer to the Indus River mouth could reflect a number of processes. For example short-term changes in erosion patterns related to climate change and reproducibility of Nd isotope composition, generally considered to be ± 1 ENd (Jonell et al., 2015).
- 35 al., 2018), could also result in significant variability. We suggest that when analyses from the Laxmi Basin depart by >1 ε_{Nd} from the trend of the Indus Marine A-1, Sulaiman/Kirthar or western Indus fan Ocean Drilling Program (ODP) samples then they likely do not represent only flux from the Indus River. This is especially true when ε_{Nd} values increase, suggesting mixing with sediment from the neighboring isotopically positive Deccan Plateau, consistent with the interpretation of Yu et al (2019) since 0.6 Ma at IODP Site U1457. Significant variations in Indus River ε_{Nd} values have been observed over recent glacial

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Sediments that have ε_{Nd} values more positive than those sediments deposited closer to the Indus River mouth require mixing with additional sources outside anything known from within the Indus catchment. Such analyses are considered to be at least

- 5 partly derived from the Indian peninsula (Fig. 5). Sediments with such positive ENd values are only found in modern rivers in the Karakoram, not near the delta (Clift et al., 2002), and would require supply from a river with essentially no drainage of Himalayan source regions, which seems unlikely. Mixing with the sediment from the peninsula is more likely. Indeed, sedimentary petrography has identified peninsula-derived material in sands in the uppermost parts of the IODP section (Pandey et al., 2016b) but the potential for erosion from these areas lower in the section remains. Eolian sediment derived from Arabia
- 10 is not considered to be important, even in those parts of the section where sedimentation was slow. When sedimentation rates were ca. 10 cm/k.y., as they are in the muddier parts of the stratigraphy, then using a dry density of ca. 1.6 g/cm³, equates to a dry mass accumulation rate (MAR) 160 g/cm²/k.y. total sediment (Pandey et al., 2016a). In contrast, the nearest sediment traps from the Arabian Sea showed lithogenic MARs of only 1.05–0.29 g/cm²/k.y. and with a clear reduction away from Arabia (Honjo et al., 1999). It thus seems unlikely that wind-blown material would have contributed much of the sediment in Laxmi
- 15 Basin, even when MARs were low.

Based on our understanding of the primary sources of the Indus River the simplest explanation for the long-term increasing trend in ε_{Nd} values between 17 and 9.5 Ma would be a relative increase in erosion from either the Karakoram, Kohistan or Transhimalaya compared to Himalayan sources, followed by a reversal since 7.0 Ma. A simple two-part mixing model using ε_{Nd} = -14.7 as a representative value for the Greater Himalaya, based on the sources plotted in Figure 4 and ε_{Nd} = -8.0 for a
 combined Karakoram/Kohistan source based on bedrock analyses and the range of Indus Fan sediment compositions, yields a

range of contributions from only 15.4% Karakoram at 1.13 Ma up to 96.3% at 94.8% at 6.74 Ma. This estimate is based on equal Nd contents of the sources, consistent with modern river sediment data (Alizai et al., 2011). The detrital U-Pb zircon ages also show temporal evolution, although these data are limited to the past 8.3 Ma with the exception of a single sample of 119 grains dated at ~15.5 Ma, recovered from below the mass transport complex (Fig. 6). Because all of the sands contain
grains <120 Ma this requires derivation from the Indus River and not peninsular India (Fig. 6).

The implications of the zircon U-Pb dating can be better understood by sub-dividing the ages into provenance diagnostic groups. Zircon grains <25 Ma are found in igneous rocks at the core of the Nanga Parbat Massif (Zeitler et al., 1993). Such young grains are unknown elsewhere in the Indus catchment, although older grains dated at ~1850 and 400–500 Ma are also known to be common in the Nanga Parbat Massif (Zeitler et al., 1989), reflecting the correlation of the massif with the Lesser

- 30 Himalaya based on Nd isotopes (Whittington et al., 1999). Zircons dating 40–70 Ma are associated with both Kohistan and the Karakoram, while 70–120 Ma grains are largely found only in the Karakoram (Searle, 1996). 300–750 and 750–1250 Ma grains are found in both the Greater and Tethyan Himalaya (Alizai et al., 2011; DeCelles et al., 2000; Gehrels et al., 2011), while 1500–2300 Ma zircons are most frequent in Lesser Himalayan rocks (DeCelles et al., 2000). Zircon grains dating 1500– 2300 Ma are also found in other Himalayan units and comprise 38% of the Greater and Tethyan Himalayan analyses compiled,
- 35 however 99% of Lesser Himalayan grains fall in that age range. This observation indicates that Lesser Himalayan units are likely most critical in controlling the supply of 1500–2300 Ma grains, especially when 750–1250 Ma grains do not increase at the same time.

The major element analyses of the sands suggest that they are similar to Quaternary Indus Canyon and delta sediments and unlike western Indian shelf sediments (Fig. 3). Peninsular India is not affected by Himalayan mountain building and

40 magmatism and fission track data shows progressive_ modest cooling of western India since the Cretaceous that would not affect the U-Pb ages presented here (Gunnell et al., 2003). The youngest zircon U-Pb grain ages that can reasonably be expected from an Indian Peninsular river is ~65 Ma, derived from the Deccan Plateau (Schoene et al., 2015), but no such peak is noted in the data from the Laxmi Basin sands (Fig. 6). This is consistent with the sands not being derived from the Indian Peninsula. **Deleted:** and would require supply from a river with essentially no drainage of Himalayan source regions, which seems unlikely. Mixing with the sediment from the peninsula is preferred. Indeed, sedimentary petrography has identified peninsula-derived material in sands in the uppermost parts of the IODP section (Pandey et al., 2016) but the potential for erosion from these areas lower in section remains.

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The changing abundance in the Lesser Himalayan associated 1500–2300 Ma zircon group does not precisely mirror changes in the Nd isotope character. For example, this group leaps from 19.6% to 34.6% of the total population between 1.9 and 1.5 Ma, while at the same time ε_{Nd} values fell only slightly after a large fall between 3.4 and 2.5 Ma. Nonetheless, it is this group in particular that shows a clear relative increase in abundance over the long term since 7.0 Ma and which is also associated

- 5 with bedrock sources with very negative ε_{Nd} values. Zircons of this age comprise just 5% at 8.3 Ma, but 17% by 5.9 Ma, even before the large increase to >34% after 1.9 Ma. In contrast, the 300–750 Ma group falls slightly in abundance after 8.3 Ma. The 750–1250 Ma group shows an increase from 20% at 5.9 Ma to 32% by 3.0 Ma. This suggests that it is a source rich in 1500–2300 Ma grains, and to a lesser extent 750–1250 Ma, that is changing most in its contribution to the fan since 8.3 Ma, especially after 1.9 Ma.
- 10 Provenance evolution can be further assessed by comparing the spectra of the detrital zircons with bedrock data using a multidimensional scalar diagram (Vermeesch et al., 2016). This is a form of principle component analysis that statistically compares the U-Pb age spectra. Samples that are similar to one another plot close together on the diagram. Figure 7 shows that Himalaya sources plot on the left side of the diagram and Karakoram and Kohistan sources on the right. Nanga Parbat appears to plot in the upper center but this is based only on bedrock data from the center of the massif, not including the older zircons that
- 15 probably occur at the periphery of the massif. Not surprisingly the modern trunk Indus River plots close to the Karakoram, as do the older samples from the Laxmi Basin. There is a clear long-term trend in the detrital samples towards more and more Himalayan compositions, consistent with the analysis of the KDE plots in Figure 6. This figure indicates that the Greater and Tethyan Himalaya are likely the most important sources to the youngest samples (0.9 and 1.6 Ma), which are similar to the modern river mouth.
- 20 Sediment storage and recycling are unlikely to be affecting the erosional signal. Although storage in basins between source and sink does influence the Himalaya, this is largely of millennial duration and rarely >100 ky (Blöthe and Korup, 2013), a value still too small to affect the reconstruction presented here. Recent work on intermontane basins in the Indus catchment estimates sediment buffering on timescales of 10³-10⁴ yr (Jonell et al., 2017), while the erosional response to deglacial climate change allowed zircon travel times in the Indus to be constrained as 7–14 ky (Clift and Giosan, 2014).

25 6.2 Unroofing the Lesser Himalaya

Nanga Parbat-related 0–25 Ma grains are present throughout the sedimentary section in Laxmi Basin but are never very abundant, although they do increase from 1.8 to 5.1% of the total between 3.4 and 3.0 Ma (Fig. 3). Their modest overall contribution limits the role of Pliocene unroofing of Nanga Parbat in driving the Nd isotope evolution (Chirouze et al., 2015). However, the presence of 300–750 and 750–1250 Ma grains in all samples demonstrates the long-term flux from the Greater

- 30 and Tethyan Himalaya to the Indus Fan and allows us to rule out large-scale drainage capture of eastern tributaries as the cause of falling ε_{Nd} values since 5.7 Ma (Clift and Blusztajn, 2005). We note that ε_{Nd} values started to fall after 5.7 Ma, close to the time when the first Inner Lesser Himalayan units, characterized by their very negative ε_{Nd} values (< -22), were first exposed (Najman et al., 2009) but also at a time when the 750–1250 Ma zircon grains became more abundant. Although 1500–2300 Ma grains, characteristic of the Lesser Himalaya, started to become more abundant after 8.3 Ma, the sharpest increase in their</p>
- population was after 1.9 Ma. The shift towards the Lesser Himalaya is shown by the fact that at 7.8 Ma the 750–1250 Ma (Greater/Tethyan Himalaya) and 1500–2300 Ma (Lesser Himalaya) age populations accounted for 26% and 17% of the total zircons respectively, while by 0.9 Ma the proportions had changed to 32% and 38%. Such a change can only be achieved by increasing Lesser Himalayan erosion even more than erosion in the Greater/Tethyan Himalaya, which also rose. Sedimentation rates in the submarine fan increased from the Late Miocene into the Pleistocene (Clift, 2006) so this change in relative influence
 cannot indicate reduced erosional flux from the Greater/Tethyan Himalaya.

Furthermore, the abundance of zircons in the source rocks that contribute to the Indus mainstream is relatively low compared to the eastern Himalaya-draining tributaries. Zr can be used as a proxy for zircon content because of its high concentration in

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this mineral (Amidon et al., 2005). The Indus mainstream sample from Attock records a Zr concentration of ~18 ppm compared to a range of 14 to 63 ppm for the Himalayan tributaries (Alizai et al., 2011). Using water discharge data as a proxy for sediment transport capacity we estimate an average Zr concentration for the modern Punjabi tributaries of 40 ppm, more than double that in the mainstream (Alizai et al., 2011). Zr is even higher in the Laxmi Basin sediment (142–223 ppm) reflecting hydrodynamic sorting during transport.

This discrepancy in zircon source abundance is important because although the youngest Laxmi Basin sediment contains 79% of grains older than 300 Ma this does not mean that 79% of the erosion is from the Himalaya, rather than the Karakoram, Kohistan and the Transhimalaya. Indeed, petrographic data (Garzanti et al., 2005) and Nd isotope constraints (Clift et al., 2001) indicate that only ~39% and 41% respectively of the total flux to the Arabian Sea is now from Himalayan sources, with

- 10 the Karakoram accounting for much of the rest. The low zircon fertility along the mainstream upstream of Attock means that it is unlikely that uplift of Nanga Parbat could be driving the changes in zircon reported in this study despite the fact that there are Lesser Himalayan rocks within the massif. It is noteworthy that the same is not true of Nd concentrations, because the Indus mainstream contains 27 ppm Nd at Attock, compared to a range of 20–29 ppm in the Himalayan tributaries (Alizai et al., 2011). Because the two areas have similar Nd concentrations the evolution in Nd isotope composition <u>can</u> be interpreted
- 15 to indicate more erosion from both the Lesser Himalaya and Nanga Parbat since 5.7 Ma. The modern river data indicates that ~10% of the total modern zircon flux in the trunk river upstream of Attock is from Himalayan bedrocks (>300 Ma), i.e., not from the eastern tributaries. The >300 Ma zircons account for just 16% of the zircon flux from the upper mainstream. In contrast, the mainstream supplies 61% of the total sediment flux to the Arabian Sea based on Nd isotope constraints (Alizai et al., 2011). 96% of the zircon flux from the eastern Punjabi tributaries is >300 Ma but these streams account for only 39% of
- 20 the total Indus sediment load reaching the Arabian Sea (Alizai et al., 2011). If we estimate that the Punjabi tributaries are 2.2 times more enriched in zircon, but around the same in Nd, compared to the mainstream Indus River then this implies that the evolution to more ε_{Nd} negative values seen offshore is 35% driven by erosion from Nanga Parbat and 65% driven by erosion in the Lesser Himalaya.
- The lack of <u>a tight</u> correlation between Nd and U-Pb zircon data reflects the fact that these different analyses were not both
 performed on the same samples, although they both show the same long-term drift to more erosion of older Himalayan crust and less from the Karakoram/Kohistan. Short lived changes in erosion pattern would account for mismatches in provenance between samples with similar but different depositional ages. We also recognize that Nd and Zr are not uniformly concentrated in the sources along the mainstream and Punjabi tributaries, and that the fine-grained sediments analyzed for Nd may have different provenance than the sandy zircon material, e.g., coarser material may be preferentially derived from the Karakoram,
 while finer material is more Himalayan (Jonell et al., 2018), or derived from peninsular India.

6.3 Controls on Erosion

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<u>The</u> importance of Karakoram/Kohistan to the erosional flux in zircon grains is clear <u>until after 7.0 Ma</u> and may be related to motion on the Karakoram Fault that started after 16 Ma (Phillips et al., 2013) and that drove rock uplift and unroofing along the length of that structure. <u>The</u> reduction in erosion is despite thermochronology data from the K2 mountain region that emphasize Pliocene cooling of the central Karakoram after slower exhumation earlier in the Cenozoic (Foster et al., 1994).

- 35 emphasize Pliocene cooling of the central Karakoram after slower exhumation earlier in the Cenozoic (Foster et al., 1994). The lack of any detectable signature in the fan related to material from K2 likely reflects its small area compared to the other potential sources to the Indus Fan and the fact that much of the sediment is transported north into the Tarim Basin (Clift et al., 2017). The fan record provides a basin-wide average image of erosion but is not good at identifying dramatic tectonic events in limited areas that do not produce much sediment, just as also noted in the Nanga Parbat Massif. Away from the Karakoram
- 40 Fault itself the gneiss domes of the southern Karakoram show rapid uplift and cooling after 20–25 Ma (Rolland et al., 2001), potentially contributing to the trend to more positive ε_{Nd} values from 17 to 9.5 Ma. However, exhumation rates in the southern Karakoram continued to be rapid into the Pleistocene (Mahéo et al., 2004), while the sediment data require them to be outpaced

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by Himalayan erosion since 7.0 Ma. However, erosion in the eastern Karakoram has a markedly different history, with faster rates of exhumation in the Middle and Late Miocene followed by a slowing after 5 Ma (Wallis et al., 2016), consistent with the zircon and Nd isotope evolution presented here. Because the summer monsoon does not reach the Karakoram (Karim and Veizer, 2002) changes in the erosion of those mountains is unlikely to be linked to changes in the summer rains, but could be

- 5 related to the strength and location of the Westerly Jet which supplies the glaciers that control erosion in that region. Neither Nd isotopes nor zircon U-Pb ages exhibit large changes in provenance between 15.5 and 8.3 Ma, implying that the Tethyan and maybe the Greater Himalaya were already unroofed by ~15 Ma and continued to erode through that time interval. It is noteworthy that the initiation of greater relative contributions from the Lesser Himalaya after 7.0 Ma as tracked by zircon populations and after 5.9 Ma based on Nd isotopes commenced at a time of reducing monsoon rainfall across the foreland
- 10 basin as tracked by the relative abundance of C3 versus C4 vegetation (Dettman et al., 2001; Quade et al., 1989; Singh et al., 2011)(Fig. §). The Lesser Himalayan-related 1500–2300 Ma grains increased from 5% to 17% of the total zircon population between 8.3 and 7.8 Ma, while the 750–1250 Ma group only rose from 23 to 26% and the 300–750 Ma group fell from 26 to 20%. The trend in erosion at this time is strongly towards the Lesser Himalaya. Subsequently, the 750–1250 Ma group shows a steadier rise from 5.9 Ma to 3.0 Ma, while the 300–750 Ma group is more erratic and peaked at 5.9 Ma. The start of a
- 15 progressive increase in zircon flux from the Himalaya after 8.3 Ma occurred during a time of overall slowing erosion and drying climate, as inferred from the regional sediment flux budget to the fan (Clift, 2006)(Fig. 7). Falling intensities of chemical weathering and slowing total sediment delivery rates from the Late Miocene to the Pliocene (Clift et al., 2008) mean that the total weathering flux must be decreasing. Consequently, if Himalayan uplift has caused more silicate weathering, drawing down CO₂ and cooling the global environment (Raymo and Ruddiman, 1992) this is because of processes in the Eastern 20 Himalaya, and not those in the Indus catchment.
- Lesser Himalayan erosion must be considered in two phases. The Outer Lesser Himalaya were likely eroding in NW India since 16 Ma (Colleps et al., 2018). The structurally overlying Inner Lesser Himalayan units lie directly under the Greater Himalaya Sequences and contrast in Nd isotope character (ENd <-22) with the Outer Lesser Himalaya that overlap with the Greater and Tethyan Himalaya in terms of Nd isotopes (ENd >-18; Fig. 4)(Najman et al., 2009). Nd isotope work on foreland basin sedimentary rocks has indicated that the unmetamorphosed Inner Lesser Himalaya were first exposed after 9 Ma, at least
- Locally in the Beas River area (Najman et al., 2009) and in Pakistan (Meigs et al., 1995)(Fig. 1), earlier than the fall in ε_{Nd} values in the new marine data after 5.9 Ma. These foreland Nd data further indicate that the very ε_{Nd} negative rocks of the metamorphosed Inner Lesser Himalaya were exposed after 6 Ma (Fig. 2), which correlates more closely with the offshore record of a decrease in ε_{Nd} values at that time. However, the Beas area foreland records do not record the steep rise in Inner
- 30 Lesser Himalayan erosion after 3.4 Ma observed in the Indus Fan (Najman et al., 2009). This implies that although the metamorphosed Inner Lesser Himalaya were exposed locally in the Beas area earlier in the Miocene, as shown by the change in isotope character in the fluvial sediments in that part of the foreland basin, their widespread unroofing across the entire Indus catchment is younger than previously recognized. This prediction is however consistent with recent palinspastic structural reconstructions of the western Himalaya (Webb, 2013). Thermochronology from the bedrocks of the Lesser
- 35 Himalaya in the Indus catchment indicate that exhumation was rapid from 10 to 6 Ma, but that the rocks now exposed at the surface were still deeply buried (12–15 km) at that time (Caddick et al., 2007). Lower temperature thermochronometers including zircon and apatite fission track constraints from the Lesser Himalaya in the Sutlej Valley point to continued rapid cooling after 6 Ma (Vannay et al., 2004) but we can only constrain the exhumation of the rocks now exposed at the surface. They cannot inform us of when these units were first unroofed. This is the role of the erosional record we present here.
- 40 It is not apparent how the evolution in exhumation since the Late Miocene might be linked to climatic evolution. More vigorous Lesser Himalayan erosion parallels a weakening monsoon and slower overall erosion (Fig. §). When the monsoon is strong the occurrence of extreme summer rainfall increases (Turner and Annamalai, 2012) and when this occurs strong rainfall penetrates further north into Himalayan valleys and drives erosion of those rock units (Bookhagen et al., 2005). Consequently,

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Late Miocene monsoon weakening would be expected to involve a southward retreat of strong rainfall from the edge of the Tibetan Plateau, resulting in more rainfall and erosion over the area where the Lesser Himalayan rocks are exposed. Inner Lesser Himalayan thrust sheets were imbricated above a ramp in the basal decollement (Webb, 2013), This stacking

- resulted in surface uplift and generated a break in the topography against which the summer monsoon rains began to precipitate,
 focusing erosion and bringing these rocks to the surface. A positive feedback is likely because modelling has shown that focused denudation can encourage the formation of duplexes in thrust belts (Malavieille, 2010). Exposure of the Inner Lesser Himalaya on a regional scale happened later than has been previously recognized (Meigs et al., 1995; Najman et al., 2009). Zircon (U-Th)/He thermochronology argues for a start of cooling in the Inner Lesser Himalaya after 11 Ma (Colleps et al., 2018) but this does not require widespread unroofing of this unit until the later times indicated by this study, i.e., 3.4–1.9 Ma.
- 10 The final regional exposure does not correlate either with changes in the <u>vegetation in the foreland basin</u> and only roughly with the onset of Northern Hemisphere Glaciation that is often believed to enhance erosion at 2–4 Ma (Zhang et al., 2001). Although it does correlate with a strengthening of the monsoon derived from nitrogen records from the same drill sites (Tripathi et al., 2017) it is unclear if this marine biogenic production record is closely related to Himalayan rainfall or not. Even if the Lesser Himalayan unroofing is not linked with monsoon intensity this does not preclude the older Greater Himalaya (Fig. 1C)
- 15 from being more intimately linked to summer monsoon development. However, the evidence of this study suggests that the monsoon has been reacting more passively to structural changes in the Himalaya driven by solid Earth tectonic processes since the Late Miocene and possibly since ~15 Ma.

6 Author Contributions

Peter Clift helped organize the drilling expedition, designed the analytical program and led the writing. Peng Zhou did much of the sample preparation work. Daniel Stockli was responsible for the dating of the zircon grains. Jerzy Blusztajn performed the Nd and Sr isotope analysis. All authors contributed to the writing and interpretation.

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Figure Captions

Figure 1: A) Shaded bathymetric and topographic map of the Arabian Sea area showing the location of the drilling sites within the Laxmi Basin, considered by this study. Map also shows the primary source terranes and the major tributary systems of the

- 5 Indus River, as well as smaller peninsular India rivers that may have provided material to the drill sites. B) Inset map shows detail of the Laxmi Basin and location of the drill sites considered in this study. Numbered red circles indicate existing scientific boreholes from Deep Sea Drilling Project (DSDP) and Ocean Drilling Program (ODP). Magnetic anomalies are from Miles et al. (1993). White dashed lines show transform faults. KK = Karakoram; NP = Nanga Parbat. C) Geological map of the western Himalaya showing the major tectonic units that are eroded by the Indus River and its tributaries. Map is modified
- after Garzanti et al. (2005). Rivers as shown in thick black lines. ISZ = Indus Suture Zone, MCT = Main Central Thrust, MBT
 = Main Boundary Thrust and MFT = Main Frontal Thrust. Thick black line shows the boundary of the Indus drainage, while thinner lines demark the limits of the major Himalayan tributaries.

Figure 2: A) Grainsize range of all samples analyzed for U-Pb zircon dating from the Laxmi Basin shown on the scheme ofFolk (1974). B) The detailed grainsize spectra of the analyzed samples.

Figure 3: (A) Geochemical signature of the analyzed samples illustrated by a CN-A-K ternary diagram (Fedo et al., 1995). CN denotes the mole weight of Na₂O and CaO* (CaO* represent the CaO associated with silicate, excluding all the carbonate). A and K indicate the content of Al₂O₃ and K₂O respectively. Samples closer to A are rich in kaolinite, chlorite and/or gibbsite

20 (representing by Kao, Chl and Gib). CIA values are also calculated and shown on the left side, with its values correlating with the CN-A-K. Samples from the delta have the lowest values of CIA and indicates high contents of CaO and Na₂O and plagioclase. Abbreviations: sm (smectite), pl (plagioclase), ksp (K-feldspar), il (illite), m (muscovite). B) Geochemical classification sediments from the Indus delta (Clift et al., 2010), Indus Canyon (Li et al., 2018) and western India peninsular rivers (Kurian et al., 2013) following the scheme of Herron (1988).

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Figure 4: Diagram showing the Nd and Sr isotopic compositions of sediments recovered from the Laxmi basin drill sites and Indus Marine A-1 compared to the major possible source terrains. Inset map shows the full isotopic range of the source ranges. Data from the Deccan Plateau compiled from Georoc (http://georoc.mpch-mainz.gwdg.de/georoc/). Pink squares show the composition of selected Peninsula India rivers (Goswami et al., 2012). Compositions for the Indus Delta since 15 ka are from

- 30 Clift et al. (2010). Data from the Kirthar and Sulaiman ranges are from Zhuang et al. (2015). Transhimalaya data are from Rolland et al. (2002), Singh et al. (2002), and Khan et al. (1997). Greater Himalayan data are from Ahmad et al. (2000), Deniel et al. (1987), Inger et al. (1993) and Parrish and Hodges (1996). Karakoram data are from Crawford and Searle (1992) and Schärer et al. (1990).
- Figure 5: Evolution of Indus related Nd isotope composition since 17 Ma. (A) New data from IODP Sites U1456 and U1457 and Indus Marine A-1 together with existing data from the latter, from <u>JODP Site U1457 measured at high resolution by Yu e tal. (2019), from ODP sites in the western Arabian Sea and from the Kirthar and Sulaiman Ranges.</u> The yellow-shaded area shows the ±1 ε_{Nd} uncertainty for those data not at Sites U1456 and U1457. Samples plotting to the right of this range could be influenced by flux from peninsular India. (B) Data from all the sources in the previous diagram with peninsular-related samples
 excluded and a five-point running average plotted with the. ±1 ε_{Nd} uncertainty. Note the decline in ε_{Nd} values after 6 Ma and especially after 3 Ma.

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Figure 6: Kernel density estimate (KDE) plots for the zircon U-Pb ages from Laxmi Basin compared with some of the major source terrains in the western Himalayas, from the compilation of Alizai et al. (2011). Colored strips depict characteristic age peaks that can be linked to specific source areas within the Western Himalayas. Left column shows KDE plots to 3000 Ma and the right-hand column shows the data from 200 Ma.

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Figure 7. Multidimensional scalar (MDS) plot comparing the detrital samples from the Laxmi Basin (green dots) with potential source regions onshore (red dots) and both the modern river mouth and mainstream Indus downstream of Nanga Parbat (blue dots). Data sources are as for Figure 6. Note progressive migration away from Karakoram/Kohistan end members and towards the Greater and Tethyan Himalaya.

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Figure 8: Comparison of climate, erosion and exhumation proxies in the Himalaya. (a) Smooth Nd isotope history for the Indus River from the Arabian Sea with grey background showing effective uncertainties from Figure 5. (b) Breakdown of detrial zircon U-Pb age populations with each source diagnostic group shown as its proportion of the total. Percentage is recalculated to exclude non-diagnostic aged grains. (c) Carbon isotope character of pedogenic carbonate in Pakistan as an

15 indicator of dominant vegetation in the Potwar Plateau of Pakistan (Quade et al., 1989), and NW India (Singh et al., 2011). (d) Relative exhumation rates of the Greater Himalaya tracked by bedrock Ar-Ar dating (Clift et al., 2008) and zircon fission track from foreland basin sediment (Chirouze et al., 2015). (e) Rates of sediment supply to the Arabian Sea calculated from regional seismic (Clift, 2006). KK = Karakoram, LH = Lesser Himalaya, LHCS = Lesser Himalayan Crystalline Series.

20 Supplement Table 1. Grainsize data from those samples from which zircons were dated by U-Pb methods.

Supplement Table 2. Major element analysis by ICP-ES of those samples from which zircons were dated by U-Pb methods.

Supplement Table 3. Sr and Nd isotope data from the samples considered in this study.

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Supplement Table 4. U-Pb zircon analytical data and calculated ages for samples considered in this study.

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Figure 4



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10 Figure 8