Responses to Comments on the Manuscript:

"A new method of resolving annual precipitation for the past millennia from Tibetan ice cores"

(MS No.: tc-2021-115)

We sincerely thank Prof. Jihong Cole-Dai for his thoughtful comments. Below we have made point-to-point responses to the comments. The comments are in black, and our responses in blue.

The paper's main objective is to reconstruct precipitation records from ice cores. The steps to accomplish the objective (Linea 80-88) are (1) determining thickness of annual layers in ice cores, (2) modeling layer thinning caused by ice flow, and (3) combining the results of (1) and (2) to reconstruct past precipitation. The authors call this process a "new method" to develop annual precipitation records from ice cores.

I am not sure what is new in the items presented in the paper. For Step 1, the way to identify annual layers in this work is the use of measurement of chemical impurities in ice cores that show annual oscillations in amount or concentration. But this is not new. Numerous papers have documented the identification of annual layers in ice cores using various chemical species measured at reasonable or acceptable temporal resolution and the use of the identified annual layers to derive layer thickness and the number of annual layers in a core (dating). Perhaps the authors mean that the chemical analysis technique (LA-ICP-MS) is new to ice cores. But a measurement technique (elemental measurement) for Ca different from the other techniques (e.g., ion chromatography) does not necessarily make the method to identify annual layers new. For example, when hydrogen peroxide was first measured in ice cores and its concentration was found to oscillate annually, H_2O_2 measurement for annual layer counting was not considered a new method of dating or annual layer determination (Sigg and Neftel, 1980 and Sigg et al., 1994). Maybe the authors believe the millimeter LA-ICP-MS analysis resolution is new; but measurements such as electric conductivity (ECM) and dielectric profiling (DEP), which have been used for quite a while, can also be of high-resolution (mm or sub-mm).

Response: Although ice core annual layer identification and layer thinning modeling are not new, they have not been used to reconstruct detailed (e.g. annual) precipitation time series beyond a few tens of years from Tibetan ice cores due to technical challenges and limitations. The new method presented in study has the potential to develop annual precipitation time series at millennial time scale from alpine ice cores, which has not been achieved so far from any previous studies.

We agree with Prof. Cole-Dai that numerous papers have documented the identification of annual layers in ice cores using various chemical species measured at reasonable or acceptable temporal resolution. However, due to rapid layer thinning, annual layer identification was only possible for the top sections (meters or tens meters) of the Tibetan ice cores from the conventional discrete measurements (e.g., water stable isotopes, major ions, dust) used in these studies, with a typical sampling resolution of centimeters. Although ECM and DEP can be of high-resolution (mm or sub-mm) and have been widely used for polar ice cores (Rasmussen et al., 2006), both techniques have not been used for the Tibetan ice cores to establish their chronology. This is largely because the high dust background of these ice cores causes a high noise/signal ratio that obscures the seasonality of their ECM and DEP profiles.

Our study is indeed the first attempt to use LA-ICP-MS on a Tibetan ice core. Given the ultra-high sampling resolution of LA-ICP-MS (153 μm per sample in our study), two orders of magnitude higher than the sampling resolution (centimeters) of the conventional chemical measurements, this technique allows us to identify annual cycles in deeper sections of the Chongce ice core (back to 2.5 ka B.P. in this study, in comparison to the usual tens years based on the conventional discrete measurements).

It is also worth pointing out that the ^{14}C measurements were first applied to establish the chronology of Tibetan Chongce ice cores (Hou et al., 2018, 2019 and 2021). This provides the necessary information of layer thinning to develop the new method. We wish that this method will be used to reconstruct a continuous high-resolution precipitation record at millennial time scale when LA-ICP-MS is performed on the entire Tibetan ice cores.

The paper title suggests that the authors think this is the first time ("new") that annual layer identification using high-resolution chemical measurement is applied to mountain glacier ice cores. But that point is not apparent or explicit in the paper body.

Response: It is indeed the first time that the ultra-high-resolution LA-ICP-MS measurement is applied to mountain glacier ice cores, making it possible to identify annual layers at deeper sections of these cores. It is also the first time that such annual layer identification is combined with layer thinning modeling to reconstruct annual precipitation records at the millennial time scale from the Tibetan ice cores. This new method has potential to be applied to other alpine ice cores. We have clarified these points in the revision.

For Step 2, the modeling of ice flow including the method to quantify layer thinning rate is obviously not new. So, reconstruction of original layer thickness using measured layer thickness and modeled thinning curve should not be considered new, if the layer identification method is not new. In fact, reconstruction of accumulation records from measured annual layer thickness and modeled and experimentally determined layer thinning has been used frequently for polar ice cores, with records as long as tens of thousands of years. Yet, the authors seem to suggest (Lines 59-60: "challenging to develop annually resolved accumulation records covering longer (e.g. millennial) time periods") that this is a rare accomplishment. Again, this would sound reasonable when referring to mountain glacier ice cores (but not so for polar ice cores).

Response: We agree with the reviewer that the novelty of the method only applies to mountain glacier ice cores. Annual layer identification can be achieved for the Greenland summit ice cores for the past tens of thousands of years (Rasmussen et al., 2006), but this is not the case regarding the mountain glacier ice cores due to their relatively short length and rapid thinning. Our study is indeed the first to apply this method, i.e. taking account of both annual layer identification at the millennial time scale and annual layer thinning, for the Tibetan ice cores. It has also potential to be applied to other alpine ice cores. We have clarified these points in the revision.

The authors state (Lines 187-198) that identification of annual layers using concentration cycles of the selected elements was "verified" with counting by the StratiCounter program. But the StratiCounter algorithm is not designed to count layers accurately. It is meant to facilitate the counting process and make counting less subjective. Therefore, counting with StratiCount ought not to be used to verify manual counting. In fact, the algorithm is supposed to be used only when the annual signals are clear and consistent (i.e., highly unambiguous; Sigl et al., 2015). In this case, with many annual signals highly ambiguous (multiple peaks in one annual layer; see Figure 1), result of Straticounting cannot be viewed as verification of the result of manual counting.

Response: We agree with the reviewer's concern on the StratiCounter program. Indeed, in this study, we relied on visual identification and manual counting as our primary method, using StratiCounter only for comparison purposes. In our case, the results from manual counting and StratiCounter are highly consistent, reinforcing the validity of our results. We revised the wording in the manuscript to clarify this.

The precision (i.e., uncertainty) of annual layer counting (ALC) is critically important to the success of layer thickness determination and ice core dating. Often times, researchers compare the number of annual layers counted to a certain depth where a time stratigraphic marker, such as a known volcanic eruption or the radioactivity signal of nuclear debris) is present to the number of expected years established with the time marker; however, this comparison only indicates the accuracy of counting, not precision. In practice, the counting is often revised by reclassifying ambiguous layers, if its result differs significantly from the expected number of years, to reconcile with the age of the layer where the time marker is, in order to improve accuracy. To address precision directly, researchers have tried several approaches to get a quantitative sense of ALC precision/uncertainty. For example, Alley et al. (1997) estimated ALC uncertainty based on the difference of the number of layers counted by different individuals and/or at different times by the same individual. Rasmussen et al. (2006) and Ferris et al. (2012) derived ALC uncertainty by summing up the number of ambiguous (possible and possibly not) annual layers.

This paper by Zhang et al. does not discuss the uncertainty in the identification of the annual layers using elemental data (concentrations ought to be proportional to counts per second in mass spectrometry). Usually, an annual cycle is defined as one maximum and one minimum of the measured concentration of the chemical species (in some instances a measured physical property may be used), at least this is the case for polar ice cores. For cores analyzed with high temporal resolution, cases of ambiguous annual layers are rare, leading to small uncertainties. Because the deposition processes of chemical impurities on non-polar mountain glaciers are probably different (Lines 188-189) from those common in the polar regions, the authors use a definition of annual signal different from that often used on polar ice cores. In this work, the authors define (Lines 187-188) annual layers as groups of peaks (of element concentration) "separated by a prolonged section of low element concentrations". One reason for this is that multiple peaks of a given element are found in each annual layer (Lines 188-190). This definition of an annual layer may work in some cases or cores, but raises questions on what an annual layer is, such as how many peaks constitute "a group of peaks" and how long a section is considered "prolonged". The data presented in Figure S7 look very obvious that two years of accumulation are in about 4 cm of the core. But I don't see many years in Figure 1 which are so obvious. Additionally, how does one decide where the annual layer starts where it ends, when multiple chemical species do not agree with each other? An example of ambiguous or inconsistent layers in different species can be seen in the depth interval of 108.09-108.15 m in Section II (Figure 1). With this type of data and the definition of annual layers, the determination of annual layers and thickness is quite subjective, at least more so than in polar ice cores where the definition is clear and annual signals are often unambiguous.

The authors acknowledge the limitation, resulting from this annual layer definition, on the precision of layer determination in Line 192: "identification of annual layers requires expert judgment". To me, this means that researchers with varying degree of experience or "expertise" or different perspectives will count differently. The results will be different, not only in the total number of annual layers counted (accuracy), but also in the thickness of individual layers. This is critically important to the objective of this work – accumulation history – as uncertainty in layer thickness leads directly to uncertainty in accumulation of individual years as well as long-term accumulation trends.

I would like to see some discussion, hopefully supported with data, on the uncertainty of the annual layer thickness determination in the core(s) studied in this work and using the counting method. Also, it would be helpful to explain what the expertise is to make "expert judgment", so as to provide a measure of the objectivity of layer counting.

Response: We agree with the reviewer that the accuracy and uncertainty of annual layer counting (ALC) is critically important to the success of layer thickness determination and ice core dating. ALC is always accompanied by a certain level of subjectivity. Such as the example given by the reviewer, Alley et al. (1997) estimated ALC uncertainty based on the difference of the number of layers counted by different individuals and/or at different times by the same individual. This could be considered "expert judgment". This is also why we included the results of annual layer identification from the StratiCounter algorithm to provide a relatively "objective" mathematic expression in comparison with our subjective visual identifications.

In the revision, we follow the approach successfully employed for Greenland ice cores by Rasmussen et al. (2006) to quantify counting uncertainty from uncertain layers. In this approach, we count uncertain layers as 0.5 ±

0.5 years, and estimate the maximum counting error (MCE) from the number of uncertain layers (N) as N × 0.5 years. Using the records for four elements (i.e., Al, Ca, Fe, Mg), we define "uncertain annual layer boundaries" as those without synchronous peaks of all four elements. Annual layer peaks for each of the three sections and their respective uncertainties are shown in Fig. 1. The number of annual layers for Section I, II, and III is 8 ± 2, 19 ± 3, and 23 ± 3. The derived average annual layer thickness for Section I, II, and III is thus $38.30 \pm ^{9.57}_{6.38}$ mm (corresponding to $30.96 \pm ^{7.74}_{5.16}$ mm w.e.), $18.42 \pm ^{3.45}_{2.51}$ mm ($14.74 \pm ^{2.76}_{2.01}$ mm w.e.), and $12.71 \pm ^{1.91}_{1.47}$ mm ($10.16 \pm ^{1.52}_{1.17}$ mm w.e.), respectively. We included these updated results in the revision.

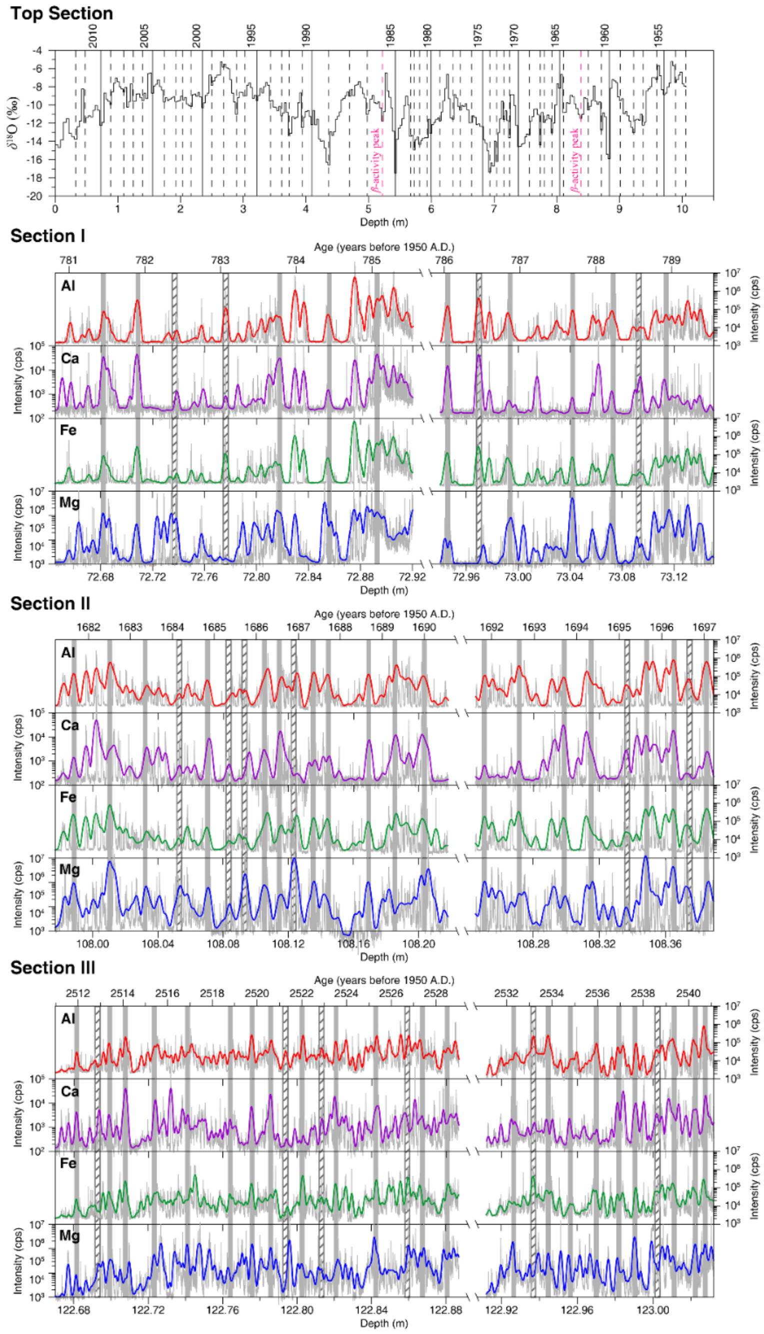

Figure 1. Annual layer counting for the Top Section, Section I, II, and III (top to bottom). The annual layers of the Top Section are identified based on the seasonality of $\delta^{18}O$ and two β-activity peaks (An et al., 2016). The annual layers of Section I, II, and III are marked at the winter/spring peaks (grey bars) of Al, Ca, Fe, and Mg concentrations. The open grey bars filled with forward slashes indicate uncertain annual layers. Thin grey lines indicate raw data, and thick colored lines represent 200-point Gaussian smoothing. LA-ICP-MS intensity is reported as counts per

second.

I am curious why the authors decided to present data and offer discussion on only three very short sections of Core 2 of approximately 135 meters. It appears that the chemical analysis and layer counting were done for most, if not all, of the core: the number of years counted extends to the deepest of the core (Figure 1).

Response: So far, we have only performed the LA-ICP-MS measurement on the three trial sections. We plan to perform more LA-ICP-MS measurements in the near future.

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