

Response to Anonymous Reviewer #2 to manuscript TC-2020-55

Title: Giant dust particles at Nevado Illimani: a proxy of summertime deep convection over the Bolivian Altiplano

Authors: Filipe G. L. Lindau, Jefferson C. Simões, Barbara Delmonte, Patrick Ginot, Giovanni Baccolo, Chiara I. Paleari, Elena Di Stefano, Elena Korotkikh, Douglas S. Introne, Valter Maggi, Eduardo Garzanti, Sergio Andò

We thank the Anonymous Referee #2 for his comments and suggestions. All line numbers correspond to the discussion paper and all added texts to the discussion paper are marked blue.

Specific Comment #1: L 70: The authors state that this is the first time giant particles have been addressed in Andean ice. Is this the first time they have been observed, or simply the first time they have been interpreted? If it's the first time they have been observed, why is that? Is there something special about the analysis here, or site, or time period, that would be unique? I cannot recall ever seeing such large particles presented and interpreted at any high elevation or polar ice core site, so some kind of brief literature review would be helpful. Have such particles ever been observed and explained at high elevation observation stations? Any evidence that would corroborate the existing and transport mechanisms would help orient the reader to what might be happening here.

[Answer]: *We changed the text in order to fill this gap of information:*

L 70: An interesting result concerns the presence of giant dust particles (presenting a diameter larger than 20 μm), whose variability is correlated with the stable isotope record. Very large mineral dust particles were generally neglected in climate studies and underrepresented or non-represented in global climate models, because of their generally local origin with respect to the sampling site, and their relatively low number concentration (Albani et al., 2014; Adebiyi and Kok, 2020). The recent observation of such large dust grains even at great distance from the source puts into question the physical models used to estimate settling velocities, and suggests some additional mechanisms such as strong turbulence and upper-level outflow are needed to keep these dust particles aloft (van der Does et al., 2018). As a consequence, there is now a growing interest in such relatively less abundant but volumetrically important dust grains, which can play an important role in biogeochemical cycles, in cloud microphysics, in the ocean carbon cycle and atmospheric radiation budget (van der Does et al., 2018; Ryder et al., 2019). A few studies have also considered large mineral particles in snow and ice, obtaining interesting results, in particular related to the relationships existing between coarse particles and the atmospheric patterns responsible for their deflation, transport and deposition (Kutuzov et al., 2016; Wu et al., 2009, 2010; Simonsen et al., 2019).

Our data show that the concentration of giant dust particles into firn is correlated with regional meteorological observations, and in particular with atmospheric deep convection over the Bolivian Altiplano during summer. This study shows for the first time that the presence of giant dust particles in Andean firn and ice is strictly

controlled by climatic processes. We found clear evidence that the convective activity over the Altiplano, reconstructed through the analysis of giant particles, is enhanced during summer periods and in particular during La Niña years, in agreement with observations concerning atmospheric circulation anomalies in the area (Vuille, 1999).

Specific Comment #2: L 150: The authors assert that any particles larger than 5 microns are of obvious local origin. I have a hard time understanding what is local and what is distant in this geographic context. If local means very close (i.e., within the glacier basin) to the ice core site, then how does a convective mechanism make sense? Local wind stress seems a much likelier scenario. A better location map showing dust sources and some kind of proposed transport pathway would be helpful. And because the focus of the paper is on the giant particles, why not collect mineralogy data on them? It seems logical to compare/contrast the fine and giant particles to establish some basis for local/remote origin.

[Answer]: We added a definition of local and regional sources in Sect. 1:

L 55: Previous ice core studies from the Central Andes (Correia et al., 2003; Knüsel et al., 2005; Osmont et al., 2019) reveal that the aerosol content of ice is dominated by local (i.e. glacier basins from Nevado Illimani) and regional (the Altiplano's area) mineral dust during the winter, when black carbon from biomass burning in the Amazon basin is also present.

[Answer]: Then, we added the potential dust sources, discussed in Sect. 3.2 of the discussion paper, to Fig. 1:



Figure 1: Location of the Nevado Illimani, the Zongo Valley and the Quelccaya ice cap. The numbers indicate the location of the meteorological stations used for comparison with our results: 1 – El Alto; 2 – Calacoto; 3 – Patacamaya; 4 – Oruro and 5 – Potosi. The red area indicates the Altiplano-Puna Volcanic Complex (Lindsay et al., 2001). The gray squares and gray triangles denote to potential dust source areas in the salars of the Altiplano and in the Puna, respectively (Gaiero et al., 2013). The land basemap was derived from satellite data (Natural Earth I with Shaded Relief from <http://www.naturalearthdata.com>).

[Answer]: Finally, we fixed the text in Sect. 2.4:

L 150: We used single-grain Raman spectroscopy to identify mineralogy of dust particles having a diameter smaller than 5 μm . This because this kind of analysis was carried out for provenance purposes, **thus considering particles expected to travel over longer distances.**

Specific Comment #3: L 200–215: The entire logic of the paper relies on the explanation of the stable isotope signal in terms of convective activity. Yet, the paragraph begins by stating the Andean isotope signal has divergent explanations. Has the convective explanation been explicitly tested at Nevado Illimani? And do the amount effect and convective explanations occur in unison, or are they mutually exclusive? Sentences on modeling and satellite observations are interesting, but it is not clear to me that this has been tested and verified at Illimani with in situ data. Merely stating that you assume convection is the dominant control (lines 213-214) is, to me, not sufficient in this case.

[Answer]: *The "amount effect" and the convective hypothesis occur in unison and therefore are not mutually exclusive. This is known from the literature, especially in the Illimani area. Indeed, climatic controls on the isotope signal in precipitation were deeply studied nearby Nevado Illimani, at the Zongo Valley (about 60 km northwest Nevado Illimani). Over Zongo, deep atmospheric convection was found to be an important control on δD during the rainy season (Vimeux et al., 2011), and represents the primary factor controlling δD , in addition to the cumulative rainfall. This gives further support to our conclusions. We have modified the text as follows:*

L 200–215: Seasonal variations of the water stable isotopes in snow precipitated over the Andes are also useful for dating. However, the Andean isotopic signal led to divergent interpretations (Vimeux et al., 2009). Whereas in polar ice cores the water isotopic signature is chiefly related to temperature (Uemura et al., 2012), the isotopic composition of tropical precipitation can be affected by a larger number of factors (Hoffmann et al., 2003). It is well known that the so called “amount effect” leads to an anti-correlation between the amount of precipitation and the proportion of heavier isotopes in the precipitation. This effect is in turn related to an ensemble of physical and microphysical processes producing a robust signal on the isotopic composition of precipitation (Dansgaard, 1964; Risi et al., 2008; Vuille et al., 2003). In this context, deep atmospheric convection also plays a role on stable isotope composition (Vimeux et al., 2005). **Along the Zongo Valley (Fig. 1, located nearby Nevado Illimani), in particular during the summer season, the cumulative rainfall along air mass trajectory is a second order parameter in the control of isotopic depletion, being it primarily modulated by regional convective activity (Vimeux et al., 2011). In agreement,** modeling studies (e.g. Bony et al., 2008; Risi et al., 2008) reveal that the stronger the convective activity during a particular event, the higher the total amount of precipitation and thus the more depleted the isotopic composition of precipitation. **In addition,** satellite data (Samuels-Crow et al., 2014) reveal that during the summer season the isotopic composition of water vapor strongly depends on convective activity. These observations lead the authors to conclude that the isotopic composition of snow from the tropical Andes mainly reflects tropical convection.

Specific Comment #4: L 216: I would like to see a better statistical treatment of the data throughout the paper. The authors note a “close correspondence” of stable isotopes and giant particles in Fig. 2a. To my eye, the r value must be at least -0.8, which is remarkable in ice core data that normally have a fairly low signal/noise ratio. I don’t understand what the PCA value adds.

[Answer]: *We removed the PCA from the text. Then, we changed the Fig.3 that was presented in the Response to Anonymous Reviewer #1:*

L 216: Interestingly, we note a close correspondence between the variability of stable isotopes and the proportion of giant particles in firn (Fig. 3): oscillations of the stable isotope record (δD) closely follow the percentage of giant dust particles (GPPnb). During the dry season, giant particles are proportionally less abundant (average GPPnb 0.5%) whereas the isotopic composition of snow is less negative (average -113‰ for δD ; -15‰ for $\delta^{18}O$). Conversely, during the wet season when giant dust particles are at their annual maximum (average GPPnb 1%) the isotopic composition of snow is more depleted (-141‰ for δD , -18‰ for $\delta^{18}O$), reaching its minimum.

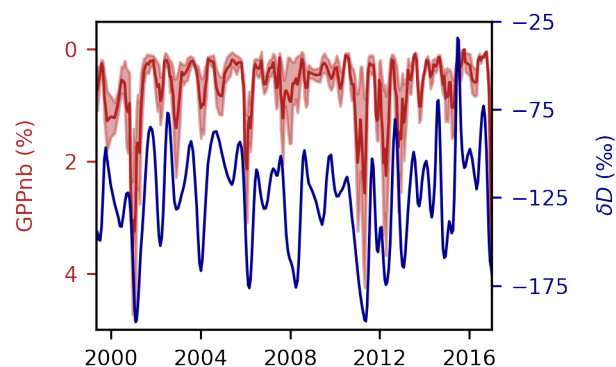


Figure 3: Relationship over the 18-years record between the percentage of giant particles with respect to the total dust particles number (GPPnb, reverse scale) and the δD . Uncertainties for each GPPnb value (expressed by the red shaded area) are relative to the standard deviation between Coulter Counter measurements. All data are reported as 3-point running average of the data previously re-sampled at 0.05 m w.eq.

[Answer]: Although ice core data normally have a fairly low signal/noise ratio, we consider that our correlation was weaker than -0.8 due to giant particles measurement uncertainties. These uncertainties were explained in the Response to Anonymous Reviewer #1, leading to the addition of the following sentences in Sect 2.2:

Section 2.2: The higher deviation for the mass in comparison to the total number of particles was expected due to the presence of heavy giant particles having diameters $>20 \mu m$ (coarse silt), for which small differences in size estimation lead to higher uncertainties. Indeed, when considering only the giant particles the mean RSDs were 55% and 63% for the number and mass distributions, respectively. Thus, the proportion (%) of giant particles (GPPnb) as well as total particle concentration, were calculated from the number size distribution. Approximately 14% of the samples showed very large uncertainties (RSD $>100\%$) for GPPnb and were discarded. The mean RSD for GPPnb was 45%.

Specific Comment #5: L 223–225: I don't understand the relationship among GPP, Ca, and total dust concentration. The correlation between total dust, Ca, and GPP appears poor (statistics would help confirm this), particularly in the dry season. The dry season values are quite variable in dust and Ca, but quite consistent in GPP. What is responsible, and how does it bear on local vs. regional sources?

[Answer]: This part of the original discussion paper was changed in the Response to Anonymous Reviewer #1, including a new Fig. 2 (as showed below). Fig. 2 shows that giant and total particles have a similar variability during the dry season. Ca^{2+} indeed features some differences. We believe these differences seem justified considering our high temporal resolution. We expect that the composition of dust in the core is not always the same, and the relative amount of calcium ready to go into solution may be different from one sample to another according to their mineralogical composition. We have modified the text as follows:

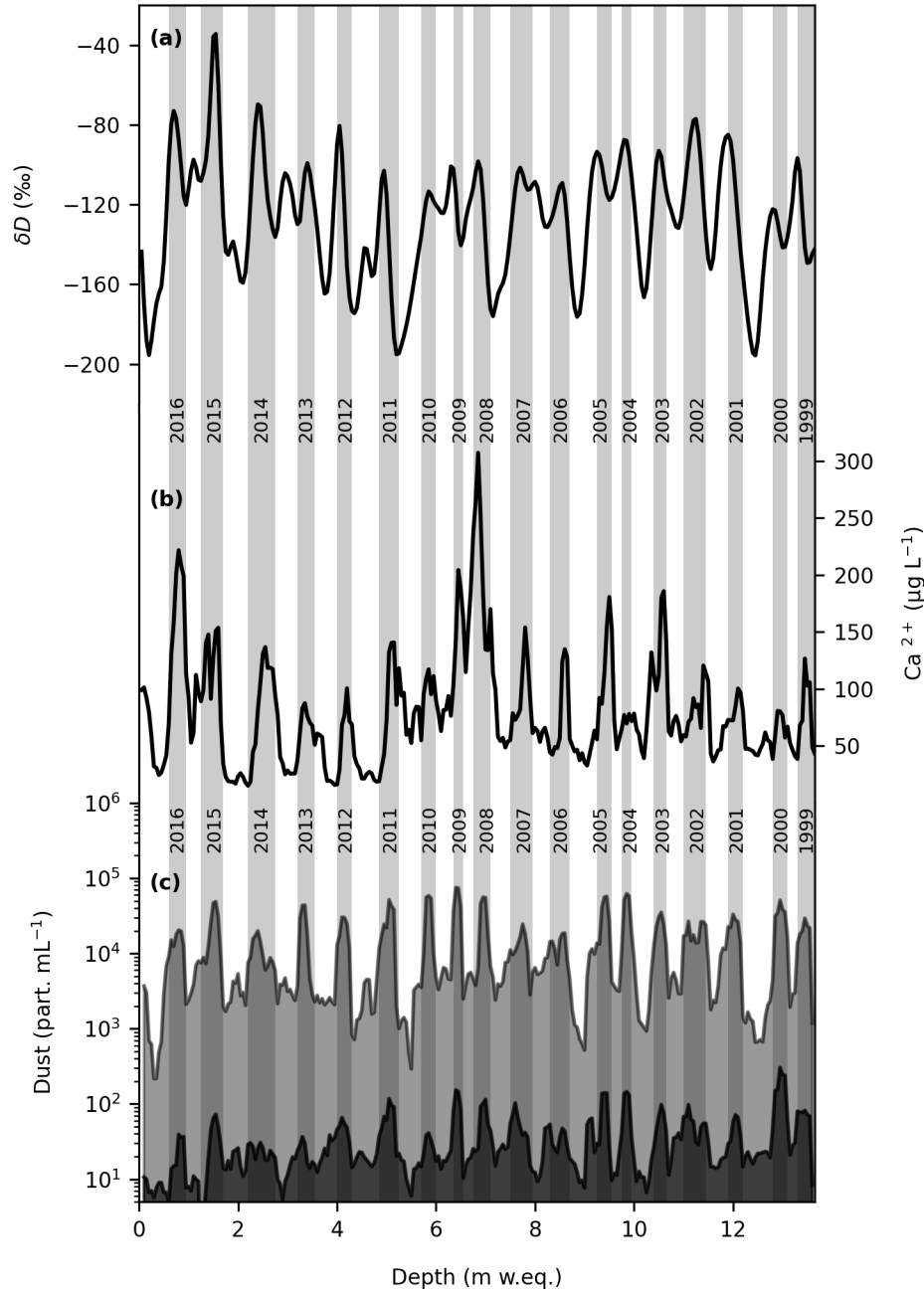


Figure 2: Dating of the Nevado Illimani firn core by annual layer counting (ALC) based on different proxies discussed in the text: (a) δD , (b) ionic Calcium, and (c) total and giant dust particles concentrations (light and dark gray, respectively, both are in logarithmic scale). Gray shaded vertical bands correspond to the dry season for each calendar year. All data are reported as 3-point running average of data re-sampled at 0.05 m w.eq.

L 178: Dust concentration varies from $\sim 2,000$ particles mL^{-1} (hereafter part. mL^{-1}) during the wetter season, to $\sim 10,000$ part. mL^{-1} during the dryer season (median values). When considering extreme values, the variation range exceeds three orders of magnitude, being the lowest concentration during the wet season 150 part. mL^{-1} and the highest one during the dry season $140,000$ part. mL^{-1} . By considering just the giant particles we also

observed a seasonal pattern, but with smaller variations. Giant particles median concentration varies from 15 part. mL⁻¹ during the wet season and 30 part. mL⁻¹ during the dry season. This suggests that giant particles deposition at the Nevado Illimani is less reduced than finer particles deposition during the wet season.

Our results are in agreement with average dust concentrations from Quelccaya ice cap during the 20th century, ~10,000 part. mL⁻¹ and ~25,000 part. mL⁻¹ for the size ranges of 1.6–16 µm and 0.6–20 µm, respectively (Thompson et al., 1986, 2013). The well-defined oscillatory pattern of dust concentration variability reflects the extreme seasonality of precipitation over both local and regional dust sources, and the succession of dry and wet conditions. Therefore, each sample was classified as belonging to the wet or to the dry season according to dust concentration. Sublimation has a limited influence on seasonality (Ginot et al., 2002).

The seasonality of dust concentration is in accordance with the Ca²⁺ record and also with literature studies (Knüsel et al., 2005). However, both records show differences, in particular during the dry season when they are not significantly correlated at the 95% level. Considering our high temporal sampling resolution, this might be associated with slight changes in dust mineralogy, possibly affecting the amount of calcium to be solubilized. Ionic calcium can be primarily associated to calcium sulphate (CaSO₄) and/or calcium carbonate (CaCO₃) (Kutuzov et al., 2019). Because scarcity of calcium carbonates was revealed by mineralogical analyses (Fig. 4, see below), we argue that most of the ionic calcium observed in firn samples is present as a soluble species, probably CaSO₄, and not detectable through Raman spectroscopy on single insoluble particles. However, we consider the possibility of calcium carbonate depletion due to scavenging during dust transport and/or dissolution during the melting of the samples, as discussed by Wu et al. (2016) based on ice core samples from the Tibetan Plateau. In addition, we cannot exclude that Ca-bearing aerosols might have been initially a mixture of pure gypsum and calcium carbonates that successively reacted with atmospheric H₂SO₄, both in the atmosphere or within the snow-pack as the result of post-depositional processes (Röthlisberger et al., 2000; Iizuka et al., 2008).

Specific Comment #5: L 279–280: Again, correlation does not mean that the convection hypothesis is confirmed for either isotopes or giant particles.

[Answer]: We tried to improve our discussion also considering more evidences from literature. We have modified the text as follows:

L 281: Seasonal mean values (Fig. 6) show the significant correlation between these two proxies. Data located in the bottom right corner of Fig. 6 refer to dust deposited onto the Nevado Illimani glacier during the wet season, when more intense convection leads to higher percentage of giant particles and more depleted isotopic composition of precipitation. Points located in the upper left corner, conversely, are associated to winter dry periods when convection and precipitation are reduced and the relative proportion of giant particles is lower. Convective activity is known to significantly affect the isotopic composition of tropical precipitation. Intense regional convection leads to more isotopically-depleted precipitation (e.g. Risi et al., 2008). A proposed mechanism is that convective downdraft promotes the subsidence of higher level water vapor, causing isotopic depletion of low level vapor crossing the eastern Andes (Vimeux et al., 2011). Convective downdrafts, in turn, are often associated with density currents, offering an efficient mechanism for dust lifting (Flamant et al., 2007). Indeed, the leading edge of the density current is characterized by strong turbulent winds that can mobilize dust and mix it through a deep layer (Knippertz et al., 2007). In accordance, events of giant dust particles suspension and transport, as detected by aircraft measurements in north Africa, were related to the occurrence of convective systems (Ryder et al., 2013). Therefore, giant particles on the Nevado Illimani glacier can be reasonably used as

proxy for deep summer convective precipitation. Given their size and geochemical/mineralogical fingerprint, we confidently associate them to local/regional convective activity near the Bolivian Altiplano.

Specific Comment #6: L 281: The poor correlations with dry season values may be driven by the single outlier (upper left portion of Fig. 5). I would like to see the r values in Table 2 reported with and without that outlier. I suspect the story may change significantly.

[Answer]: There is indeed a significant correlation during the dry season. This was adjusted in the Response to Anonymous Reviewer #1, where both Fig. 5 (hereafter Fig. 6) and Table 2 were changed:

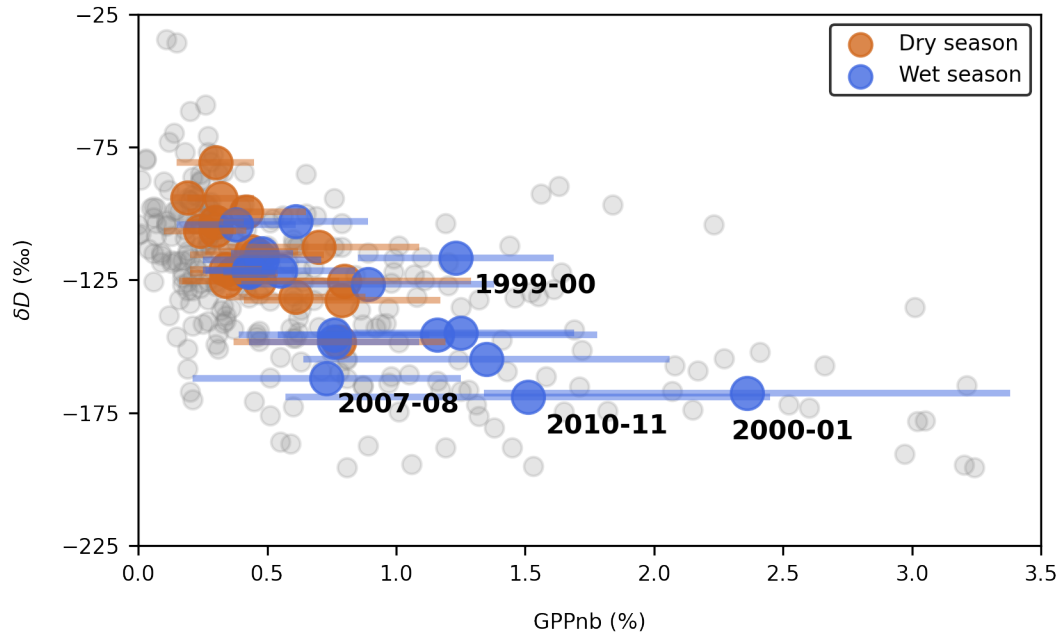


Figure 6: Seasonal mean GPPnb and δD for the dry seasons (orange circle) and the wet season (blue circle). Error bars (horizontal bands) for GPPnb are based on the mean relative standard deviation for the samples integrating each season. Light gray dots on background are raw data. The La Niña events discussed in the text (1999-00, 2000-01, 2007-08, and 2010-11) are reported.

Table 2: Seasonal correlations between Giant Particles Percentage (GPPnb) and δD , rainfall observations and outgoing longwave radiation (OLR). The period between December and February was defined as the wet season, and the period between June and August as the dry season. Significant correlations at 95% level are shown in bold.

GPPnb	δD	El Alto	Calacoto	Patacamaya	Oruro	Potosi	OLR
Wet	-0.71	0.47	0.39	0.76	0.25	0.41	-0.69
Dry	-0.70	0.04	-0.21	-0.09	-0.08	-0.23	0.07

[Answer]: Then we explained the dry season relationships in the following paragraph:

L 306: We conclude that the more intense is summer convection, the higher is the relative number of giant dust particles suspended in the atmosphere and the more depleted is the δD . Winter δD , conversely, is more influenced by the local amount effect, as re-evaporation plays a greater role in such a dry environment (Vimeux

et al., 2011). Although we observed a significant correlation between dry season GPPnb and δD , the rare winter convection seems also to have a low influence in GPPnb variability, as indicated by its lack of significant correlations with both JJA precipitation and OLR (Table 2).

Specific Comment #7: L 290–320: I am struggling to understand the link between convective precipitation and the meteorological data. I would think that convective activity is by nature episodic, so how does monthly precipitation data accurately capture this? And how does one then link convective activity to the entrainment of dust? If it's raining hard from convective activity, how does one get giant particles into the upper atmosphere? And then, without some kind of mechanistic link, I think it is a stretch to conclude that GPP=convective activity=La Nina years.

[Answer]: *First, we changed the text in order to explain the relationship between convective activity and precipitation:*

L 290: In order to test the hypothesis of a relationship between giant particles and convective precipitation, we analyzed monthly precipitation and wind speed records from five meteorological stations located in the central Andes (Fig. 1). Data was provided by SENAMHI, Bolivia (www.senamhi.gob.bo/sismet), whereas monthly outgoing longwave radiation (OLR) data on a $2.5^\circ \times 2.5^\circ$ grid box (Liebmann and Smith, 1996) was obtained from NOAA/OAR/ESRL PSD, Boulder, Colorado, USA (<https://www.esrl.noaa.gov/psd/>). Low OLR values correspond to cold and high clouds which denote enhanced convection. OLR data centered at $17.5^\circ S$, $70^\circ W$ was used as an index of the convective precipitation over the Altiplano, as it presents strong negative correlations with regional rainfall observations (Garreaud and Aceituno, 2001). In addition, both OLR and precipitation data provided similar results when linking δD and regional convection at the Zongo Valley (Vimeux et al., 2011). In fact, it is estimated that deep convection provides 65% of the precipitation over this region, as the orographic lifting of moisture from the Amazon basin through Andes trigger condensation, latent heat release, and strong convective updrafts during the summer (Insel et al., 2010).

Meteorological and OLR datasets were re-sampled into DJF (December to February) and JJA (June to August) time series and compared with our seasonally resolved GPPnb series. For each wet and dry season, defined by dust concentration variability (Sect. 3.1), a mean GPPnb was obtained.

[Answer]: *Then, we have modified the paragraph explaining the possible relations with La Niña as follows:*

Fig. 6 shows that over the 18-years period analyzed in this work, the summer seasons showing the most intense levels of convection (considering both GPPnb and δD) were 2000–2001 and 2010–2011. Both correspond to La Niña periods, as indicated by their DJF Oceanic Niño Index (ONI) of -0.7 and -1.4, respectively. It is well known that the El Niño-Southern Oscillation phenomenon has a significant impact on climate over the Altiplano, especially during the summer season. In particular, meteorological data show that La Niña conditions intensify the meridional pressure gradient on the northern side of the Bolivian High, leading to stronger high troposphere easterly winds, increased eastward upslope flow and enhanced moisture transport (Garreaud, 1999; Vuille, 1999). However, the strong DJF La Niña events of 1999–2000 (ONI = -1.7), and 2007–2008 (-1.6) do not show higher GPPnb or more depleted δD compared to other values of the wet season (Fig. 6). We believe this was due to competing mechanisms controlling moisture transport from the Amazon basin to the Altiplano. In addition to the role played by the upper troposphere easterly winds, the meridional circulation between the tropical North Atlantic Ocean and the western tropical South America also influences the DJF precipitation over the Central Andes, especially in the last two decades (Segura et al., 2020). Evidence based on reanalysis data indicates that

when this meridional circulation is enhanced, the atmospheric stability between the mid and the upper troposphere over the Altiplano is reduced, resulting in increased moisture transport from the Amazon basin (Segura et al., 2020). Thus, we propose a new approach for future studies in tropical Andean glaciers based on giant particles and stable isotopes of snow. This can be used as a complement to a number of other climate proxies and modeling experiments, providing insights into past atmospheric circulation over tropical South America.

General Comment #1: I do not feel that any of the explanations provided for the isotope or particle interpretations stand on their own, and the correlation itself is not sufficient for me to accept the particle-convective activity link.

[Answer]: *Our particle and isotope interpretations are corroborated by regional meteorological data and also by previous studies carried out near the Illimani. Also, both particle-convection and isotopes-convection relationships are in agreement with studies specifically dealing with these links. Our study is a first exploratory work interpreting the giant dust particles content in a tropical Andean firn core, therefore we believe stronger correlations might be obtained by reducing uncertainties (which are extensively discussed in our manuscript) in giant particles measurements, but this is something that we will explore in the future and goes beyond the aim of this work.*

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