Response to Anonymous Reviewers #1 and #2 to manuscript TC-2020-55

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

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This document shows a point-by-point reply to comments from both reviewers (RC1 and RC2). It is followed by a marked-up version of the manuscript showing the changes we have made. All line numbers correspond to the marked-up manuscript.

RC1, Specific Comment #1: Line 40: while moist air advection from the east is suppressed.

[Answer]: We fixed this sentence.

RC2, Specific Comment #1: L 79–80: 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.

RC1, Specific Comment #2: L 105: Explain ice layer formation

[Answer]: We added an explanation about ice layer formation to the text.

RC1, Specific Comment #3: L 120: Give some more details about the standard protocol: how is settling of giant particles prevented, stirring, the mean standard deviation of what?

RC1, General Comment #1: The correlation between giant particles and δD , considered as proxy for convection is based on the relative mass proportion of giant particles. Considering the fact that determining giant particles in a liquid sample is tricky, because they tend to settle, you need to substantiate the robustness of your results better. Specify if the given mean standard deviation between the two measurements with the coulter counter (lower than 3%) applies also to the giant particles alone.

[Answer]: We added details about the standard protocol to the text. Then, we added a discussion about the deviations between the measurements for the giant particles. Number distributions showed a lower uncertainty than mass distributions. Therefore, we converted (along all the text) the proportions of giant particles in terms of mass (GPPms) to terms of number (GPPnb).

RC1, Specific Comment #4: L 155: LOD: concentration in dust or ice sample?

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

RC1, General Comment #2: Show examples of the size distributions for the wet and dry season.

[Answer]: We added a new Figure to the Supplement (Fig. S2) showing these examples.

RC2, Specific Comment #2: L 170–171: 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 L 52–53. Then, we added the location of the potential dust sources, discussed in Sect. 3.2, to Fig. 1. Finally, we fixed the text in L 170–171.

RC1, Specific Comment #5: Fig. 2: Show PC1 separately, in the current figure it is difficult to distinguish δD and GPPms.

RC2, **Specific Comment #3:** I don't understand what the PCA value adds.

[Answer]: We removed the PCA from the text.

RC1, General Comment #3: Show in addition the record of mass or number of giant particles for every sample. From the presented data it is unclear if the number or mass of giant particles also has a seasonal or any other variability or not.

Since total particle mass concentrations are low during the wet season (when you observe the correlation), the relative mass proportion is the ratio of two small numbers, probably having a large uncertainty. Add uncertainty bars.

[Answer]: In Fig. 2 we plotted the number of giant particles together with the total number of particles. We have also included a discussion of the variability of its concentration in L 206–207. The proportion of giant dust particles (GPPnb) was plotted in a new Fig. 3.

RC1, Specific Comment #6: L **216–217**: Calcium carbonate is also soluble in water (solubility 13 mg/l), and most likely therefore not detected in mineralogical analyses.

RC2, Specific Comment #4: L 211–222: 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]: 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.

RC1, Specific Comment #7: L 232: Massive dust deposition – I think this is exaggerated. Have you compared dust concentrations at Illimani with that in other high-alpine ice cores? Are the dust layers visible in the core?

[Answer]: We changed the word massive. Then, we compared our dust concentration record with that of another Andean ice core (Quelccaya).

RC2, Specific Comment #5: L 234–251: 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.

RC2, Specific Comment #6: L 259: 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.

[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 L 126–131.

RC2, Specific Comment #7: L 323–325: 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 (L 332-339).

RC2, Specific Comment #8: 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]: When we started working with number distributions (GPPnb) instead of mass distributions (GPPns), we changed both Fig. 5 (hereafter Fig. 6) and Table 2. Indeed, there is a significant correlation between GPPnb and δD during the dry season. The dry season relationships were explained in L 381–386.

RC1, General Comment #4: Before being lifted up by convection, dust particles need to be mobilized from the ground, which requires strong wind (advection). Have you checked wind speeds in the dust source areas? Dust source areas are located SE of Illimani, whereas humidity in the wet season originates in the Amazon Basin, due to stronger easterly winds and eastward upslope flow (especially enhanced during La Niña conditions). The link between local dust sources and easterly upslope flow is not immediately obvious. Your hypothesis would require large-scale convective processes also affecting the Altiplano. Do you have indications for that? Your precipitation data show the opposite. Hurley et al. (2015, 2016) offer a different hypothesis for depleted stable isotope ratios, i.e. the amount effect is associated with South American cold air incursions, linking synoptic-scale disturbances and monsoon dynamics to tropical ice core δ^{18} O. Have you considered that as potential explanation for dust mobilization/uplift? How was the attribution to wet and dry season or even DJF, JJA for the ice core values conducted? This is critical and needs to be explained.

[Answer]: We have added a more detailed explanation of our proposed mechanism for dust mobilization/uplift in L 358–379. A better description of the method we used to assign the wet and dry seasons to the firn core values was included in Sect. 3.1 (L 255–258).

RC2, Specific Comment #9: L 387–408: 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]: We changed the text to explain the relationship between convective activity and precipitation (L 344–350). Then, we modified the paragraph that explains the possible relations with La Niña (L 387–408).

RC2, 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.

RC1, Specific Comment #8: Table S2: Give also ice core concentrations for comparison with other publications.

[Answer]: We added a comparison in Table S2.

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

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Abstract. A deeper understanding of past atmospheric circulation variability in the Central Andes is a high-priority topic in paleoclimatology, mainly because of the necessity to validate climate models used to predict future precipitation trends and to develop mitigation and/or adaptation strategies for future climate change scenarios in this region. Within this context, we here investigate an 18-years firn core drilled at the Nevado Illimani in order to interpret its mineral dust record in relation to seasonal

- 5 processes, in particular atmospheric circulation and deep convection. The core was dated by annual layer counting based on seasonal oscillations of dust, calcium and stable isotopes. Geochemical and mineralogical data show that dust is regionallysourced in winter and summer. During austral summer (wet season) an increase in the relative proportion of giant dust particles $(\emptyset > 20 \,\mu\text{m})$ is observed, in association with oscillations of stable isotope records (δ D, δ^{18} O). It seems that at Nevado Illimani both the deposition of dust and the isotopic signature of precipitation are influenced by atmospheric deep convection, which is
- 10 also related to the total amount of precipitation in the area. This hypothesis is corroborated by local-regional meteorological data. The interpretation of giant particle and stable isotope records suggests that during La Niña years, summer convection activity is enhanced, in agreement with atmospheric circulation studies downdrafts due to convective activity promote turbulent conditions capable of suspending giant particles in the vicinity of the Nevado Illimani. Giant particles and stable isotopes, when considered together, can be therefore used as a new proxy for obtaining information about deep convective activity in the
- 15 past, which is ultimately related to paleo-ENSO variability.

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1 Introduction

Climate variability in the Central Andes and the Bolivian Altiplano has a strong link with atmospheric circulation and rainfall anomalies over the rest of tropical South America (e.g., Vuille, 1999). Over the Altiplano, a semi-arid plateau in the Central

- 20 Andes with a mean elevation of 3800 m above the sea level (a.s.l.) (Fig. 1), climate variations have a direct effect on the availability of water resources, with severe economic and social impacts (Garreaud and Aceituno, 2001). Recent Andean glaciers retreat The recent retreat of Andean glaciers due to global climate change (Rabatel et al., 2013) poses issues not only for water availability (Soruco et al., 2015), but also for the preservation of glaciers as natural archives that could be soon lost. For example, in the period between years 1963 and 2009, Nevado Illimani (Fig. 1) lost approximately 35% (9.49km²) of its total area of
- 25 ice (Ribeiro et al., 2013), while on (Ribeiro et al., 2013). On Quelccaya ice cap (13°54'S, 70°48'W, 5670 m a.s.l., Fig. 1), the seasonal variations in stable isotopes were attenuated in of stable isotopes began to deteriorate because of the percolation of meltwater through firm, affecting the record corresponding to the latter half of the twentieth centuryrecord due to melting and percolation through the firm, although the seasonality in dust was of the dust record is still preserved (Thompson et al., 2017). Precipitation on the Bolivian Altiplano is largely concentrated in the summer months Garreaud et al. (2003)(Garreaud et al., 2003)
- 30 , in response to the peak phase of the South American Summer Monsoon (SAMS). During summer (December-January-February, DJF), the intensification and southward displacement of the Bolivian High Anticyclone (thereafter referred as Bolivian High) promotes strong easterly winds and a turbulent entrainment of easterly air masses over the Andean ridge. In addition, the upward motion over western Amazon, which is part of the meridional circulation between the tropical North Atlantic and western tropical South America, leads to increased convection and reduced tropospheric stability over the Central Andes
- 35 (Segura et al., 2020). Such an atmospheric context favours the establishment of an eastward upslope air-flow and the advection of moisture from the Amazon basin toward the Andes (Zhou and Lau, 1998). Accordingly, summer precipitation is strongly associated with deep convective activity, enhanced by high amounts of water vapor in the boundary layer which destabilize the tropospheric column over the Altiplano (Garreaud, 1999). Precipitation during the rest of the year is scarce: during winter months (June–July–August, JJA) the Altiplano is generally very dry, and advection of dry air from the Pacific region is
- 40 promoted, being moist air advection the advection of moist air from the east is suppressed. Strong winter westerly winds and dry conditions allow massive local transport and deposition of dust over the Central Andean glaciers, and for this reason high seasonal contrast exists between wet summer and dry winter snow layers in terms of dust and aerosol content (Knüsel et al., 2005).

Besides seasonal variability, year-to-year climate over the Altiplano is also influenced by conditions in the tropical Pacific

- 45 Ocean. During warm phase of the El Niño-Southern Oscillation (ENSO), the Altiplano climate is dry. Dry summers associated with El Niño events in the tropical Pacific are characterized by enhanced westerly flow over the tropical Andes inhibiting moisture advection from the Amazon basin (Knüsel et al., 2005; Thompson et al., 2013). Conversely, wet summers associated to with a cooling of the tropical Pacific (La Niña events) promote further ingression of humid easterly air masses from the Amazon Basin.
- 50 Developing an annually-resolved ice core record from the Altiplano is an opportunity to enhance our knowledge about present and past climate variability in the tropical Andes region. 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 and regional (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. During the summer, conversely, the concentration of aerosol and

55 particulate matter is much lower while impurities of anthropogenic origin (e.g. Cu, As and Cd) are observed in higher proportions (Correia et al., 2003).

With the aim to enhance our knowledge about past and present climate variability in the tropical Andes region, a new shallow firn core (23.8 m long) was drilled on the Nevado Illimani (Cordillera Real, Eastern Bolivian AltiplanoEastern Cordillera, Central Andes), as an integration of the Ice Memory project (https://www.ice-memory.org). In this study we investigate mineral

- 60 dust aerosol variability and provenance in this firn core, through the analysis of dust concentration, grain size, geochemistry and mineralogy. The very pronounced seasonal variations of the analyzed proxies allowed for the development of a precise chronology, which covers the 1999–2017 period, and for the investigation of correlation between dust records and other proxies. Dust particles entrapped in firn samples seem to originate from regional sources during both winter and summer, despite minor mineralogical differences between the two seasons are observed.
- 65 An interesting result concerns the presence of giant dust particles (presenting a diameter larger than 20 μm), whose variability is correlated to the stable isotope record. Our data and local meteorological observations suggest that this relationship could be explained as a consequence of 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
- 70 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). A
- 75 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 proportion of giant dust particles into firm is correlated with local meteorological observations, and in

particular with atmospheric deep convection over the Bolivian Altiplano during summer. This study shows for the first time that

- 80 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 periodsand in particular during La Niña years, in agreement with observations concerning atmospheric circulation anomalies in the area (Vuille, 1999). From this perspective, this study demonstrates the great potential of giant particles records, which is strongly influenced by climatic and meteorological processes at high altitude continental glaciers. This is a first exploratory
- 85 work, analysis of a longer ice core would be desirable in the future to investigate the relationships between giant dust particles deposition, atmospheric deep convection and periodic climatic phenomena (ENSOLa Niña).

2 Material and Method

2.1 Field campaign and firn core sampling

- 90 Nevado Illimani (16°37'S, 67°46'W, 6438 m a.s.l.) is located 50 km southeast of the Bolivian capital, La Paz, and 180 km southeast of Lake Titicaca (Fig. 1). Its approximate dimensions are 10 per 4 km, with some peaks above 6,000 m a.s.l. Nevado Illimani consists of a granodiorite pluton of Late Oligocene age, with a short belt composed by a coeval dacitic flow located near the south-western border of the pluton (McBride et al., 1983; Jiménez and López-Velásquez, 2008). In June 2017, a 23.8 m firn core (corresponding to 13.75 m water equivalent) was drilled at an altitude of 6350 m a.s.l. on the saddle between the
- 95 two Nevado Illimani summits, approximately where two deep ice cores were recovered in June 1999 (Knüsel et al., 2003). The expedition was coordinated by a French, Russian, Bolivian and Brazilian team and integrated the Ice Memory project (Université Grenoble Alpes Foundation). An EM-100-1000 electromechanical ice core drill (Cryosphere Research Solutions, Columbus, Ohio, USA) was used for the drilling and three cores were extracted, two down the bedrock (136 and 134 m) and the core for this study (23.8 m).
- 100 The cores (diameter of 10 cm), consisting of 24 sections of approximately 1 m length, were transported by mountain porters from the drilling site to the base camp during the night in order to prevent melting. Once at the base camp, they were immediately transported to a refrigerated container located in La Paz, where temperature was set at -20 °C. After the drilling campaign, the container was shipped to the Institut des Géosciences de l'Environnement (IGE, Université Grenoble Alpes, France) where the core sections were weighted and cut longitudinally using a vertical band saw in a cold room (at -15 °C).
- 105 Ice layers (less than 5 cm thick) occurred frequently along the firn core (Fig. S1), generally in closely spaced groups of 2 or 3 individual layers. In addition, less than 1 cm thick ice layers (or possibly wind crusts, hardly distinguishable from the former at greater depths) commonly occurred along the core. These features indicate few events of meltwater percolation, and ensure the proxies recorded in the firn core are well-preserved.

One quarter of the original core was dedicated to dust analyses and transported for this purpose to the EUROCOLD facility of

110 the University of Milano-Bicocca (Italy). There, firn sections were transversely cut at 5 cm using a horizontal band saw with a cobalt steel blade and 464 samples were obtained. These were manually decontaminated by mechanical scraping with a clean ceramic knife inside a laminar flow high-efficiency particle air (HEPA) ISO 5 Class bench located in an ISO 6 Class cold room. Once decontaminated, the samples were put into clean Corning centrifuge tubes and kept frozen until the measurements.

115 2.2 Coulter Counter Analysis

Samples were melted at room temperature, and a ~ 10 mL aliquot from each was transferred to an Accuvette Beckman Coulter vial, previously washed with Millipore Q-POD[®] Element ultra-pure water (in an ISO 5 Class laminar flow bench located inside an ISO 6 Class clean laboratory). Each sample was treated following standardized protocols (Delmonte et al., 2002). A Beckman Coulter Multisizer 4 equipped with a 100 μ m orifice was used to measure dust concentration and grain size (400 size

120 channels within the 2–60 µm interval of spherical equivalent diameter). Each sample was measured twice (consuming 0.5 per

measurement), the mean standard deviation between the two measurements being lower than 3% Samples were continuously stirred until the moment of the analysis, as the larger particles tend to settle rapidly. Systematic analysis of ultra-pure water blanks allows estimating a mean signal to noise ratio around 97. Each sample was measured twice, consuming 0.5 mL per measurement. The mean relative standard deviation (RSD) between these two measurements considering both the number and

- 125 the mass of particles was 7% and 29%, respectively. 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 μ 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
- 130 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%.

2.3 Instrumental Neutron Activation Analysis

- A set of 10 samples was dedicated to Instrumental Neutron Activation Analysis (INAA). Samples were selected from different depth intervals along the core (see Table 1, "N" series, and Table S1 for precise depths), in order to be representative of both the dry and the wet seasons. The samples were filtered using PTFE Millipore[®] membranes (0.45 µm pore size, 11.3 mm diameter) previously rinsed in an ultra-pure 5% solution of bi-distilled HNO₃— according to the procedures adopted by Baccolo et al. (2015). The filtration took place in an ISO 5 Class laminar flow bench. Two blank membranes, that underwent the same cleaning procedures, were prepared by filtering 300 mL of MilliQ (Millipore[®]) water. For calibration and quality
- 140 control, we used certified solid standards: USGS AGV2 (ground andesite), USGS BCR2 (ground basalt), NIST SRM 2709a (San Joaquin soil) and NIST SRM 2710a (Montana soil). In addition, standard acid solutions for each analyzed element were prepared with concentrations in the order of $\mu g g^{-1}$. Blanks for the empty flask and for the ultra-pure acid solution used to prepare the liquid standards were also measured. Samples, standards and blanks were irradiated at the Applied Nuclear Energy Laboratory (LENA, University of Pavia, Italy) by a Triga Mark II reactor of 250 kW. The "Lazy Susan" channel, neutron flux
- equal to $2.40 \pm 0.24 \text{ x } 10^{12} \text{ s}^{-1} \text{ cm}^{-1}$, was used to identify Ce, Cs, Eu, Hf, La, Sc, Sm, Th, and Yb. Samples were successively transferred at the Radioactivity Laboratory of the University of Milano-Bicocca, in order to acquire gamma spectra by means of a high-purity Germanium detector HpGe (ORTEC, GWL series), following the standardized procedure developed for low-background INAA (Baccolo et al., 2016).

The masses of the elements in each sample were determined by comparing spectra related to standards and samples (Baccolo

150 et al., 2016). In order to compare different spectra, the time of acquisition, the radionuclide decay constant, the cooling time and a factor considering radioactive decay during the acquisition were kept into account. The detection limits were calculated considering three times the standard deviation of the blank signal. The uncertainties for each element were calculated based on the mass measurements, the adjustment for the spectrum, the subtraction of the blanks and the standard concentration uncertainties. Errors for the elemental concentrations in our samples ranged from 3% for La to 17% for Cs, and the detection 155 limits ranged from 0.1 μ g per gram of dust for Sm to 7 μ g g⁻¹ for Ce (Table S2). Full analytical details can be found in Baccolo et al. (2016).

The Enrichment Factor (EF) normalization was calculated for each element considering as a reference the mean composition of the upper continental crust (UCC) (Rudnick and Gao, 2003). Scandium (Sc) was chosen as the crustal reference element following Eq. (1):

160

$$EF(x) = \frac{\left(\frac{[X]}{[Sc]}\right)_{sample}}{\left(\frac{[X]}{[Sc]}\right)_{UCC}} \tag{1}$$

Scandium was chosen as reference element because it is poorly affected by processes altering its mobility in hosting minerals, and its biogeochemical cycle is almost unaffected by anthropogenic activities (Sen and Peucker-Ehrenbrink, 2012). In addition, Sc is highly correlated with other lithogenic elements, such as Ce (r = 0.997), used by Eichler et al. (2015) as a crustal

165 reference for Nevado Illimani samples, and La (r = 0.989). The choice of Sc has been also determined by its easy and precise determination through INAA.

2.4 Micro-Raman Spectroscopy

We used single-grain Raman spectroscopy to identify mineralogy of dust particles having a diameter smaller than 5 µm. This

- 170 because this kind of analysis was carried out for provenance purposes, thus excluding the large and the giant particles of obvious local originconsidering particles expected to travel over longer distances. A set of four samples (see Table 1, "R" series, and Table S1) was prepared, following the procedure described in previous studies (Delmonte et al., 2017; Paleari et al., 2019) specifically developed for small dust grains. Two samples are representative of mineral dust deposited in the dry season (high dust concentration) whereas two represent dust from the wet season (low dust concentration, or "background"). Measurements
- 175 were performed by using an InVia Renishaw micro-Raman spectrometer (Nd YAG laser source, $\lambda = 532$ nm) available at the Laboratory for Provenance Studies (UNIMIB). We identified the mineralogy of more than 630 grains, excluding organic particles possibly related to contamination and particles with an undetermined spectrum or with no signal.

2.5 Stable Isotope and Ion Chromatography Analysis

180 The dust analyses described above used one quarter of the longitudinally-cut firn core. A second quarter was shipped in a frozen state to the Climate Change Institute (CCI, University of Maine, USA) for ion chromatography (IC) and water stable isotope analysis.

At the CCI, in a cold room set at -20 $^{\circ}$ C, we cut longitudinal sections of the core with a vertical band saw to separate an inner and an outer part. The latter was sampled by transverse cuts approximately every 12 cm using a stainless-steel hand saw

185 (resulting in 190 samples) and stored in plastic bottles for stable isotope ratio determination. Decontamination of the inner

part was performed by scraping with a clean ceramic knife under a laminar flow HEPA bench inside the cold room. Then, the decontaminated inner part was sampled for IC analysis by a continuous melter system (Osterberg et al., 2006) also in an ISO 6 Class clean room. The mean sample resolution was 3 cm, resulting in 767 samples. We measured Ca^{2+} concentration using a ThermoScientificTMDionexTM Ion Chromatograph ICS-6000 analytical system fitted with suppressed conductivity detectors

with a Dionex AS-HV autosampler. The method detection limit (MDL) was defined as 3 times the standard deviation of the blank samples (MilliQ water, 10 blank samples). The detection limit for Ca²⁺ was 21.05 µg L⁻¹. The δD and the δ¹⁸O were determined using a Picarro L2130-i wavelength-scanned cavity ring-down spectroscopy instrument (Picarro Inc., USA) with a precision of 0.1‰.

195 **3 Results and discussion**

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3.1 Seasonal variability of proxies and firn core chronology

We established a chronology for the Nevado Illimani firn core based on annual layer counting (ALC), considering the pronounced seasonal oscillation of dust concentration, calcium and water stable isotopes (Fig. 2). Dust concentration variations, which are recognized for being useful for ALC in tropical and continental ice cores (Ramirez et al., 2003; Kutuzov et al., 2019), span about two-orders of magnitude between the summer and the winter. Dust concentration varies from \sim 850 \sim 2,000 particles mL⁻¹ (hereafter part. mL⁻¹) during the wetter season, to \sim 4,800 \sim 10,000 part. mL⁻¹ during the dryer season (median val-

- ues). The two size distributions shown in Fig. S2, illustrate this variability. When considering extreme values, the variation range exceeds three orders of magnitude, being the lowest concentration during the wet season 70-150 part. mL⁻¹ and the highest one during the dry season 20-30-140,000 part. mL⁻¹. Our results are in agreement with average dust concentrations
- 205 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). By considering just the giant particles we also observed a seasonal pattern, with median concentrations of 15 part. mL^{-1} during the wet season and 30 part. mL^{-1} during the dry season. The well-defined oscillatory pattern of dust concentration variability reflects the extreme seasonality of precipitation over the Altiplano both local and regional dust sources, and the succession of dry and wet conditions. Sublimation has a limited influ-
- ence to this seasonality (Ginot et al., 2002).

Dust concentration is in accordance with the Ca²⁺ record and in agreement 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

215 (CaSO₄) and/or calcium carbonate (CaCO₃) (Kutuzov et al., 2019). Because scarcity of calcium carbonates was revealed by mineralogical analyses (Fig. 34, 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,

220 we cannot exclude that Ca-bearing aerosols might have been initially a mixture of pure gypsum and calcium carbonates that successively reacted with atmospheric H_2SO_4 (Röthlisberger et al., 2000), both in the atmosphere or within the snow-pack as the result of post-depositional processes (Röthlisberger et al., 2000; Iizuka et al., 2008).

The regular succession of dry dusty periods and wet periods can be associated with the seasonal onset and decay of the Bolivian High, a high pressure system which is well developed and centered over Bolivia (Lenters and Cook, 1997). When the Bolivian

- vian High is particularly strong and displaced southward of its climatological position, easterly flow in the high troposphere is enhanced as well as moisture advection from the interior of the continent to the Altiplano. This moisture transport from the Amazon Basin toward the Altiplano induces a notable amount of precipitation over the Altiplano (wet season), associated with strong summer convection. The relatively low dust concentration found in the Illimani snow during the summer period is therefore related to particle dilution in the snowpack because of increased precipitation and reduced regional dust mobilization
- 230 deriving from wetter soil conditions. Conversely, during winter (JJA) months, conditions over the Altiplano are typically dry, leading to higher dust availability. At that time of the year, the winter westerly flow over the entire region promotes eastward dust transport towards the Nevado Illimani, leading to massive significant higher dust deposition in the firn layers representing on average on average about 85% of the total annual dust mass-particles there deposited.
- 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)(Dansgaard, 1964; Vuille et al., 2003; Risi et al., 2008). In this context, deep atmospheric convection also plays a role on stable isotope composition (Vimeux et al., 2005). Modelling 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, modelling studies (e.g., Bony et al., 2008; Risi et al., 2008)
- 245 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. Satellite 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. Convective precipitation over the Bolivian Altiplano is enhanced during the wet summer season, leading to the
- emergence of clear seasonal oscillations in the stable isotope records of Nevado Illimani firn core which can be used for ALC and to develop a chronology.

Considering the pronounced seasonal changes in dust concentration, Ca^{2+} and stable isotopes, it is possible to assign to the base of the Illimani firm core an age corresponding to the beginning of 1999 AD. The firm record thus covers the 18-years

period from early 2017 to early 1999 and the average accumulation rate can be estimated in the order of approximately 750

255 mm of water equivalent per year, slightly higher than the one inferred by Knüsel et al. (2003). In addition, we classified each sample as belonging to the wet or to the dry season according to concentration levels for all dust, Ca^{2+} and stable isotopes. The samples belonging to the depth intervals in the gray areas of Fig. 2 were classified as dry season samples. All other samples were classified as belonging to the wet season.

Interestingly, we note a close correspondence between the variability of stable isotopes and the proportion of giant particles

- 260 in firn (Fig. 2a3): oscillations of the stable isotope record ($\delta Din Fig. 2a$) closely follow the percentage of giant dust particles (GPPms in Fig. 2aGPPnb). During the dry season, giant particles are proportionally less abundant (average GPPms 28GPPnb 0.5%) whereas the isotopic composition of snow is less negative (average -111113% for δD ; -15% for $\delta^{18}O$). Conversely, during the wet season when giant dust particles are at their annual maximum (average GPPms 48GPPnb 1%) the isotopic composition of snow is more depleted (-136141% for δD ; -18% for $\delta^{18}O$), reaching its minimum. The first principal
- 265 component of these two series (GPPms and δD), also reported in Fig. 2a, explains approximately 83% of the variance of the records.Considering the pronounced seasonal changes in dust concentration, and stable isotopes, it is possible to assign to the base of the Illimani firn core an age corresponding to the beginning of 1999 AD. The firn record thus covers the 18-years period from early 2017 to early 1999 and the average accumulation rate can be estimated in the order of approximately 750 mm of water equivalent per year, slightly higher than the one inferred by Knüsel et al. (2003).

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3.2 Dust provenance: mineralogy and geochemistry

The mineralogical composition of fine dust (<5 µm) deposited onto the Illimani firn layers reveals that the most abundant mineral phases are quartz, feldspars (alkali feldspars and plagioclase) and phyllosilicates (Fig. 34, Table S3). Phyllosilicates are mainly represented by muscovite-illite (and/or smectite, hardly distinguishable from illite by their Raman spectra) and secondarily by kaolinite (representing 7.5% in the dry season and 1.5% in the wet season). Altogether, quartz, feldspars and phyllosilicates account for 75–78% of mineral particles during both the wet and the dry season, but phyllosilicates are particularly abundant during the wet season, when they represent approximately 44% of minerals (Fig. 3a4a). We believe that the increased abundance of muscovite-illite during the wet season is related to different depositional regimes. During the dry winter, aerodynamic platy-like phyllosilicates can remain in the atmosphere for longer periods and are only partially deposited. On

- 280 the contrary, during the wet summer strong scavenging is associated to heavy precipitations at Nevado Illimani (Bonnaveira, 2004), enhancing the removal of mineral particles from the atmosphere, including phyllosilicates. Titanium oxides and iron oxides/hydroxides are present in all samples. Hematite is twice as abundance as goethite. This is typical in regions dominated by arid conditions or where a prolonged warm and dry season is followed by a shorter and wetter period (Journet et al., 2014). Accessory minerals include carbonate and tourmaline, and very rare pyroxenes (Table S3). Such
- 285 a mineralogical composition is coherent with the felsic to intermediate plutonic volcanic source rocks, suggesting that most of the dust deposited at Nevado Illimani has a local/regional provenance, both in the wet and in the dry seasons. Low-background INAA analyses allowed determining the EFs for different rare earth elements (REEs), which are non-mobile

and therefore widely used as provenance tracers (McLennan, 1989; Moreno et al., 2006; Gabrielli et al., 2010). In Fig. 4a 5a the Yb/La and the Eu/Sm elemental ratios are used to compare dust samples retrieved from Nevado Illimani firn samples

- and literature data concerning geological samples from the Altiplano-Puna Volcanic Complex (Ort et al., 1996; Lindsay et al., 2001) and potential source areas (PSAs) in South America (Gaiero et al., 2004, 2013). The Yb/La ratio can be used to appreciate wether heavy and light REEs are enriched or depleted with respect to each other, whereas the Eu/Sm ratio is a proxy for the europium anomaly, usually calculated considering Gd, not detected by our analytical method. The comparison reveals that Nevado Illimani dust has a composition similar to APVC crystal-rich ignimbrites, pointing to a correspondence with samples
- 295 from the Northern Puna region, and not with samples from the salt lakes present in the Altiplano (Uyuni and Coipasa salars). These pieces of evidence agree with previous analyses of strontium and neodymium isotopes on Nevado Illimani ice core dust (Delmonte et al., 2010) and with the geochemical signature of local sources in the Altiplano (Gili et al., 2017), supporting the hypothesis that dust deposited at Nevado Illimani is sourced by sediments present in Southern Altiplano-Northen Puna areas. The EFs of Ce, La, Sm, Eu and Yb is similar between samples with higher percentage of giant particles (GPPnb > 1%, charac-
- 300 teristic from the wet season) and samples with background GPPnb (Fig. 4b5b), supporting what has been observed in relation to the mineralogical composition of samples from the wet and dry seasons. However, two important exceptions to this pattern occur in relation to Hf and Cs. High GPPnb samples show an anomalous Hf enrichment when compared to background samples, a feature that was observed also in Saharan dust samples (Castillo et al., 2008) and attributed to the presence of detrital zircon (Zr, Hf)SiO₄ in samples showing the coarsest grain size. As discussed by Vlastelic et al. (2015) the Hf enrichment observed
- 305 in samples where the percentage of giant particles is high might be related to the presence of a few silt-sized zircon grains, which would in turn require high energy (turbulence) to lift them and keep them in suspension into the atmosphere. In this study, however, zircon grains were not detected by Raman Spectroscopy, which indicated greater abundance of phyllosilicates in dust deposited during the wet summer season. The Hf enrichment in these samples may thus be related instead to tiny zircon inclusions within phyllosilicate particles.
- 310 Another deviation from background is related to cesium. Because of the very common Cs-K exchange into interlayer sites in illite-muscovite, the Cs enrichment in samples from the wet season may also be related to the greater abundance of phyllosilicates (Cremers et al., 1988; Rosso et al., 2001).

3.3 Relationship between the giant particles and deep convection

- The absolute concentration of dust in firn and ice cores depends on many factors including the snow accumulation rate, the dust source strength (which includes soil aridity/wetness, vegetation cover and any other factor influencing the quantity of particles available for deflation), as well as transport processes, which also affect the residence time of particles in the atmosphere (mainly in the case of long-range transport). Conversely, the dust size distribution and the relative proportion of particles within a given grain size merely depends on transport conditions (Delmonte et al., 2004, 2017).
- 320 In the case of Nevado Illimani, where dust is mostly regionally-sourced, we believe that the dust concentration is primarily modulated by the seasonally-varying source strength, mainly depending on source aridity and humidity, and by accumulation

rate. Conversely, the dust size distribution, in particular the relative mass (or number (or mass) of giant particles is related to the relative strength of local turbulence and deep convection. Indeed, we observe an interesting correlation (Fig. 2a3) between the relative mass number of giant particle percentage (GPPms) and δD GPPnb) and δD , both related to convective activity.

Seasonal mean values (Fig. 5) show that the 6) show the significant correlation between these two proxies is significant only in the summer wet season. Data located in the bottom right corner of Fig. 5–6 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

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- 330 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 Cordillera of the Central Andes (Vimeux et al., 2011). This downdraft is generated by the cooling of air due to the reevaporation of the
- 335 falling precipitation, which is favored by the often dry (unsaturated) conditions over the Central Andes. Convective downdrafts 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 par-
- 340 ticles 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.

In order to test the hypothesis of a relationship between giant particles and convective precipitation, we analyzed monthly precipitation records from five meteorological stations located in the central Andes (Fig. 1). Data, and monthly outgoing longwave

- 345 radiation (OLR) centered at 17.5°S, 70°W. Low OLR values correspond to cold and high clouds which denote enhanced convection. 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). In agreement, OLR shows strong negative correlations with regional rainfall observations over the Bolivian Altiplano (Garreaud and Aceituno, 2001). Furthermore, both OLR and precipitation data provided similar
- 350 results when linking δD and regional convection at the Zongo Valley (Vimeux et al., 2011). Precipitation data was provided by SENAMHI, Bolivia (www.senamhi.gob.bo/sismet), whereas monthly outgoing longwave radiation (OLR) OLR data on a 2.5° x 2.5° grid box (Liebmann and Smith, 1996) was obtained from NOAA/OAR/ESRL PSD, Boulder, Colorado, USA (https://www.esrl.noaa.gov/psd/). OLR data centered at 17.5S, 70W 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).
- 355 These datasets were re-sampled into DJF (December to February) and JJA (June to August) time series and compared with our GPPms series. seasonally resolved GPPnb series. For each wet and dry season, defined according to dust concentration, Ca²⁺,

and δD records (Sect. 3.1), a mean GPPnb was obtained.

Results reported in Table 2 clearly show that during wet season GPPms GPPnb is positively correlated (at 95% level) to with DJF precipitation at El Alto (16.5S, 68.2W, 4034 a.s.l.) and Patacamaya (17.2°S, 67.9°W, 4498 m a.s.l.), which are closer

- 360 to Nevado Illimani than the other meteorological stations. Because of the convective nature of precipitation episodes and high spatial variability in precipitation over the Bolivian Altiplano(Aceituno, 1996). Rainfall variability over the Altiplano is strongly dependent on the intensity of the moisture transport over the eastern slope of the Eastern Cordillera, but also depends on the local amount of near-surface water vapor (Garreaud, 2000). This later responds to the complex topography of the Central Andes, resulting in differences between the precipitation records of Altiplano's meteorological stations (Aceituno, 1996). Thus,
- 365 it is expected that only precipitation data from stations in the closest vicinity of the Nevado Illimani show good agreement correspondence with glaciological data. In agreement, Knüsel et al. (2005) observed that the Patacamaya precipitation record was better correlated (compared to El Alto) with the dust related ions record from Nevado Illimani, suggesting that the precipitation regime in the area south of Nevado Illimani influences its dust record.
- Also, GPPms As convective clouds are formed over the Eastern Cordillera, the reevaporation of precipitation falling through the
 dry atmospheric boundary layer on its western slope leads to a downward flow of cold air over the highly complex terrain. This is driven by differences in density in relation to the environment, similar to the mechanism proposed by Knippertz et al. (2007). The leading edge of this density current is characterized by strong turbulent winds, suggesting giant dust particles mobilization over areas in the vicinity of the Nevado Illimani, where these particles are also deposited. In accrodance, GPPnb is negatively correlated with the DJF OLR centered over the Altiplano (Table 2), indicating that deep convection increases giant
- 375 particle entrainment and suspension, humidity and precipitation over the region. During dry seasons, conversely, GPPms is not significantly correlated with δD , as shown by seasonal mean values reported in Fig.5. Curiously, we found no significant relationships between GPPnb and wind speeds at the meteorological stations. This might be due to the short lifetimes of the density currents related to convection, observed to be in the order of a few hours (Knippertz et al., 2007). Considering our seasonal resolution analysis, we suggest that these high turbulence events had a low influence on the mean DJF wind speed.
- We conclude that the more intense is summer convection, the higher is the relative mass-number of giant dust particles suspended in the atmosphere and the more depleted is the δD . Interestingly, Fig. 5 Differently from the wet season, when the major control of δD variability in Zongo (and therefore probably in Illimani) is the progressive depletion of water vapor by unsaturated convective downdrafts, the δD variability of the winter responds mainly to the intense reevaporation processes that occur in a dry atmosphere, with a low convective activity (Vimeux et al., 2011). Although we observed a significant correlation
- 385 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).

Fig. 6 shows that over the 18-years period spanned by the Nevado Illimani firn core analyzed in this work, the summer seasons showing the most intense levels of convection were 1999–2000, (considering both GPPnb and δ D) were 2000–2001, 2005–2006, 2007–2008, 2010–2011 and 2011–2012. These all and 2010–2011. Both correspond to La Niña years, as it is also

390 clear from Fig. S2, where seasonal PC1 (proxy for convection) is compared to the periods, as indicated by their DJF Oceanic Niño Index . Some months after cooling of surface waters in the Pacific Ocean along the western coast of South America, we

observe intensification of convective activity over the Altiplano(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). Our data show that during summer seasons characterized by However, the strong DJF La Niña events , the atmospheric patterns typical of wet summer seasons are emphasized 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

- 405 a new approach for future studies aimed at the reconstruction of paleo-ENSO at high elevation glaciological sites in the Andes in tropical Andean glaciers based on giant particles and stable isotopes of snow, which. This can be used as a complement to a number of other climate proxies and modeling experiments, providing insights into past ENSO variability atmospheric circulation over tropical South America.
- 410 *Data availability.* Dust (concentration, grain size, geochemistry and mineralogy), stable water isotopes and calcium data can be made available for scientific purposes upon request to the authors (contact filipelindau@hotmail.com, jefferson.simoes@ufrgs.br or barbara.delmonte@unimib.it).

Author contributions. FGLL, JCS, BD, GB wrote the original manuscript. JCS and BD designed the research. PG designed and led the drilling campaign. FGLL and PG sampled the core. FGLL, BD, GB, CIP and EDS conducted dust analyses. EK and DSI carried out the ionic and the isotopic measurements, respectively. BD, EG, SA, GB and CIP advice on data collection and interpretation. VM, CIP, PG, EG and SA provided comments to the original manuscript. VM, EG and SA provided analytical resources.

Competing interests. The authors declare that they have no conflict of interest.

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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 white 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 Bolivia, Chile and Argentina indicate the salars from of the Altiplano and in the Punaregions, respectively (Gaiero et al., 2013). The land basemap was derived from satellite data (Natural Earth I with Shaded Relief from http://www.naturalearthdata.com).



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 concentration particles concentrations (light and dark gray, respectively, both are in logarithmic scale). Grey-Gray shaded areas-vertical bands correspond to the dry season for each calendar year. On (a) the percentage the percentage of giant particles with respect to the total dust mass (GPPms, reverse scale) is compared to the δD , and the first principal component of these two series (representing 83% of variance) is also reported. All data are reported as 3-point running average of data re-sampled at 0.05 m m w.eq.



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 δ D. 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 weg.



Figure 4. Changes in dust mineralogy between (a) the wet and (b) the dry seasons. The lower plots highlight the mineralogy of the phyllosilicates.



Figure 5. Geochemical signature of Nevado Illimani firn samples. (a) Relationship between the REE (normalized considering the UCC composition Rudnick and Gao2003) from the Nevado Illimani firn core (this work) and sediments/soils from potential dust sources (corresponding to $< 63 \mu m$ grain size for top soils). Data from Northern Puna, and Uyuni and Coipasa salars are from Gaiero et al. (2013). Data from Patagonia are from Gaiero et al. (2004). Data for the APVC refer to geological samples from Lindsay et al. (2001) and Ort et al. (1996). (b) Enrichment Factors (EF) for different elements and standard deviations. Samples with a high GPPnb (blue circles) show anomalous enrichment for Hf and to a lesser extent for Cs (see text).



Figure 6. Seasonal mean GPPms 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 brown gray dots on background are raw data. The La Niña years events discussed in the text (1999–00, 2000–01, 2007–08, and 2010–11) are reported and marked with a black star.

Sample	Year	Season	Dust (ppbpart. mL^{-1})	GPPms-GPPnb (%)
N1	2016	Dry	2790 - <u>14,737</u>	22.6 0.1
N2	2015 - 2016	Wet	590 -7,283	14.0 0.2
N3	2013 - 2014	Wet – Dry	1330.6,340	23.1 0.3
N4	2012	Dry	9050 -30,405	19.7 0.2
N5	2010 - 2011	Wet	830 -2,605	4 1.6 1.3
N6	2008	Dry	20060-25,816	18.4 0.7
N7	2007 - 2008	Wet – Dry	2430 <u>6</u>,424	38.7 1
N8	2003	Dry	10080-29,923	17.7 0.2
N9	2002	Dry	5350 -21,057	16.6 0.2
N10	2000 - 2001	Wet	680 -4, <u>180</u>	51.0 1.9
R1	2010	Dry	4740 - <u>17,409</u>	17.70<u>.1</u>
R2	2009 - 2010	Wet	940-8,234	26.00.2
R3	2004	Dry	7340 -86,918	19.40.2
R4	2003 - 2004	Wet	830 -2, <u>303</u>	38.4 0.9

Table 1. Characterization of the Nevado Illimani samples analyzed for elemental (N1 to N10) and mineralogical (R1 to R4) composition.

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 the 95% level are shown in bold.

GPPms GPPnb	δD	El Alto	Calacoto	Patacamaya	Oruro	Potosi	OLR
Wet	- 0.80 -0.71	0.49 0.47	0.35 <u>0.39</u>	0.540.76	0.26 <u>0.25</u>	0.32_0.41	- 0.48-0.69
Dry	0.03 -0.70	-0.43 <u>0.04</u>	-0.23_0.21	-0.62 -0.09	-0.49 0.08	-0.30 0.23	0.30 0.07

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

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Figure S1. The Nevado Illimani firn core density profile. The right panel refer to depth intervals where groups of ice layers were observed.

Table S1.	Sections of the	e Nevado Illima	ni firn core anal	lysed for elemental	(N1 to N10) a	nd mineralogica	l (R1 to R4) cor	nposition.
				2	\[0	\[

Sample	N1	N2	N3	N4	N5	N6	N7	N8	N9	N10	R1	R2	R3	R4
Top depth (m)	1.67	2.31	5.57	8.67	10.56	13.60	13.69	19.10	20.29	21.41	5.79	5.91	9.80	10.00
Bottom depth (m)	2.31	3.78	7.02	8.97	11.66	13.69	14.78	19.25	20.48	22.50	5.82	6.00	9.84	10.17

Table S2. Average elemental concentrations measured by Instrumental Neutron Activation Analysis, and the procedural errors and detection limits (DL). For comparison, we also present mean concentrations of dust particles from high elevation ice cores in the Alps (Thevenon et al., 2009) and in Tibetan plateau (Wu et al., 2009).

Element	Ce	Cs	Eu	Hf	La	Sc	Sm	Th	Yb
Concentration (ppm)	89.8	21.1	1.6	13.1	40.8	13.7	8.14	17.6	2.71
Error (%)	7.24	13.8	6.65	9.97	4.05	3.97	7.27	7.16	10.8
DL (ppm)	7	4	0.8	3	0.5	0.3	0.1	4	0.1
Colle Gnifetti ice core, Alps (ppm)	1.76	1.20	1.78	5.24	2.74	18.6	4.63	3.49	<u>6.25</u>
Dunde ice core, Tibetan Plateau (ppm)	<u>65.7</u>		1.09	4.04	32.6		5.51	14.3	2.93



Figure S2. Comparison of the seasonal PC1 The number size distribuition of CPPms and δD (black line) and a typical sample from the Oceanic Niño Index (ONI) (blue line) typically used for identifying El Niño (warma) dry and La Niña (coolb) events in the tropical Pacificwet season. It is possible to note that Red areas highlight the highest convection giant particles (low PC1between 20 and 60 µm) is associated to summer seasons of La Niña years.

	R1	R2	R3	R4
Year	2010	2009–10	2004–05	2004
Season	dry	wet	wet	dry
Particles analyzed	167	161	138	167
	N (%)	N (%)	N (%)	N (%)
Quartz and polymorphs	31.7	21.7	13.8	23.4
K-feldspars	7.2	10.6	5.1	11.4
Plagioclases	10.8	9.3	7.2	18.6
Muscovite-illite-smectite	23.4	29.2	59.4	19.2
Other phyllosilicates	0.6	1.2	0	1.2
Clay minerals (kaolinite)	0.6	1.2	0	3
Clinopyroxenes	0	0.6	0	0
Orthopyroxenes	0	0	0	0
Amphiboles	0	0	0	0
Rutile	2.4	2.5	0	0.6
Anatase	4.2	6.8	4.3	2.4
Hematite	9	8.7	7.2	14.4
Goethite	8.4	5.6	1.4	3
Carbonates	0	1.2	0	0
Epidotes	0	0	0	0
Titanite	0	0	0	0
Tourmaline	1.2	0	0.7	0.6
Other heavy minerals	0.6	1.2	0.7	1.2
Other light minerals	0	0	0	1.2
Total	100	100	100	100

Table S3. Mineral phases and relative abundance (number of grains, %) in Illimani firn from different time periods.

References

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