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1 **Glacier melting increases the solute concentrations of Himalayan glacial lakes**

2 Franco Salerno,^{*1,3} Michela Rogora,² Raffaella Balestrini,¹ Andrea Lami,^{2,3} Gabriele A. Tartari,² Sudeep Thakuri,¹
3 Danilo Godone,⁴ Michele Freppaz,⁵ and Gianni Tartari^{1,3}

4 ¹ CNR – Water Research Institute (IRSA), Via del Mulino 19, Brugherio (MB) 20861, Italy

5 ² CNR – Institute of Ecosystem Study (ISE), Largo Tonolli 50, Verbania Pallanza (VB) 28922, Italy

6 ³ Ev-K2-CNR Committee, Via San Bernardino, 145, Bergamo 24126, Italy

7 ⁴CNR IRPI - Geohazard Monitoring Group, Strada delle Cacce, 73, 10135 Torino, Italy

8 ⁵Università di Torino, DISAFA and NatRisk, Largo Paolo Braccini 2, 10095, Grugliasco (TO), Italy

9 *Correspondence to Franco Salerno (salerno@irsa.cnr.it) Water Research Institute – National Research Council (IRSA-
10 CNR), Via del Mulino, 19, 20861 Brugherio, MB, Italy TEL +39 039 21694221 FAX +39 039 2004692

11 **ABSTRACT** Over the past two decades, we observed a substantial rise in ionic content that was mainly determined by
12 the sulfate concentration at 20 remote high elevation lakes located in central southern Himalaya. At LCN9, which was
13 monitored on an annual basis for the last twenty years, the sulfate concentrations increased over 4-fold. Among the
14 main causes, we exclude a change in the composition of wet atmospheric deposition, as well as a possible influence of
15 decrease in seasonal snow cover duration, which could have exposed larger basin surfaces to alteration processes.
16 Glacier retreat likely was the main factor responsible for the observed increase of sulfate concentrations. We attribute
17 this chemical changes mainly to the sulfide oxidation processes that occur in subglacial environments. Moreover, we
18 observe that the weakened monsoon of the past two decades has only partially contributed to the lakes enrichment
19 through runoff waters that are more concentrated in solutes or lowering the water-table, resulting in more rock exposed
20 to air and enhanced mineral oxidation.

21 **INTRODUCTION**

22 High mountain ecosystems are especially vulnerable to climate change, and lakes in particular may act as sentinels.^{1,2}
23 An evaluable opportunity for a fine-scale investigation is particularly evident on the south slope of Mt. Everest (Nepal),
24 which is one of the most heavily glacierized parts of Himalaya³. At the same time, this region is the most densely
25 populated by glacial lakes in the overall Hindu-Kush-Himalaya range⁴. In this region, Lami et al.⁵ observed a significant
26 relationship between the increase in the annual temperature recorded in the area and the enhanced conductivity detected
27 in two glacial lakes located above 5000 m a.s.l. in the last decades, but without inferring any undergoing physical
28 process or source of ionic release. At the same time, an increase in conductivity and sulfate concentrations has been

observed in a few lakes of the European Alps, but in this case, the changes have been specifically ascribed to the solute release from an active rock glacier as a response to climate warming⁶. An increase in sulfate from melting ice during drought years is also reported by Williams et al.⁷ in the outflow of a rock glacier in the Colorado Front Range and by Mast et al.⁸ in southwestern Colorado. Even in this last case the authors consider temperature as the main climatic driver of change and sulfate as largely derived from pyrite (watershed weathering sources), although the undergoing physical processes are only supposed.

In this study, after exploiting the recently reconstructed⁹ daily temperature and precipitation time series and the available accurate tracing of the glacier shrinkage in the region¹⁰, we discuss changes in the water chemistry of 20 remote lakes located on the south slopes of central Himalaya that were sampled approximately after twenty years, starting in the early 1990s. Furthermore, during this period, two selected lakes (named Lake Cadastre Number (LCN)¹¹ 9 and 10) were monitored yearly. We consider several drivers of changes of the in-lake chemistry, including temperature, precipitation, atmospheric deposition, rocks and soil weathering processes (periglacial environment), seasonal snow cover duration, and, in particular, the role of glaciers (subglacial and surrounding periglacial components). With regard to permafrost, little is known about its distribution in Himalaya.¹² Therefore the subsurface ice and its possible degradation is not included in this analysis, although its influence on in-lake chemistry evolution cannot be excluded as reported by other authors^{13,14} in the Colorado Front Range.

MATERIAL AND METHODS

Study area. The monitored glacial lakes are located in Sagarmatha (Mt. Everest) National Park (SNP), Nepal, which lies in the central southern Himalayas (Fig. 1a, 1c). The SNP (1148 km²), crossed by the Khumbu valley, is the world's highest protected area, extending from an elevation of 2845 to 8848 m a.s.l..^{15,16} According to Searle et al.¹⁷, the Khumbu valley substratum is dominantly made up of crystalline rocks (e.g., sillimanite gneisses calc-silicates, amphibolites and K-feldspar augen gneisses) and leucogranites that intrude the metasediments as dykes, sills and bodies. The overlying Everest series (weakly metamorphosed shales and pelites with limestone bands), the Yellow band unit (limestones, marbles and calc-silicates) and the Ordovician limestones are only present in the eastern part of the basin. Extensive moraine debris of different ages, alluvial deposits and glacio-fluvial deposits host sub-surface aquifers in close connection with the hydrographic network.

The land cover classification shows that almost one-third of the territory is characterized by temperate debris-covered glaciers, with 75% of glacier surface area lying between 5000 m and 6500 m a.s.l..¹⁰ and with less than 10% of the park area being forested^{15,16}. The soils in the high valleys are primarily Entisols.¹⁸

58 Salerno et al.¹⁹ reported the presence of 170 glacial lakes and provided an accurate description of their topographic
59 characteristics. Most of them are small (median surface of 0.01 km²) and within an elevation zone ranging from 4800 to
60 5300 m a.s.l.. Table 1 shows the main morphometric features of the 20 survey lakes. They have been selected to cover
61 wide elevation (4466-5427 m a.s.l.) (Fig. 1b), basin slope (9°-35°), and basin aspect (120°-231°) ranges. The mean
62 glacial coverage is 18%, with three basins without glaciers (LCN40, LCN66, and LCN70). Two sub-basins (LCN10 and
63 LCN67) does not have glaciers within the sub-basin, which directly drain into the lakes, while in this case their
64 upstream lake basin (LCN9 and LCN68, respectively) presents a glacier coverage (all data are reported in Table SI1).
65 Details on these lakes and their basins can be found in Tartari et al.²⁰. All glaciers lying within the survey lake basins
66 are debris-free and without clearly detectable accumulation and ablation zones.

67 The climate is characterized by the monsoons, with a prevailing S-N direction.²¹ During the last twenty years at
68 the Pyramid meteorological station (5050 m a.s.l., Fig. 1c)⁹, which is located just 500 m from LCN9 (Fig. 1c), 90% of
69 the precipitation is concentrated during June-September, and the probability of snowfall during these months is very
70 low (4%), whereas the amount of snowfall at an annual level reaches 20%. Snow as a percentage of total precipitation
71 increases with the elevation gradient justifying the wide glacier coverage in SNP. Recently, the daily temperature and
72 precipitation time series of the last twenty years (1994-2013) were reconstructed by Salerno et al.⁹ for these altitudes
73 (Fig. 1b), which correspond to the median elevation of the 20 survey lakes. The total annual cumulated precipitation at
74 this elevation is 446 mm, with a mean annual temperature of -2.45 °C. The mean annual air temperature has increased
75 by 0.9 °C since the early 1990s. The significant increases were experienced just for the months of April, November, and
76 December.⁹ As the air temperature increased, the average elevation of the freezing line (0 °C isotherm) has moved
77 upward. The major changes occurred in April, where it moved upward by 225 m, passing from around the mean
78 elevation of glaciers terminuses to the mean elevation of the surfaces of the same glaciers located in the basins of the 20
79 survey lakes (Fig. 1c). The increase in air temperature observed at these high elevations during April fits with the
80 warming reported by Pal and Al-Tabbaa²², who observed that only April shows significant changes in all Indian regions
81 and the West Himalaya (1901–2003 period). As for precipitation, a substantial reduction of rainfall (47%) and in the
82 probability of snowfall (-10%) has been observed in this area over the last twenty years⁹. . According to Yao et al.²³
83 there is strong evidence of the general monsoon weakening even in the overall Himalayas.

84 **Water chemistry analysis.** The chemical composition of 20 remote lakes located in SNP was evaluated in the period
85 between 1990 and 1997 (hereafter early 1990s) and successively from 2008 to 2012 (hereafter early 2010s). Lake
86 sampling was carried out in autumn, at the end of the monsoon season. Samples were collected from the lake shore,
87 possibly close to the lake outlet. Moreover, for the lakes LCN 9 and LCN10, sampling and chemical analyses have been
88 performed annually since 1990 (1991 for LCN9), usually in late September or October, with the exception of the

89 missing years 1995-96 and 1998. For these lakes, three samples were generally collected along the water column from
90 an inflatable boat in the deepest part of the lake. Here we took their average. For detailed information on the
91 hydrochemical and biological characteristics of these lakes, see the work of Lami et al.⁶.

92 All samples were collected using prewashed plastic bottles and stored at 4 °C for successive chemical analysis; these
93 were performed during the 1990s by the CNR Water Research Institute and subsequently by the CNR Institute of
94 Ecosystem Study (Italy). Several intercomparisons have been conducted to ensure data quality and comparability
95 between the data produced by the two laboratories. Samples were analyzed for pH, conductivity, total alkalinity (using
96 the Gran method for acidimetric titration), ammonium, total nitrogen, total phosphorus and reactive silica (by
97 spectrophotometry), sulfate, nitrate, chloride, calcium, magnesium, sodium and potassium (by ion chromatography).
98 Data were checked for internal consistency by means of the ionic balance and the comparison between measured and
99 calculated conductivity. Details on the analytical methods and on the quality controls adopted in the laboratory can be
100 found in Lami et al.⁵.

101 In this paper, we focus on conductivity values, as an index of total solute concentrations of lakes, H^+ , and major ions:
102 calcium + magnesium ($Ca^{2+}+Mg^{2+}$), sulfate (SO_4^{2-}) and bicarbonate (HCO_3^-). These are indeed the prevailing ions in all
103 sampled lakes, representing more than 80% of the total ionic content (Fig. SI1). We assume alkalinity as corresponding
104 to bicarbonate ions, because at circum-neutral pH, as those of the study lakes, dissolved HCO_3^- is the dominant species
105 contributing to alkalinity.

106 **Morphometric and hydrological analysis.** Seasonal snow cover duration of each survey lake basin has been attributed
107 as a mean for the 2002-2012 period of the daily MODIS (Moderate Resolution Imaging Spectroradiometer) imagery
108 (pixel 250 m) (<http://modis.gsfc.nasa.gov/>). The glacier surface areas within the survey lake basins were derived for the
109 early 1990s from Landsat 5 TM (17 November 1992, pixel 30 m) and for the early 2010s from Landsat 11 ETM (30
110 November 2011, pixel 15 m) remote imagery. Data and methods are detailed in Thakuri et al.¹⁰. We interpreted the land
111 cover of the remaining part of each basin through a visual interpretation of the Advanced Land Observation Satellite
112 (ALOS, October 2008, pixel 10 m) remote imagery and distinguished terrain coverage by bare rocks, bare soils, and
113 shrub vegetation. No rock glaciers were detected in these basins by visual interpretation.

114 Because we were interested in understanding whether the changes that occurred over the last twenty years in glacier
115 surface areas and snow cover have influenced the solute variations observed during the same period, we had to
116 overcome, as shown below, such limits linked with the resolution and availability of the satellite data.

117 Regarding glaciers, the satellite resolution of remote imagery available for covering the analyzed period provides a
118 large uncertainty in estimating changes in surface areas, especially for small glaciers, such as those located within the
119 basins of the survey lakes; thus, the estimated changes are dubious¹⁹. This limit has been overcome by enlarging the

120 scale of analysis to the overall SNP. At this scale, the 29 glaciers (with a surface area $>1 \text{ km}^2$) that were already traced
121 by Thakuri et al.¹⁰ are more than 30 times larger ($5.7 \pm 0.40 \text{ km}^2$ as median) than those located within the survey lakes
122 ($0.31 \pm 0.10 \text{ km}^2$ as median); thus, changes are more representative and less uncertain¹⁹.

123 Concerning the snow cover, the MODIS imagery is available only since the early 2010s. For the previous decade,
124 we deduced the trend in the coverage based on the estimated snowfall decrease reported in this area since the early
125 1990s⁹. The increase of temperature observed outside the monsoon period, when the precipitation is almost completely
126 composed by snow, brought a significant decrease (-14% , $p < 0.05$) of the snowfall probability for the last twenty years
127 at 5050 m a.s.l.. The trend reported above has been calculated by these authors according to Fujita and Sakai²⁴ and
128 Ueno et al.²⁵ assuming that the probability of snowfall and rainfall depends on mean daily air temperature, using as
129 thresholds 0 and 4 °C, respectively. The variations that occurred in the 2002-2012 period were computed as the
130 difference between the first five and the last five years. Even in this case, the analysis was enlarged to the overall SNP
131 to increase the representativeness within an elevation ranging from 4500 to 5500 m a.s.l. (6152 MODIS pixels), i.e., the
132 altitudinal band of the survey lakes (Fig. 1b).

133 At this wide scale, we determined that changes in glacier surface areas and seasonal snow cover duration are related
134 to certain morphometric boundary conditions. Elevation, aspect, and slope have been investigated for snow cover
135 duration and glaciers, but for glaciers, we also considered the maximum, minimum and mean elevation, and the down
136 and upstream slopes of the glaciers following the experience of Thakuri et al.¹⁰ and Salerno et al.¹⁹. Detailed
137 computational methods of these morphometric variables are reported in the works of these authors. In the same way, at
138 the scale of each survey lake basin, we determined that the differences that occurred in SO_4^{2-} and HCO_3^- are related to
139 the same morphometric boundary conditions investigated for glaciers and snow cover duration at a wider scale.

140 For simulating the daily melting of the glacier located in the LCN9 basin we used a T-index model²⁶. This model
141 uses the mean daily air temperature, the glacier elevation bands, and a melt factor ($0.0087 \text{ m d}^{-1} \text{ }^\circ\text{C}^{-1}$) provided by
142 Kayastha et al.²⁷ from a field study (Glacier AX010) located close to the SNP.

143 **Statistical analysis.** We conducted a Principal Component Analysis (PCA) among the lake chemical components and
144 the basin morphometric features to obtain information on the relationships among the data and to look for reasons that
145 could justify the observed changes in major ion concentrations²⁸. The test was performed by using the “princomp” and
146 the “biplot” functions in the R Project environment²⁹. The degree of correlation among data was verified through the
147 correlation coefficient (r) after testing that the quantile-quantile plot of model residuals follows a normal distribution.
148 Otherwise data were log-transformed to meet the statistical requirements for normal distribution; then the residuals of
149 the regressions were tested for homoscedasticity (not shown here) (e.g.,²⁹). The chemical concentrations of the 20
150 survey lakes, measured in the early 1990s and the early 2010s, were compared, and the differences between the two

151 periods were tested by using the non-parametric ANOVA for paired comparisons (Friedman test) at $p < 0.05^{28,30}$.
152 Differences in glacier surface areas and snow cover duration were analyzed by applying the same statistical test. The
153 significance of annual chemical trends was evaluated with the annual Mann Kendall test at $p < 0.05$.³¹

154 RESULTS AND DISCUSSION

155 **Survey lakes.** In the early 1990s, most of the survey lakes exhibited low values of conductivity ($25 \mu\text{S cm}^{-1}$ as
156 median) (Table 2, Fig. 2a). After approximately 20 years, the conductivity increased significantly, by 47% ($p < 0.001$) on
157 average. In some cases, the values even doubled. Lakes became significantly ($p < 0.001$) enriched in solutes, primarily in
158 SO_4^{2-} (+57%), then in $\text{Ca}^{2+} + \text{Mg}^{2+}$ (+41%) and less in HCO_3^- (+11%) (Table 2, Fig. 2b,c,d). The increase in SO_4^{2-} was
159 even 4-fold in LCN9 (from 84 to $327 \mu\text{eq L}^{-1}$) and LCN10 (from 73 to $285 \mu\text{eq L}^{-1}$) (Fig. 2b). For these lakes, a
160 significant increasing annual trend ($p < 0.05$) was observed both for conductivity, SO_4^{2-} , $\text{Ca}^{2+} + \text{Mg}^{2+}$, and HCO_3^-
161 concentrations (Fig. 3b,c,d), with two main peaks in 1998 and 2010 (discussed in the last paragraph). All survey lakes
162 (Table 2 and Fig. 2e), as well as LCN9 and LCN10 (Fig. 3e), exhibited a significant increase of hydrogen ion (H^+)
163 concentrations (126%, $p < 0.01$).

164 The temporal change of the relative contribution of the various ions to the total ionic content of LCN 9 can be
165 observed in Figure SI1: sulfate passed from 22 to 36%, calcium remained stable (41%), while alkalinity contribution
166 decreased from 24 to 11%. To assess the relative change in the ionic composition of the survey lakes, we calculated the
167 c-ratio $\text{HCO}_3^- / (\text{HCO}_3^- + \text{SO}_4^{2-})$, which is a geochemical indicator that is used to evaluate the relative dominance of the
168 carbonation reaction and sulfide oxidation in meltwaters.³² The c-ratio decreased significantly in the survey lakes (-
169 18%, $p < 0.01$) (Table 2, Fig. 2f). Particularly pronounced is the c-ratio decline observed in LCN9, from 0.56 to 0.29 (-
170 48%), and in LCN10, from 0.59 to 0.31 (-47%) (Fig. 3f). The only exception with respect to the general pattern is
171 LCN31 (Fig. 2f). The Duwo glacier, which is located within the LCN31 basin, is surging and shows a divergent
172 response to climate change with respect to all of the other glaciers located in the region.¹⁰ Therefore, we think that the
173 two anomalies could be associated. All data presented in this section are reported in Table SI1.

174 In the following, we discuss potential factors that may have contributed to the increase of solute concentrations in
175 the lakes, with special attention towards SO_4^{2-} and HCO_3^- ions: atmospheric deposition, weathering, precipitation,
176 glaciation, and snow cover duration.

177 **Atmospheric Deposition.** The chemistry of wet deposition was investigated at the Pyramid station during 1991-1992³³
178 and 2007-2008³⁴. The precipitation contents for all ions were the lowest ones among those measured at high elevation
179 sites around the world³². The comparison between the studied periods revealed no substantial variations in SO_4^{2-} and
180 other compounds (NO_3^- and NH_4^+) that resulted from anthropic activities, thus suggesting that the tropospheric

181 background concentrations are constant in the Himalayas³⁴. These findings indicate that the atmospheric deposition
182 cannot play significant role in the solute increasing trend recorded in Himalayan lakes.

183 **Land cover and Snow cover duration.** We tested the possible dependence of SO_4^{2-} and HCO_3^- concentrations for the
184 survey lakes sampled in 1990s with respect to the basin land cover and seasonal snow cover duration. We preferred to
185 use 1990s data in order to investigate the dependences between these variables before a period (1990s-2010s)
186 influenced decidedly more by higher climate-driven changes⁹. The correlation matrix among all of these variables is
187 presented in Fig. SI3, and an overall overview of their mutual relationships is provided with the PCA in Fig. 4a. The
188 main finding is that the SO_4^{2-} concentrations are directly related to glacier coverage ($r=0.72$, $p<0.001$) (Fig. 4b), i.e.,
189 higher lake solute concentrations are found for basins with more glacial coverage. No relationship can be observed with
190 the other selected variables and in particular with the snow cover duration. Furthermore, we note that the HCO_3^-
191 concentrations do not have significant relationship with the land cover and the snow cover duration.

192 When observing the morphometric features of the LCN9 basin and the LCN10 sub-basin (Table 1), it becomes clear
193 that the discriminating feature between these lakes is the presence of a small glacier of 0.11 km² lying within the LCN9
194 basin, whereas the LCN10, which is located downstream, receives the glacial melting indirectly from LCN9 (Graphical
195 abstract). Concurrently, LCN10 has lower solute concentrations and conductivity (Fig. 3a, 3b, 3c, 3d). This fact can be
196 interpreted as further evidence that glaciers (subglacial and surrounding periglacial components) are the most important
197 driver of changes in both SO_4^{2-} concentrations and that the runoff in the non glaciated areas of the LCN10 sub-basin as
198 well as the possible groundwater discharge from headwater areas of the basin transports lower concentrations, thereby
199 diluting the LCN10 content. This interpretation is corroborated by the further evidence provided by lakes LCN68 and
200 LCN67. From Table SI1 we observe that the downstream lake (LCN67) has, among the other solutes, lower sulfate
201 concentrations (733 $\mu\text{eq L}^{-1}$ respect to 926 $\mu\text{eq L}^{-1}$ for LCN68). Even in this case the discriminating feature is the
202 presence of a glacier lying within the upstream lake basin (see Fig. SI2 and Salerno et al.¹⁹- Fig. SI2d-).

203 **Changes in glacier surface areas.** Thakuri et al.¹⁰, considering glaciers larger than 1 km², reported a surface area loss
204 of -8.7%, $p<0.01$ for the overall SNP during the 1992-2011 period (uncertainty $\pm 2.8\%$). Applying here the same remote
205 imagery and methods reported by these authors, but at glaciers located within the basins of the 20 survey lakes
206 (0.31 ± 0.10 km² as median), we found a higher decrease in glacier area, but with an uncertainty connected with the
207 satellite resolution that increased by of an order of magnitude (-19.7%, $p<0.01$, uncertainty $\pm 23.9\%$). The glacier within
208 the LCN9 basin lost 14% (uncertainty $\pm 22\%$) of its surface area. According to many authors^{10,35,36,37}, even in SNP and
209 in the Himalayas in general, the main losses in area over the last decades have been observed for smaller glaciers
210 (glacier size vs glacier area change $r=0.58$, $p<0.01$, Fig. SI4). Therefore, the survey lake basins are subject to a glacier

211 shrinkage twice that observed for the glacial masses in the SNP region. However, the reduced size of these glaciers
212 causes the area loss estimation to be affected by a large uncertainty that is even higher than its magnitude, and the
213 estimation is thus too rough to be directly compared with the chemical variations observed in the relevant downstream
214 lakes. Therefore, as described above, we conducted two parallel analyses: at the SNP level and at the scale of a survey
215 lake basin.

216 At a larger scale, considering the data from Thakuri et al.¹⁰, we analyzed which morphometric factors were able to
217 cause changes in surface areas in the last twenty years (Fig. 5a). The correlation matrix among all of these variables is
218 presented in Fig. SI4. From the PCA analysis shown in Fig. 5a, we can see that the downstream slope ($r=-0.66$,
219 $p<0.001$) (Fig. 5c) and the relevant aspect ($r=-0.75$, $p<0.001$) of glaciers are the main factors responsible for the surface
220 area losses, whereas elevation seems to have less influence. These findings agree with the few other studies
221 investigating these relationships^{19,35,36,37}. Whereas elevation is a proxy for temperature^{35,37}, aspect is a proxy for
222 insolation³⁸, and slope is the key factor responsible for the gravitational driving stresses to which glaciers are subjected³.
223 Lower slopes are supposed to induce lower glacier ice flow, thus allowing the development of stagnant ice conditions
224 and consequently lower terminus retreat rates^{3,39,40} and the development of supraglacial lakes^{19,41}. In contrast, as we
225 found here, under the climate warming pressure observed in the region for the last decades⁸, higher slopes favor the
226 shrinkage of glaciers because they are supposed to contribute to the higher glacier velocity⁴².

227 Moreover, we observed that even glacier aspect is indirectly correlated with surface area losses, i.e., the largest
228 shrinkages have been experienced mainly for E and W facing glaciers, and the shrinkages are decidedly smaller for S
229 facing ones (Fig. 5a). Considering that S oriented glaciers are supposed to receive more solar radiation³⁸, the observed
230 spatial shrinkage seems to be counterintuitive. However, we have to consider that, as their aspects deviates from S, the
231 glaciers become significantly smaller (glacier aspect vs glacier size $r=-0.61$, $p<0.001$) and steeper (glacier aspect vs
232 glacier downstream slope $r=0.52$, $p<0.01$) (all correlations are shown in Fig. SI4) because they are located in valleys
233 perpendicular to the prevailing S-N direction of the monsoon. In our opinion, these valleys have been less hollowed,
234 and the current glaciers are thus small and steep. Therefore, the observed correlation between aspect and glacier surface
235 area losses indicates that gravitational stresses (conditioned by slopes) are higher for aspects far from S, and these, as
236 observed above, are key factors in defining the rate of retreat of glaciers under the climatic pressure.

237 Although affected by a wide uncertainty, even at the scale of 20 survey lake basins we analyzed which
238 morphometric factors were able to cause changes in surface areas. The correlation matrix among all of these variables is
239 presented in Fig. SI4. We can see that the glacier slope ($r=-0.58$, $p<0.05$) and the relevant aspect ($r=-0.49$, $p<0.05$) of
240 glaciers are the main factors responsible for the surface area losses, though, for the effect of larger uncertainty, these
241 correlations are weaker than what we found at large scale.

242 **Changes in snow cover duration.** During the 2002-2012 period, we observed a decrease of the seasonal snow cover
243 duration of -29% ($p<0.01$) within an elevation ranging from 4500 to 5500 m a.s.l. A similar behavior (even if we focus
244 on the analysis exclusively) was found within the 20 survey lake basins (-27%, $p<0.01$). We found that the largest
245 differences in snow cover duration occurred with W and S exposure and that the smallest occurred in the opposite
246 orientations. Aspect ($r=0.23$, $p<0.001$) and elevation ($r=0.24$, $p<0.001$) are the main factors responsible for changes in
247 snow cover duration, and we did not find any strong relationship with slope (all correlations are shown in Fig. SI5). In
248 contrast to what we observed for glaciers, the snow cover duration at lower elevations decreased more due to higher
249 temperatures that were recordable downstream, and the snow cover duration on S and W slopes decreased more than on
250 N and E slopes, likely as a result of the higher solar irradiance received by these sides⁴³. In the next paragraph changes
251 observed in the seasonal snow cover duration are related to changes occurred in SO_4^{2-} and HCO_3^- concentrations of 20
252 survey lakes.

253 **Changes in ionic concentrations.** Once it is defined how variations recorded in glacier and snow cover duration have
254 been conditioned by morphometric factors, we analyzed which of these factors are mainly responsible for changes
255 observed in SO_4^{2-} and HCO_3^- concentrations in the survey lakes (Fig. 5b). Concerning the HCO_3^- concentrations, we did
256 not find any relationship with the selected morphometric boundary conditions (Fig. 5b; Fig. SI6). Differently, Figure 5b
257 reveals that glacier slope ($r=0.81$, $p<0.001$) (Fig. 5d) and relevant aspect ($r=0.66$, $p<0.01$) are well correlated with SO_4^{2-}
258 concentration changes, whereas the direct relationship with the glacier surface areas losses is significant, but weaker
259 ($r=-0.49$, $p<0.05$), probably affected by the high uncertainty connected with the measurements of small glaciers (all
260 correlations are shown in Fig. SI6). From Figure SI6 we can also observe that the SO_4^{2-} concentration changes are not
261 correlated with the basin slopes ($r=0.18$, $p>0.10$), as well as the direct relationship between glaciers slopes and basins
262 slopes is weak ($r=0.44$, $p<0.05$) enforcing the causal link between the SO_4^{2-} variations and the glacier slopes. As
263 indicated above, the glacier slope is also the main morphological feature able to explain the changes of the glacier
264 surface area at higher and more representative scale (Fig. 5a and Fig. 5b). Therefore, we can assert that lake basins with
265 steep glaciers oriented far from S have experienced higher glacier surface area losses and a concomitant SO_4^{2-} increases.
266 Differently, the main decreases in snow cover duration occurred at lower elevations and on land with a S orientation,
267 and they are less strictly dependent on the terrain slope (Fig. SI5 and Fig. SI6). Therefore, the changes observed in snow
268 cover duration seem not to be the main cause of the observed SO_4^{2-} increases.

269 LCN9 experienced a particularly evident increase in ionic concentrations, which was even 4-fold in the case of SO_4^{2-}
270 concentration (Fig. 3a). In this case, the reason for this huge change can also be found in that the LCN9 glacier has the
271 most divergent aspect (SE-E, 110°) from the south orientation and has the highest glacier slope (36°) among the 20
272 survey lakes (Table1). Moreover, a synchronous temporal pattern of solutes and conductivity can be observed in LCN9

273 and LCN10 (Fig. 3a, 3b, 3c): the increases for both lakes should be driven by the same source, i.e., the glacier located
274 within the LCN9 basin.

275 To further investigate the processes, which could have led to the SO_4^{2-} increase, we analyzed the associations
276 between the main ions and how they changed during the last twenty years. We observed a highly significant positive
277 relationship between SO_4^{2-} and $\text{Ca}^{2+} + \text{Mg}^{2+}$ in both in the early 1990s and in the early 2010s ($r = 0.85$ and 0.94 ,
278 $p < 0.001$) (Fig. SI3). On the contrary, the relationship between HCO_3^- and $\text{Ca}^{2+} + \text{Mg}^{2+}$ is weakly significant in the early
279 1990s ($r = 0.39$, $p < 0.05$) (Fig. SI3) and not significant in the early 2010s. The association between SO_4^{2-} and HCO_3^- was
280 quite low in both periods ($r = 0.45$, $p = 0.04$ and “not significant” in 2010s). The ratios of $(\text{Ca}^{2+} + \text{Mg}^{2+})/(\text{Na}^+ + \text{K}^+)$ was
281 7.2 in the early 1990s and 8.7 in the early 2010s, indicating the relative weakness of silicate dissolution. This is
282 confirmed also by the lack of correlation between Si and $\text{Na}^+ + \text{K}^+$ in the 2010s ($r = 0.17$). These results make evident an
283 excess of SO_4^{2-} with respect to HCO_3^- in waters flowing in the drainage system of glaciers. This shift towards SO_4^{2-} -
284 enriched waters is even more evident in LCN9 and LCN10, as shown by the comparison of the ionic composition of
285 lakes water in the 1990s and in the 2010s (Table SI1).

286 It is known that subglacial environments are dominated by carbonate dissolution and sulfide oxidation⁴⁴, often
287 microbially mediated^{45,46,47}. Through these processes, carbonate and sulfate concentrations in melt waters are greater
288 than might be expected from the bedrock lithology.^{48,49,50,51} A number of studies demonstrated that carbonation was
289 mainly responsible of solute acquisition in the quick-flow component of the glacier hydrological system, while the
290 enrichment in SO_4^{2-} seems to be strictly associated to delayed flow waters.⁵² For these reasons, sulfate is considered an
291 indicator of distributed type drainage beneath glaciers⁵³ and its increase, in most glacial environments, is an effect of
292 increasing residence times and rock/water interaction⁵⁴. In general, the oxidation of sulfide minerals and the dissolution
293 of carbonates are partially coupled, since the hydrogen ions, originated from the sulfide oxidations, fuel the carbonate
294 hydrolysis⁵³. Our results, showing the lack of any relationship between SO_4^{2-} and HCO_3^- , suggest that the two processes
295 are uncoupled in our system i.e. protons generated by sulfide oxidation do not dissolve carbonates. Furthermore, for
296 most lakes the c-ratio was at values lower than 0.5 in 2010s and, particularly for LCN9 and LCN10, reached 0.2 in
297 some years. These results suggest that other processes, likely in addition to carbonate dissolution and sulfide oxidation,
298 occurred in the selected lake basins.

299 A process consistent with the elevated SO_4^{2-} , Ca^{2+} and Mg^{2+} concentrations found recently in lake waters is the
300 pyrite oxidation coupled to the preferential weathering of calcium-silicate minerals which presence is documented in the
301 study area. As suggested by Williams et al.⁷ the Si produced from the weathering of calcium-silicate would be
302 precipitated due to the effect of low temperature and this could explain the lack of correlation between SO_4^{2-} and Si, and
303 the slight Si increase observed in the studied period.

304 The deglaciation observed in the study area has, first of all, increased the amount of meltwater causing a rising of
305 conductivity of lake waters. The disproportional increase of SO_4^{2-} compared to HCO_3^- may be explained by i) a change
306 in flow-paths and residence time of the meltwaters, leading to more favorable conditions for the oxidation of sulfides,
307 and ii) the pyrite oxidation followed by the weathering of calcium-silicate. We cannot exclude a role of microbial
308 populations at glacier beds if the conditions had become anoxic.^{45,46,47} Figure SI7 shows the significant correlation
309 between H^+ differences and SO_4^{2-} differences ($r=0.72$, $p<0.001$) observed for the studied lakes, which can be interpreted
310 as further evidence of the importance of sulfide oxidation connected with the glacier retreat. At this regards it is
311 interesting to observe that Andersen⁵⁵ wrote that sulfide oxidation contributes to solute fluxes from glaciers to an extent
312 greater than might be expected from mineral abundance.

313 **Weathering and precipitation.**

314 Regarding the weathering occurring on the non glaciated part of the basin (periglacial environment), we analyzed
315 the behavior of the lakes (three) without glaciers within their basins. They showed a slight increase in SO_4^{2-}
316 concentration (+17% as median) (Fig. 2b), whereas higher increases were observed for lakes with glaciers within their
317 basins (+78% as median). These differences between the two groups were significant ($p<0.01$). The slight increase in
318 SO_4^{2-} concentration of lakes without glaciers could be attributed to the intensification of weathering rates caused by the
319 reduction of the snow cover duration⁵⁶, although we already discussed that this process is not the main driver of change.
320 Particularly, during the warmer season, on the sunny slopes, previously snow covered, an increase in soil temperature
321 from 0°C to 25°C could be expected, which could intensify the weathering rates by an order of magnitude.⁵⁷
322 Weathering could be further enhanced by wetter reactive surface areas of minerals⁵⁸, but the decreasing precipitation
323 trend, as observed above over the last twenty years, does not support this possibility.

324 Nonetheless, the decreasing precipitation trend could have influenced the solute concentration in a different way:
325 lower but more concentrated runoff, could have slightly enriched the lakes⁵⁹. These solute variations were caused by
326 enhanced weathering rates, or more probably by more concentrated runoff. Another mechanism considers that the
327 possible dropping of water-table depth, due to decreased groundwater recharge, resulting in more rock exposed to air
328 and enhanced mineral oxidation.⁶⁰ However they can be considered background increases that are common to all survey
329 lakes with respect to the higher increases observed in conjunction with glacier shrinkages.

330 **Annual chemical trends for two selected lakes.** Sulfate was the ion showing the most pronounced change in LCN9
331 and LCN10 (Fig. SI1, Table SI1). Hence we tested the possible dependence of the SO_4^{2-} concentrations in LCN9 on
332 monthly mean temperature, precipitation, and glacier melting, estimated by applying the T-index model²⁶. We found
333 that SO_4^{2-} concentrations are only significantly correlated with the mean temperature in April ($r=0.74$, $p<0.001$). The

334 temperature and precipitation of the other months did not show any significant influence on the observed SO_4^{2-}
335 concentrations (Fig. SI8). As for estimated glacier melt, we found that the only month that was significantly correlated,
336 even in this case, is April ($r=0.74$, $p<0.001$) (Fig. 6a, 6b), whereas March shows a weak correlation ($r=0.45$, $p<0.1$)
337 (Fig. 6a).

338 As shown above, significant increases in mean air temperature have been experienced over the last twenty years for
339 the months of April, November and December.⁹ However, April is the only month in which the increase has been
340 effective, i.e., it has *de facto* favored the melting process. More precisely, the 0 °C isotherm in April shifted from
341 around the mean elevation of glacier fronts to the mean elevation of glaciers located in the survey lake basins (Fig. 1b).
342 In contrast, during the winter months, the temperature is so low that the 0 °C isotherm upward cannot have reached
343 these elevations, and it cannot have had an effective impact on the melting processes. As a result, the T-index model
344 estimates a significant increasing trend only for April ($p<0.01$). At this regards, the two main peaks of conductivity and
345 major ion concentrations in 1998 and 2010 observed in Figure 3 are caused by the high temperature registered for the
346 month of April of those years with subsequent high melting processes as estimated by the T-index model.

347 Previously, Lami et al.⁵ observed a significant relationship between the enhanced conductivity detected in LCN9
348 and LCN10 and the annual temperature until 2008. Here we confirm the observations of these authors until 2013
349 considering the temperature as the main climatic driver of change. Besides that, we found the temperature of April is the
350 effective drive of the observed enhanced glacier melting process, which is considered here the main factor responsible
351 for the observed increase of sulfate concentrations.

352 The chemical variations observed in lake water in the last two decades represent a response of these fragile
353 ecosystems to climate change. In particular, glacier retreat proved to be the main driver of the solute increase affecting
354 the lakes. Major ion concentrations, and sulfate at a higher extent, significantly rose in lake water. Even if these changes
355 do not pose a direct and immediate threat to the biota, they occurred in a limited time span, and significantly modified
356 the average chemical composition of lake water. For these reasons, the lakes and the main factors driving their
357 variability should be regularly monitored in the future, also in relation to the lake role as ecosystem services.

358 SUPPORTING INFORMATION

359 A table showing the chemical and topographic features of 20 survey lakes. A figure showing the relative contribution of
360 various ions to the total ionic content of LCN 9 in the early 1990s and in the early 2010s. ALOS 2008 imagery of
361 LCN68 and LCN67 basins. Correlation matrixes among chemical species, land covers, and topographic boundary
362 conditions. A figure showing the relationship between the SO_4^{2-} and the H^+ concentration changes observed in all

363 survey lakes between 1990s and 2010s. A figure showing the PCAs among SO_4^{2-} and HCO_3^- concentrations of LCN9,
364 mean temperature, and precipitation for each month of the year.

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TABLES

TABLE 1. Mean morphometric features and land cover for the 20 selected lakes (reference year 1992). For LCN 10
data are referred to the sub-basin draining directly into the lake (see graphical abstract).

Morphometry	20 survey lakes Median (range)	LCN9	LCN10
Lake elevation (m a.s.l.)	5010(4466-5427)	5209	5058
Lake area (km ²)	0.02(0.003-0.581)	0.005	0.013
Basin area (km ²)	0.8(0.1-41)	0.66	0.32
Basin slope (°)	29(9-35)	35	25
Basin aspect (°)	160(120-231)	120	120
Basin mean elevation (m a.s.l.)	5204(4843-5539)	5403	5102
Glacier area (km ²)	0.31(0.03-6.4)	0.11	-
Glacier slope (°)	26(15-36)	36	-
Glacier aspect (°)	151(110-221)	110	-
Glacier mean elevation (m a.s.l.)	5326(5087-5581)	5463	-
Land cover	20 survey lakes Mean (range)	LCN9	LCN10
Glacier (%)	18(0-50)	17	0
Bare rock (%)	28(0-66)	30	25
Bare soil (%)	40(9-74)	53	75
Shrub vegetation (%)	14(0-63)	0	0

TABLE 2. Median values of conductivity, selected ionic concentrations, and c-ratio (see the text for the explanation) of
the survey lakes sampled during the early 1990s and during the early 2010s. In brackets the standard deviation.

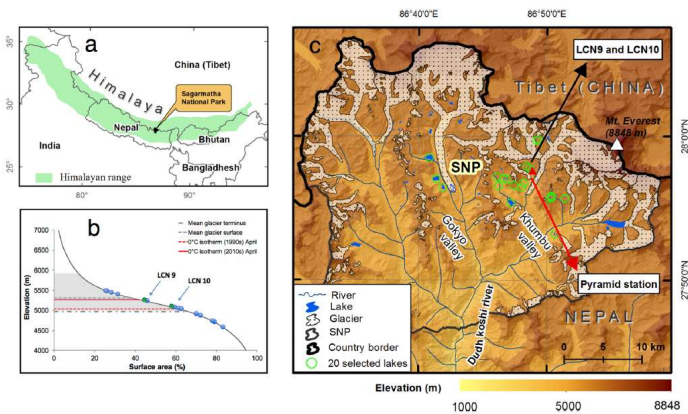
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Feature	Early 1990s	Early 2010s	Difference (%)
Conductivity ($\mu\text{S cm}^{-1}$, 20 °C)	24.7 (16)	36.4 (27)	+47 (p<0.001)
SO ₄ ²⁻ ($\mu\text{eq L}^{-1}$)	80 (129)	126 (245)	+57 (p<0.001)
HCO ₃ ⁻ ($\mu\text{eq L}^{-1}$)	128 (54)	141 (61)	+11 (p<0.001)
Ca ²⁺ +Mg ²⁺ ($\mu\text{eq L}^{-1}$)	221 (124)	331 (224)	+41 (p<0.001)
H ⁺ ($\mu\text{eq L}^{-1}$)	0.03 (0.02)	0.07 (0.02)	+ 126% (p<0.01)
c-ratio	0.66 (0.22)	0.55 (0.26)	-18 (p<0.01)

509

510 **FIGURE**

511 **FIGURE 1.** a) Location of the study area in the Himalayas. b) Hypsometric curve of SNP and altitudinal glacier
512 distribution. Along this curve, the location of 20 selected lakes is shown. The 0 °C isotherms corresponding to the mean
513 monthly temperature in April are plotted for the years 1994 and 2013 according to the observed T trends and lapse
514 rates⁹. c) Focused map on the spatial distribution of lakes and glaciers in Sagarmatha National Park (SNP) and the
515 location of the Pyramid meteorological station.



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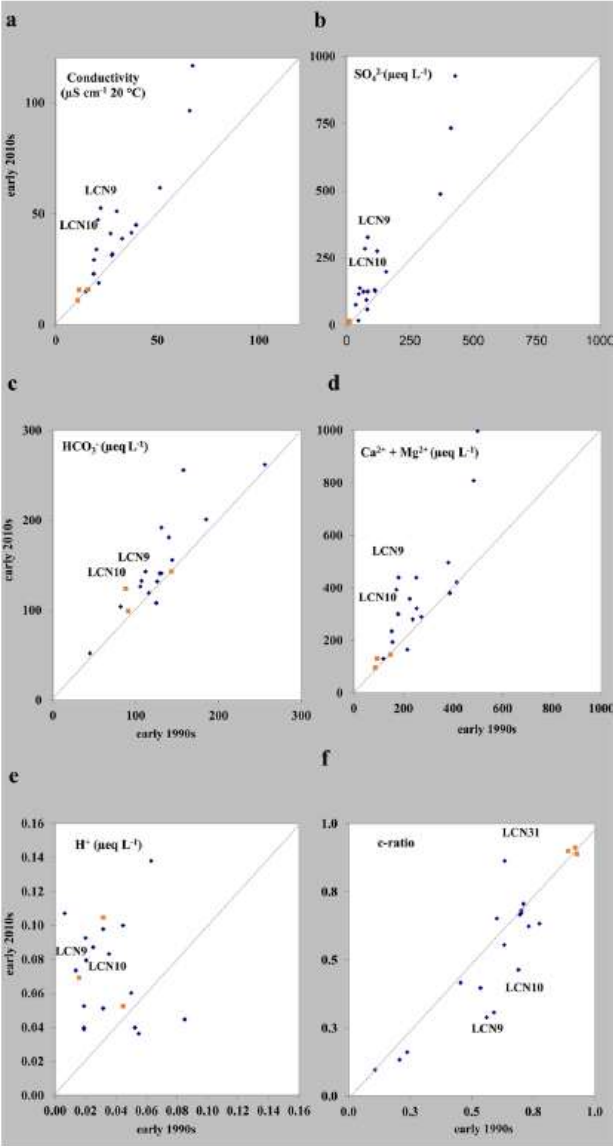
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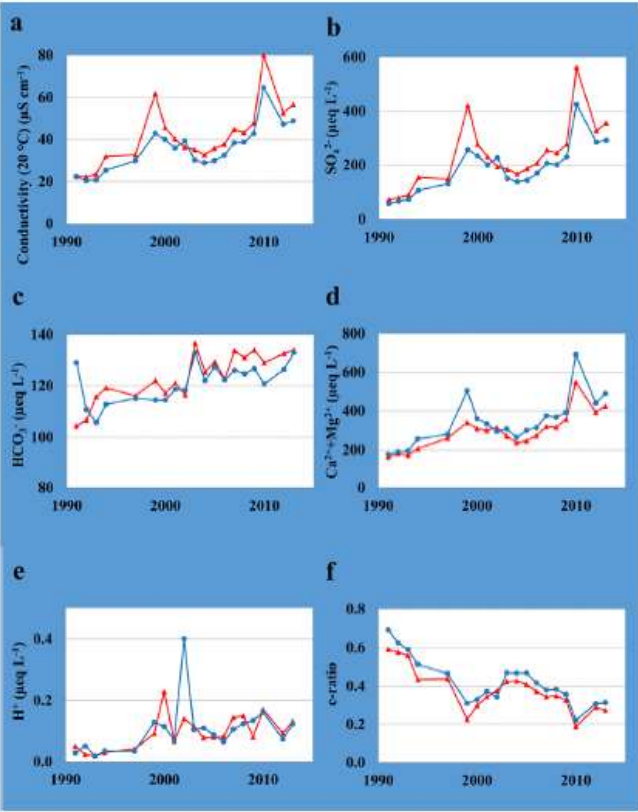
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524 FIGURE 2. Scatter-plots between a) conductivity, b) SO_4^{2-} , c) HCO_3^- , d) $\text{Ca}^{2+}+\text{Mg}^{2+}$, e) H^+ , and f) c-ratio of the survey
525 lakes sampled during the early 1990s and the early 2010s. The orange points represent lakes without glaciers within the
526 basin.



533 FIGURE 3. Long-term trends of the annual values of a) conductivity, b) SO_4^{2-} , c) HCO_3^- d) $\text{Ca}^{2+}+\text{Mg}^{2+}$, e) H^+
534 concentrations, and f) c-ratio in LCN9 (red) and LCN10 (blue).



545 FIGURE 4. a) PCA among SO_4^{2-} , HCO_3^- concentrations of the survey lakes (early 1990s), land cover, and snow cover
546 duration (*Glacier*: glacier coverage during the early1990s; *Baresoil*, *Barerock*, *Shrubs*: coverage for 2008; *Snow*: mean
547 snow cover duration for each basin during the 2010s). b) The most significant correlation that emerged from the above
548 PCA.

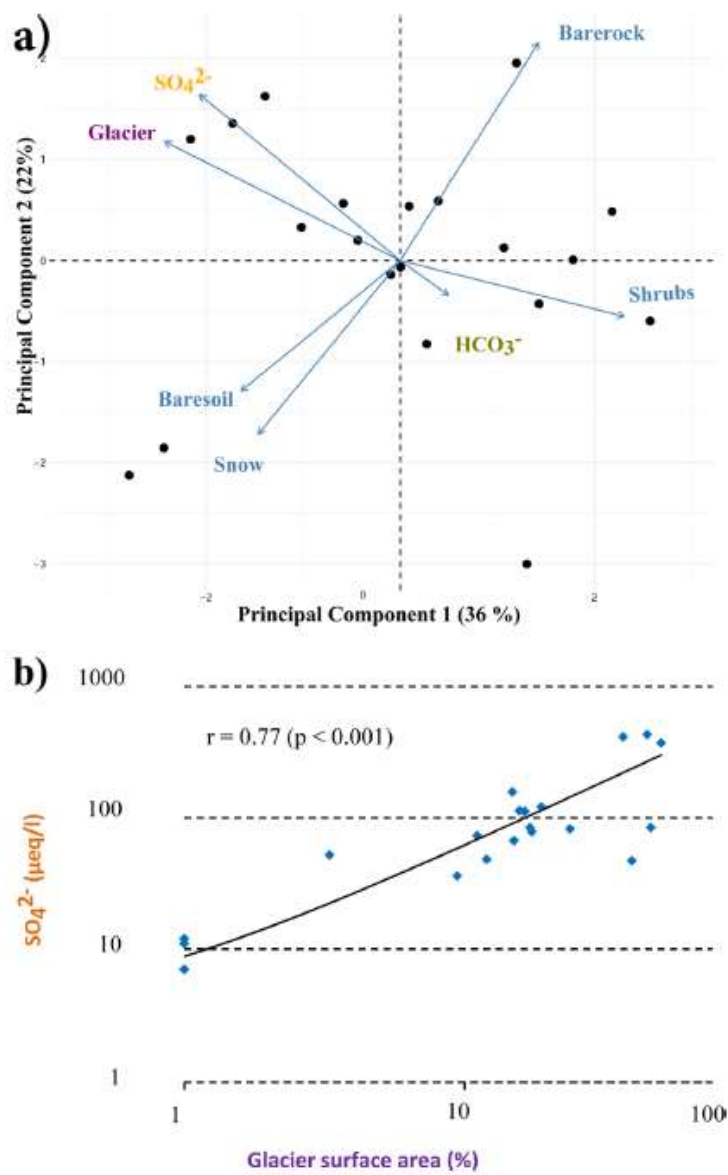
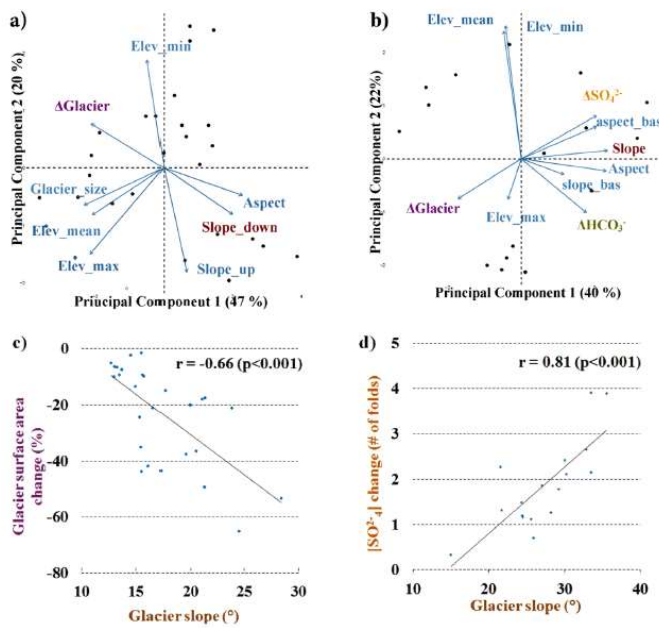
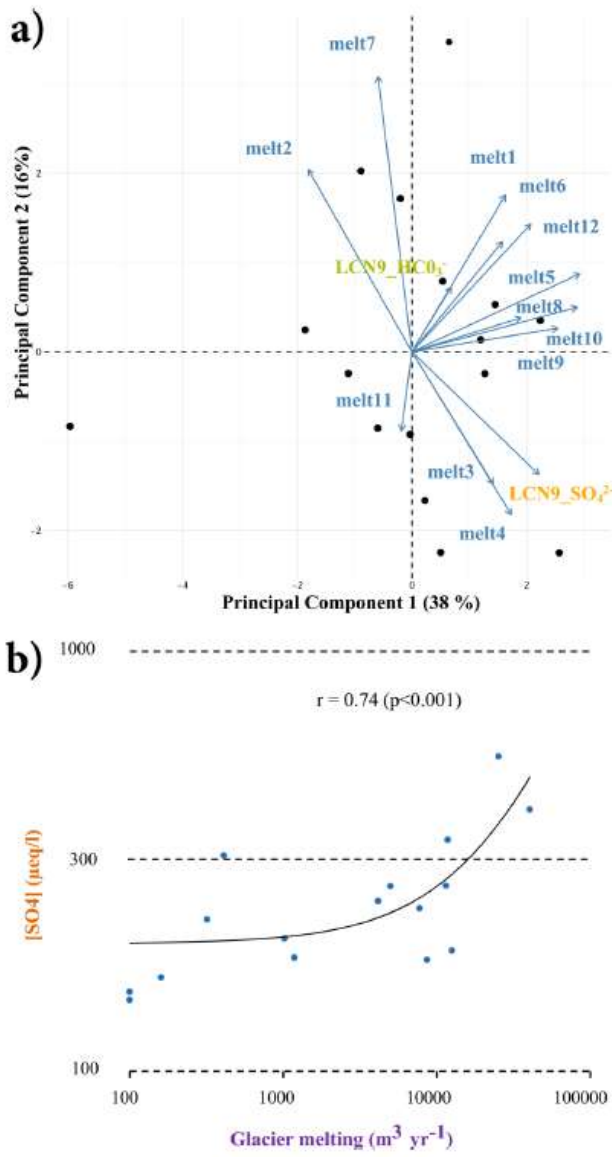


FIGURE 5. PCAs among morphometric parameters and a) glacier surface area variations ($\Delta Glacier$) that occurred in the last twenty years in the overall SNP (data from Thakuri et al.¹⁰); b) relative SO_4^{2-} and HCO_3^- variations (ΔSO_4^{2-} , ΔHCO_3^-) that occurred in the same period for the 20 survey lakes; (*Glacier_size*=size of glaciers; *Elev_max*, *Elev_mean*, *Elev_min*: maximum, mean, and minimum glacier elevation, respectively; *Slope*, *Slope_down*, *Slope_up*: mean, downstream, and upstream glacier slope, respectively; *Aspect*: mean glacier aspect). The lower graphs (c and d) show the most significant correlations that emerged from the relevant PCAs shown above.



571 FIGURE 6. a) PCA among SO_4^{2-} and HCO_3^- concentrations of LCN9 sampled annually in late September or October
572 (LCN9_SO_4^{2-} , LCN9_HCO_3^- , respectively) and the cumulated glacier melt (melt_i) modeled for each month (i) of the
573 year. b) The lower graph shows the most significant correlation that emerged from the PCA: $\text{SO}_4^{2-}\text{-LCN9}$ vs melt_4 .

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