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AN INVESTIGATION OF WATER AND ICE CHEMISTRY IN A FAST-RETREATING GLACIER IN THE DOLOMITES (NE ITALY): THE FRADUSTA GLACIER

ABSTRACT: DOSSI C., VIGANÒ A., MARTIN S., MONTICELLI D. & POZZI A., *An investigation of water and ice chemistry in a fast-retreating glacier in the Dolomites (NE Italy): the Fradusta glacier.* (IT ISSN 1724-4757, 2006).

Fradusta glacier (Pale di San Martino massif), the second largest ice-field in Italian Dolomites, is evidencing a fast retreat, with a surface area reduction of about 90% in the last 100 years, and a negative variation of glacier front of 115.5 m between 1926 and 2000. Analytical chemical studies, conducted on the field with portable instrumentation for physicochemical parameters and in the analytical laboratory for main ion determination via IC, suggested the occurrence of strong chemical interactions between melting waters and the dolomitic bedrock. The presence of ammonium ion in snow and ice samples is indicative of long-range global anthropogenic impact, which might be related to «red snow» events. Accordingly, no correlation of ammonium is found with alkalinity, indicating the absence of local pollution, as confirmed by low concentrations of sulphate and nitrate ions in snow and ice. Ammonium had a particular significance in this study, since it was proposed as an «in-situ», environmentally-friendly chemical tracer for a better understanding of hydrological karstic circulation.

KEY WORDS: Fradusta Glacier, Dolomites, Water and Ice chemistry, Anthropogenic impact.

RIASSUNTO: DOSSI C., VIGANÒ A., MARTIN S., MONTICELLI D. & POZZI A., *Studio interdisciplinare del chimismo di acqua e ghiaccio nel Ghiacciaio di Fradusta (Dolomiti, Italia).* (IT ISSN 1724-4757, 2006).

Questo studio ha messo in evidenza l'elevato e rapido ritiro del Ghiacciaio di Fradusta nel gruppo delle Pale di San Martino (Dolomiti), con una riduzione areale del ghiacciaio pari al 90% circa negli ultimi 100 anni, accompagnata da un ritiro della fronte di 115.5 m tra il 1926 ed il 2000. Gli studi chimico-analitici, effettuati sia sul campo con strumentazione portatile per i parametri chimico-fisici, che in laboratorio analitico per l'analisi degli ioni principali in IC, hanno evidenziato le interazioni chimiche che avvengono tra le acque di fusione e le rocce dolomitiche del bacino. Nel caso di neve e ghiaccio, si è potuto invece evidenziare come la presenza di elevate concentrazioni di ione ammonio sia correlata a fenomeni di inquinamento globale, tipicamente agli eventi di «neve rossa» di origine Sahariana. Questo studio ha inoltre messo in evidenza e proposto il ruolo chiave dello ione ammonio come possibile tracciante naturale a basso impatto ambientale per lo studio della circolazione carsica profonda nel massiccio delle Pale di San Martino.

TERMINI CHIAVE: Ghiacciaio di Fradusta, Dolomiti, Chimismo di acqua e ghiaccio, Impatto antropico.

INTRODUCTION

High-altitude glaciers, as well as Anctartica and Greenland, are recently receiving increasing environmental importance as historical archives, since they are storing, within their massive ice layers, paleoclimatic and paleopollution information on our past environment (Boutron, 1995; Barbante & *alii*, 2003; Caroli & *alii*, 2001; Marinoni & *alii*, 2001, Schwikowski & *alii*). The suitability of high-altitude sites for ice coring studies has been investigated and rationalized (Eichler & *alii*; Maupetit & Delmas, 1994; Preunkert & *alii*, 2000).

A different situation arises for low-altitude, small-mass glaciers, where rainfalls and summer temperature above 0°C may cause liquid water percolation and significant remixing phenomena through ice layers. In this way, all chemical (e.g. main and trace elements, organic pollutants) and physical (e.g. tritium) information are averaged within the thin ice body of the glacier with the final effect of destroying any annual historical information. Nevertheless, such small glaciers may still represent an important environmental resource. Owing to their small mass, they are

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characterized by fast response time to recent climate changes, such as global warming; at the same time, they can be considered as the ideal environment to trace the impact and the changes of local and/or long range anthropogenic sources.

The Fradusta glacier is a typical representative of such kind of glaciers. It is the second largest glacier of the Dolomites, after the Marmolada glacier, and the most important one of the Pale di San Martino massif, which is located in the Southern Dolomites (NE Italy) at the boundary of the Belluno and Trento provinces. This massif includes several peaks higher than 2500 m a.s.l., the highest being Cima della Vezzana (3193 m a.s.l.) and it is characterized by the presence of a wide plateau at an average altitude of 2500 m a.s.l. Most streams of the massif belong to the Piave river system (Biois, Mis, Cismon streams); the Travignolo stream on the northwestern side only belongs to the Adige river system (fig. 1). The Fradusta glacier is situated upon the Pale di San Martino plateau on the northern face of the Cima di Fradusta (2937 m a.s.l.) (see fig. 2). The Pale di San Martino massif is mainly composed of dolomitic rocks of Ladinic age (Dolomia dello Sciliar Formation) and dolomitic and calcareous rocks of Upper Ladinic-Carnic age (Dolomia Cassiana Formation), derived respectively from major and progradating carbonatic shelves. Volcanic rocks and basinal sediments of Ladinic age are also present in the northern part of the massif (Castiglioni, 1939). The Fradusta glacier lies both on the Dolomia dello Sciliar Formation (Pale di San Martino plateau) and the Dolomia Cassiana Formation (Cima di Fradusta) (Zampieri, 1987).

In this paper, the Fradusta glacier was mainly investigated on an analytical chemical approach, with the aim of rationalizing water and ice chemistry with respect to dolomitic bedrock, and of evaluating natural and anthropogenic impact on this important natural environment.

EXPERIMENTAL

Sampling campaign was undertaken between late spring and summer 2003. Sampling sites are indicated in figure 2.

Snow was sampled at the beginning of June, when all the plateau of the Pale di San Martino was still uniformly covered with fresh snow (FSnow sample). The presence of a thin, red layer of fine dust on the snow surface was indicative of the early occurrence of dust storm events of Sa-

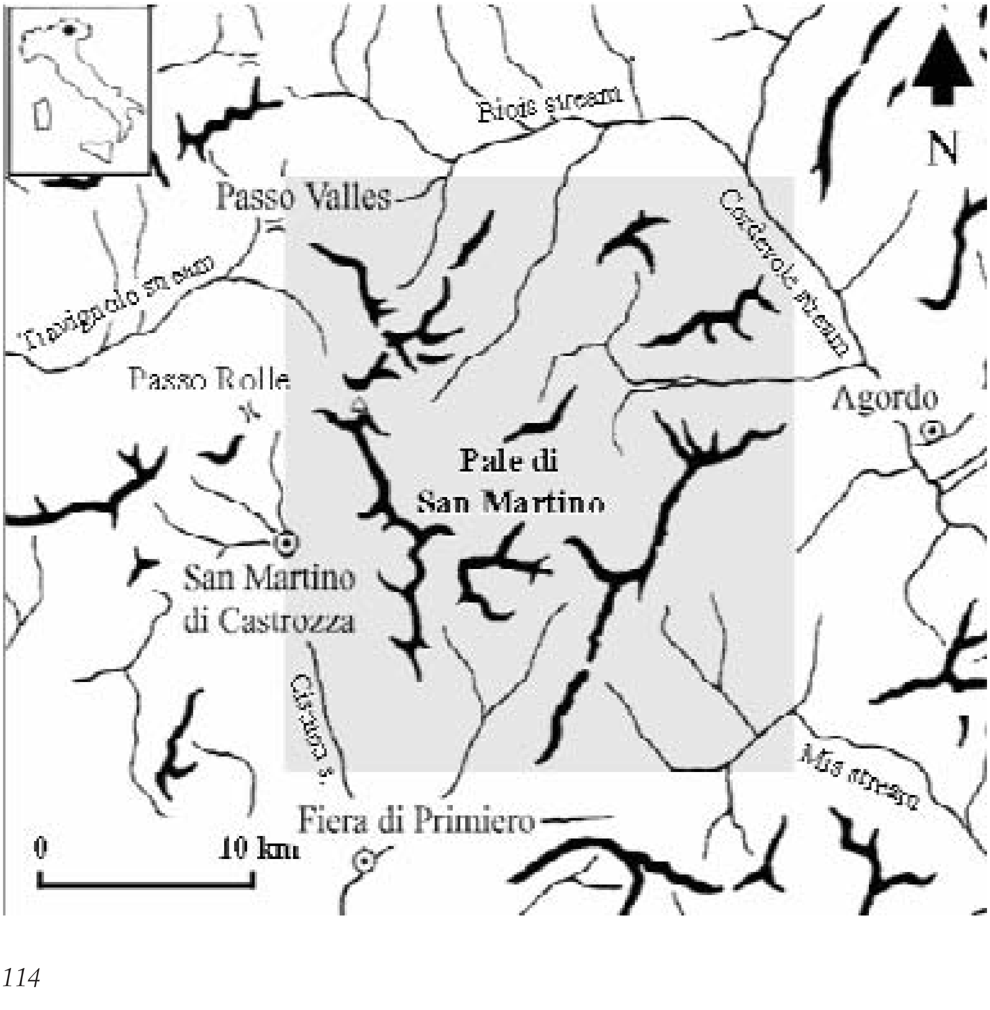

FIG. 1 - Geographic setting of the Pale di San Martino massif.

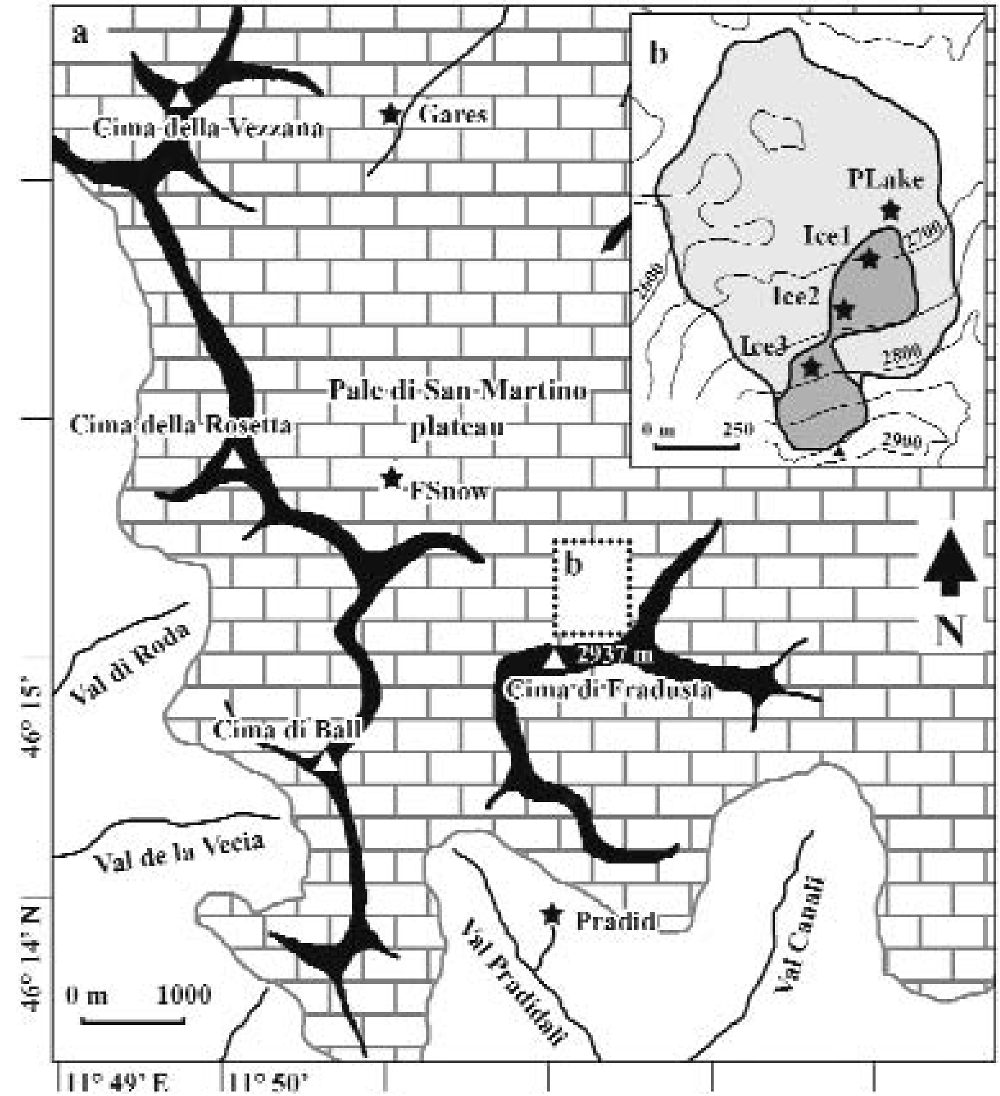

FIG. 2 - a) Simplified map of the Pale di San Martino massif. The interpreted boundary of the carbonatic shelf (brick pattern) and the sampling sites (stars with sample codes as in table 1) are represented. b) The Fradusta glacier area in 1910 (pale grey) and in 2003 (dark grey).

haran origin (Rogora & *alii*, 2004). Ice samples were collected in summer 2003, when no residual snow was left on the glacier surface. Three samples of ice, indicated as Ice1, Ice2 and Ice3, have been collected along an ideal line connecting the front with the upper part of the glacier. Water samples were collected in the proglacial lake formed at the front of the glacier (PLake sample), on the Comelle stream at the Pian delle Comelle (Gares sample) and the main water spring in Val Pradidali (Pradid sample).

Ice was sampled by a titanium ice axe, which was thoroughly cleaned with deionised water prior to each sampling: the uppermost layer of ice was discharged, as it is likely to be heavily contaminated. No depth profiling was undertaken, because all chemical and physical information is averaged through ice layers. Because of the scarce snow coverage at the end of spring season, snow was simply collected by dipping a large mouth, 250-mL bottle into it. Water was sampled against waterflow in 250-mL bottles for total alkalinity, and in 20-mL vials for cation and anion analysis. Samples for cation analysis were stabilized to pH 2 with Suprapur HNO_3 (65% Traceselect, Fluka).

Low-density polyethylene (LDPE) was used for all sampling bottles and vials. A standard cleaning protocol was undertaken, by soaking sampling bottles and vials in diluted ultrapure nitric acid (HNO_3 Suprapur, Merck) to remove all contaminants, followed by several rinsing with ultrapure MilliQ water (Millipore MilliQ System, 18 MΩ·cm resistivity) until any residual acid trace is eliminated. Sampling bottles and vials are then stored in clean place before sampling (Howard & Stratham, 1993).

Physicochemical parameters (temperature, pH and conductivity) were determined in the aqueous samples on the field. Temperature and pH were measured through a portable Hanna HI9025C pHmeter and conductivity through a portable Hanna HI9033 conductivity meter; both instruments were calibrated daily.

Major ions were determined in the laboratory by means of ion chromatography (IC761 from Metrohm, Herisau, Switzerland) with autosampler and conductimetric detection. A Phenomenex StarIon A300 column was used for the isocratic separation of chloride, nitrite, nitrate and sulphate ion with a 1.5-mL/min flow of a carbonate/bicarbonate eluent (1.8 mM Na_2CO_3 + 1.7 mM $NaHCO_3$). Sodium, ammonium, potassium, magnesium and calcium ions were separated on a Metrohm Metrosep Cation 1-2 column with an eluent composition of 4 mM tartaric acid and 1 mM dipicolinic acid and a flow of 1.0 mL/min. Standard solutions were prepared by proper dilution of a 1000 ppm standard solution (Merck); diluted solution (<0.05 mg/L) were prepared daily and acidified to pH 3 for cation analysis. Limits of detection (LOD) were estimated following the IUPAC $3s_{blank}$ formula (Mocak & *alii*, 1997) and are in the 1-3 mg/L for anions and in the 10-25 mg/L range for cations.

All data are summarized in table 2. Concentrations below detection limit are indicated as «<LOD». Nitrite ion was below detection limit in all samples, and data are not reported.

Total alkalinity was performed by acid titration with 0.01 M hydrochloric acid through a computer based automatic titrator developed in our laboratory (Dossi & *alii*, 2000). End-point was evaluated via Gran's plot (Marchetto & *alii*, 1997).

TABLE 1 - Geographic and physicochemical sampling data

Sample code	Sampling date (year 2003)	Sampling site	Height (m a.s.l.)	T (°C)	pH	Specific conductivity (µS/cm)
FSnow (fresh snow)	05 June	Pale di San Martino plateau	2640	–	5.68	3.8
Ice1 (surface ice)	26 July	Fradusta glacier	2690	–	6.72	17.4
Ice2 (surface ice)	26 July	Fradusta glacier	2725	–	8.00	37.1
Ice3 (surface ice)	26 July	Fradusta glacier	2760	–	7.61	87.3
PLake (lake water)	25 July	Fradusta proglacial lake	2680	2.6	7.73	53.1
Pradid (spring water)	10 September	Pradidali spring	1515	4.3	8.52	102.8
Gares (stream water)	11 September	Stream at the Orrido delle Comelle	1845	4.2	7.88	130.3

- Not measured

RESULTS AND DISCUSSION

The mapping of the glacier was easily undertaken in July when the ice surface was completely exposed, because of the scarce snow coverage at the end of the 2002-2003 winter and the fast snow melting promoted both by the high temperatures of the spring and summer seasons (Valt & Cagnati, 2003). The measurements evidenced a surface area of about 11.5 ha (fig. 2), with an overall reduction of almost 90% during the last one hundred years (107 ha in 1910 (Marinelli, 1910), 65 ha in 1954 (Consiglio Nazionale delle Ricerche - Comitato Glaciologico Italiano, 1962) and 23.1 ha in 1999 (ARPAV, 1999, unpublished data). At present, the glacier extends between 2890 and 2690 meters a.s.l., with an average slope of about 11 degrees. The glacial retreat was most evident in the last decade and it was also testified by the negative variation of the glacier front of 115.5 meters between 1926 and 2000.

The reduced ice thickness led to bedrock emerging and a partial division of the glacier in two different lobes. The surface of contact between these two lobes is only about ten meters wide and few meters deep. To our opinion, this division into separate ice bodies will be definitive in the next few years.

Geographical and physicochemical data of ice and water samples are summarized in tab. 1. The three water samples show pH values higher than 7.5 and increasing values of specific conductivity moving from the Fradusta proglacial lake to Gares stream. This is the result of chemical weathering of dolomite-enriched carbonatic rocks of the Pale di San Martino massif, leading to the increase of ionic concentration of Ca^{2+}, Mg^{2+} and $[HCO_3]^-$ (tab. 2). These three ions are, in fact, accounting for over 95% of overall ionic balance in water samples. Nitrate and sulphate ions are, instead, minor components with respect to bicarbonate, with the exception of fresh snow sample. Accordingly, fresh snow is characterized by low values of pH and specific conductivity, as it can be expected from the combined effect of the mineral acidity of precipitations and the limited interactions with particulate matter deriving from eolic transport, which would lead to buffering effect of bicarbonate ion. The ratio between magnesium and calcium ions in water samples («Mg/Ca ratio») is here assuming a peculiar environmental significance, since it may immediately differentiate if the water and/or ice are chemically interacting with the dolomite rocks of the basin or with the eolic dry depositions of Saharan origin (fig. 3). Saharan dust events are characterized by high calcium contents, leading to low Mg/Ca ratios in the 0 to 0.2 range.

TABLE 2 - Concentrations (as mg/L) of anions and cations in ice, snow and water samples as determined by ion chromatography

Sample code	$[HCO_3]^-$	Cl^-	$[NO_3]^-$	$[SO_4]^{2-}$	Na^+	$[NH_4]^+$	K^+	Ca^{2+}	Mg^{2+}
Fsnow	0.61	0.17	0.12	0.08	0.20	1.06	0.09	0.21	0.02
Ice1	3.66	0.19	<LOD	0.05	0.18	1.01	0.16	0.64	0.24
Ice2	9.82	0.08	<LOD	0.01	0.20	1.07	0.09	2.59	0.16
Ice3	16.59	0.02	<LOD	0.03	0.13	1.12	0.16	3.83	0.70
Plake	30.50	0.01	0.17	0.44	0.28	0.22	0.38	6.46	1.41
Pradid	56.43	0.07	2.47	1.86	0.18	0.17	<LOD	12.00	5.27
Gares	72.35	0.23	2.23	2.35	0.27	<LOD	<LOD	15.36	7.20

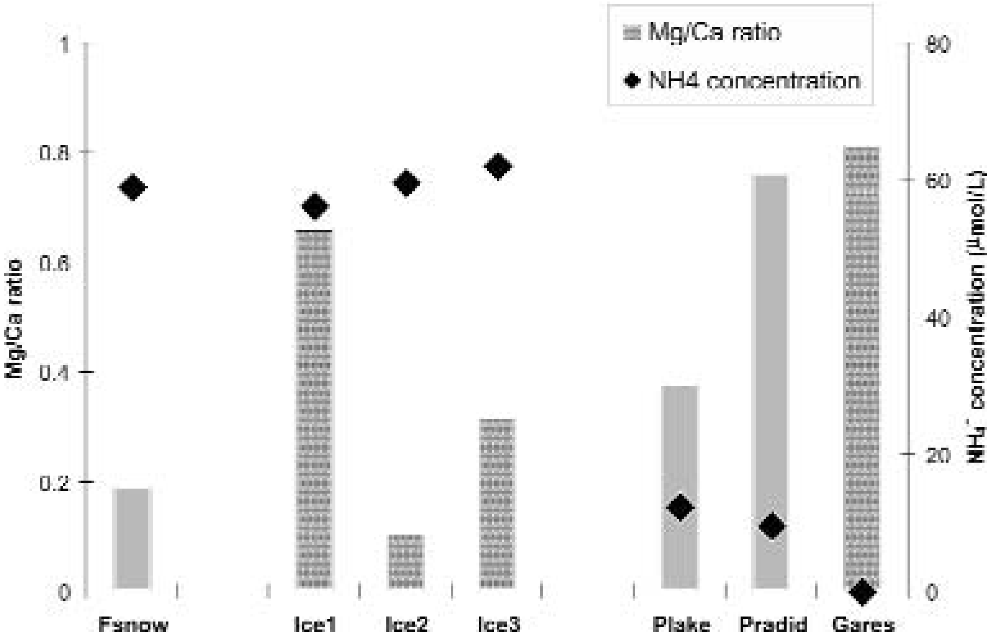

FIG. 3 - Variation of Ca/Mg ratio and NH_4^+ concentration in snow, ice and water samples.

Accordingly, fresh snow shows a Mg/Ca ratio near 0.2. In fact, all the residual snow in late spring was uniformly covered with a thin layer of red dust.

On the contrary, water samples show increasingly high Mg/Ca ratios, reaching values around 0.7-0.8 for the Pradidali spring and the Gares stream. These values are very close to the theoretical value of 1, which has to be expected in the case of weathering of pure dolomite ($CaMg(CO_3)_2$):

$$(CaMg(CO_3)_2) + 2\ H_2O + 2\ CO_2 \rightarrow Ca^{2+} + Mg^{2+} + 4\ HCO_3^-$$

The ice samples collected at different locations on the glacier show intermediate values between snow and water. They are characterized by a great variability of analytical values, particularly for specific conductivity and Mg/Ca ratios which range between 17.4 to 87.3 and between 0.10 to 0.66, respectively. This large spreading of analytical data may reflect the heterogeneity of the glacier surface. At the end of July, when residual winter snow was disappearing, a significant fraction of the glacier surface showed a spongy-like appearance, because of the fast melting of residual winter snow coverage. Particulate matter, which is depositing on the glacier surface during dry season, is then easily incorporated and slowly penetrates through ice layers, leading to increasingly higher pH, conductivity and calcium contents; this is the consequence of the chemical dissolution of finer dust. This process appears to be predominant on the upper part of the glacier, since conductivity values are linearly related to altitude.

Other chemical species, primarily ammonium, sodium, chloride ions and hydrogen peroxide, tend, instead, to concentrate on the upper layers of ice. They are in fact much better incorporated into ice during melting and refreezing steps in snowmelting (Kuhn, 2001).

Ammonium ions may be regarded as a typical seasonal marker of long-range, global anthropogenic impact, such as industrial pollution and farming activity. Ammonium concentration appears to be completely uncorrelated with total alkalinity, which is a marker of local origin (fig. 4). It could be acceptable that, in sedimentary rock basins, ammonia may play a minor, if any, role in buffering enhanced atmospheric acidification. Acid-base equilibria are then governed by the dissolution of Ca and Mg carbonates. Figure 4 may indeed evidence the direct correlation existing between the total alkalinity and the concentration of Ca^{2+} and Mg^{2+} ions.

In all snow and ice samples, ammonium concentrations are remarkably high, around 1 mg/L (59.1-62.1 µmol/L) (tab. 2, fig. 3), and closely resemble reported values in the case of the dry dust deposition event at Colle Vincent in June 1994 (Novo & Rossi, 1998). On the contrary, typical weighted mean ammonium concentrations in the snow cover, after accumulation, are significantly lower. Analytical data collected during SNOSP project in 1991-1993 years show concentrations about one order of magnitude lower, ranging between 0.045 and 0.18 µg/L (Nickus & *alii*, 1997).

We may thus infer that the scarce snow coverage in the 2002-2003 winter-spring season and the late occurrence of dry deposition events are responsible for the ammonium levels in fresh snow; interestingly, this situation appeared to common in different sites of the Italian Alps (Dossi & *alii,* unpublished results).

Ammonium ions are short-lived species, which can be oxidized to gaseous dinitrogen and nitrate ions via bacterial and/or metal-catalyzed oxidative decomposition with oxygen during hot season. At low temperatures, the oxidation kinetics of ammonium ion are relatively slow, explain-

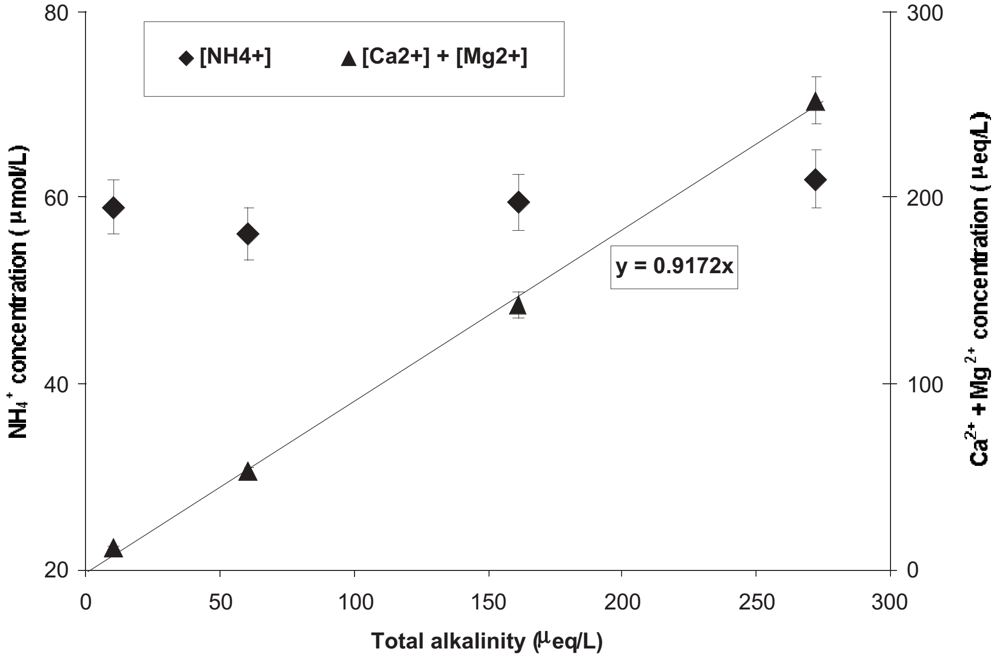

Figure 4 - Correlations between the concentration of NH_4^+, and of the sum of Ca^{2+} + Mg^{2+} with total alkalinity in snow and ice samples from Fradusta glacier.

ing its presence in the Fradusta proglacial lake, whereas in the Gares stream is totally decomposed by oxidation. Interestingly, some residual ammonium is still found in the Pradidali spring. We may reject any influence of human or farming activities on ammonia levels because of the remote location of Pradidali spring and the absence of nitrite or high levels of NaCl in springwater; which are known, in the case of the Marmolada glacier (Viganò & *alii*, 2003), to be correlated with a significant anthropogenic impact. The origin of ammonia in this springwater is then to be related to the existence of a karstic hydrological circuit which directly connects major springs in Val Pradidali with the carbonatic Pale di San Martino plateau. It is important to evidence that our hypothesis has been confirmed by recent hydrological experiments with fluorescein (Borsato *& alii*, 2002). As the consequence, rainfall and meltwaters from ice and snow rapidly percolate through the karstic circuit, reaching the Pradidali spring after few days, before its complete oxidation to N_2 and $[NO_3]^-$ has taken place. Ammonia can thus be considered a sort of a natural, «in-situ» tracer for hydrological monitoring which could prevent or better complement the use of conventional chemical tracers (fluorescein and Tinopal CBS-X) with their unwanted environmental side effects.

CONCLUSIONS

The Fradusta glacier is undergoing a very fast retreat, with an overall areal reduction of about 90% in the last century. Deglaciation is most evident in the center of the glacier, where bedrock is emerging with the partial division of the glacier in two different lobes. If the present global warming is continuing, the division into two separate ice bodies will be definitive in the next few years. The chemical data collected during the sample campaign in spring-summer 2002 gave interesting clues about the weathering mechanism of dolomitic rocks by the aggressivity of ice and meltwater. Ice and snow samples were characterized by relatively high levels of ammonium ions, around 1 mg/L, which are proposed to derive from long-range transport. The high ammonium concentrations in meltwater and the slow oxidation kinetics in karst circuits have been exploited to use $[NH_4]^+$ as an «in-situ» natural tracer. In this way, a better understanding of the hydrological karstic circulation of the Pale di San Martino plateau was reached, avoiding any utilization of environmentally-unfriendly chemical tracers such as fluorescein or Tinopal.

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