

SOLUTE ACQUISITION IN GLACIAL MELT WATERS. II. ARGENTIÈRE (FRENCH ALPS):

BULK MELT WATERS WITH OPEN-SYSTEM CHARACTERISTICS

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ABSTRACT. Variations in dissolved cations, total alkalinity, sulphate, and field pH are recorded for subglacial melt and bulk melt waters (those emerging from the portal) at Argentière (France), in peak and recession flow conditions. Calcium and bicarbonate are the major ions and the bulk melt waters are demonstrated to have acquired solutes by weathering and dissolution in a system open to atmospheric carbon dioxide. Subglacial melt waters have closed-system characteristics, are close to saturation with calcite and quartz, and must be in near-equilibrium with weathered particulates. Recession-flow bulk melt waters are chemically similar to subglacial melt but have open-system characteristics, either due to re-equilibration with the atmosphere for ground-water mixing.

RÉSUMÉ. Dissolutions par les eaux de fusion glaciaire II. Argentière (Alpes françaises): Eaux de fusion réurgentes avec caractéristiques de système ouvert. On a enregistré les variations en cations dissous, en alcalinité totale, en sulfates et en pH courant pour des eaux sous-glaciaires et des eaux de fusion réurgentes (celles qui sortent du portail sous-glaciaire) à Argentière (France), dans des conditions de crue et d'étiage. Le calcium et le bicarbonate sont les ions les plus abondants et on démontre que les eaux de fusion réurgentes ont acquis leurs ions dissous par altération et dissolution dans un système ouvert au dioxyde de carbone atmosphérique. Les eaux sous-glaciaires ont des systèmes clos caractéristiques, sont proches de la saturation en calcite et quartz et doivent être proches de l'équilibre avec les particules attaquées. Les eaux de fusion réurgentes sont chimiquement semblables aux eaux de fusion sous-glaciaires mais présentent des caractéristiques de système ouvert, soit en raison d'une remise en équilibre avec l'atmosphère soit par mélange avec des eaux d'origine souterraine.

INTRODUCTION

Chemical models for the evolution of glacial melt waters (Raiswell and Thomas, 1984; Raiswell, 1984) suggest that kinetic factors exert a critical control on the nature and magnitude of sediment-water interactions and thus on the composition of bulk melt waters (those emerging from the portal). Solute acquisition in the glacial system may follow two distinct chemical pathways, where carbon dioxide dissolution is the principal source of the hydrogen ions consumed during the weathering and dissolution of rock minerals. A closed-system pathway occurs where the rate of CO₂ consumption by weathering reactions exceeds the rate of CO₂ renewal by dissolution. This pathway has been described for melt waters at Fjallsjökull, south-east Iceland (Raiswell and Thomas, 1984). It is the purpose of this paper to describe the chemical evolution of bulk melt waters with open-system characteristics (rates of CO₂ consumption by weathering lower than rates of renewal by dissolution).

SAMPLING AND ANALYSIS

Glacier d'Argentière is on the south-west side of the Chamonix valley, in the Mont Blanc massif of the French Alps. All the bulk melt waters were collected from a site approximately 150 m down-stream from the glacier snout. The melt waters were sampled at approximately hourly intervals for 10 h (from 10.00 to 20.00 h) in April 1975, and for 36 h (from 7.00 to 19.00 h) in September 1976. The weather at both times was atypical for that time of year. During the April survey it was unusually cold and there was still extensive snow cover down to 1 000 m a.s.l. During the

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September survey, the weather was mainly warm and dry, but precipitation (snow above 3 000 m a.s.l.) occurred during the sampling period. In addition to the bulk melt waters, two samples were collected from subglacial streams near a subglacial laboratory located by the ice-rock interface (Vivian, 1975), approximately 1 500 m above the glacier snout.

ZUSAMMENFASSUNG. Lösungsanreicherung in Gletscherschmelzwasser. II. Argentière (französische Alpen): Massen-Schmelzwasser aus einem offenen System. Für subglaziales und Massen-Schmelzwasser (austretend am Gletschertor) am Argentière-Gletscher (Frankreich) wurden die Änderungen in gelösten Kationen, der Gesamtalkalität, des Schwefelgehaltes und des Feld-pH-Wertes bei maximalem und zurückgehendem Abfluss festgestellt. Calcium und Bicarbonat sind die hauptsächlichsten Ionen. Es wird gezeigt, dass die Massen-Schmelzwässer ihre Lösungen durch Verwitterung und Auflösung in einem für das Kohlendioxid der Luft zugänglichen System gewonnen haben. Subglaziale Schmelzwässer haben Charakteristiken eines geschlossenen Systems, sind mit Kalzit und Quarz beinahe gesättigt und müssen sich nahezu im Gleichgewicht mit Verwitterungspartikeln befinden. Massen-Schmelzwässer bei zurückgehendem Abfluss ähneln chemisch subglazialen Schmelzwässern, haben jedoch die Charakteristiken eines offenen Systems, entweder infolge der Rückgewinnung des Gleichgewichtes mit der Atmosphäre oder durch Vermischung mit Grundwasser.

Granitic and gneissic rocks constitute the major rock types in the Mont Blanc massif. Normative mineral computations from wet chemical data (personal communication from M.S. Brotherton) together with X-ray diffraction and optical studies, indicate that quartz and feldspars (orthoclase, albite >> anorthite) constitute 84% and 93% of the gneiss and granite respectively. A little calcite (c. 0.1%) was also present in the gneiss.

In both surveys, pH and conductivity were measured at the time of sample collection and also subsequently, in the laboratory, after deep freezing for transportation. Storage effects were small and unimportant in the present interpretations. Most remaining analyses (Na⁺, K⁺, Mg²⁺, Ca²⁺, SO₄²⁻) were determined in the laboratory, using the methods outlined in Raiswell and Thomas (1984). The only difference between the two surveys concerned total alkalinity (OH⁻ + HCO₃⁻ + CO₃²⁻) which was measured either in the field (September survey) or in the laboratory (April survey).

RESULTS

Analytical data (Table I) are given for water samples with the maximum and minimum dissolved loads, and for the mean composition, in each survey. Charge balances suggest the presence of another cation in the September data, but accurate analysis is difficult at these low concentrations. In the April data, charge balances have a less than 10% error in all except three bulk melt waters and the two subglacial samples, where another anion is indicated. Chloride was

TABLE I. ANALYTICAL DATA

Composition	Compositional range			Subglacial melt		Compositional range		
	Maximum	Mean	Minimum			Maximum	Mean	Minimum
Ca ²⁺ mmol l ⁻¹	0.238	0.214	0.202	0.084	0.132	0.050	0.033	0.010
Mg ²⁺ mmol l ⁻¹	0.033	0.030	0.023	0.086	0.003	0.001	0.0007	0.003
K ⁺ mmol l ⁻¹	0.052	0.044	0.033	0.088	0.096	0.020	0.013	0.006
Na ⁺ mmol l ⁻¹	0.089	0.077	0.056	0.257	0.094	0.026	0.015	0.010
HCO ₃ ⁻ mmol l ⁻¹	0.40	0.43	0.38	0.27	0.24	0.12	0.105	0.11
SO ₄ ²⁻ mmol l ⁻¹	0.083	0.074	0.069	0.076	0.075	0.020	0.014	0.005
Σ ⁺ meq l ⁻¹	0.68	0.61	0.54	0.68	0.46	0.146	0.098	0.053
pH	8.15	7.73	7.55	9.7	9.35	7.05	7.09	7.2
-log ₁₀ p(CO ₂)	3.83	3.42	3.30	5.74	5.36	3.19	3.28	3.37
(SI) _{calcite}	-1.18	-1.66	-1.91	-0.44	-0.56	-3.39	-3.56	-3.68

measured but was below the detection limit by ion selective electrode (1 μmol l⁻¹). Considerably higher dissolved loads (measured by Σ⁺, the sum of the cation equivalents) occur in April than in September and are associated with higher magnesium contents, but there is otherwise little compositional variation either within, or between, the two surveys (Fig. 1). Calcium is the major cation (70-80% total cation equivalents) and bicarbonate is the major anion (80-90% total anion equivalents). Clearly differences between the two surveys are a function of the magnitude, rather than the nature, of the dissolved load. In comparison with the bulk melt waters, the two subglacial samples are markedly depleted in Ca²⁺, or enriched in Na⁺ and K⁺.

The discharge during each survey was nearly constant, although very much smaller in April (0.2 m³s⁻¹) than in September (3.0 m³s⁻¹). Discharge values were recorded at the Electricité de France (EDF) gauging station (c. 400 m down-stream from the glacier snout) but are at best accurate to no more than ±15%. Within these limits the absence of characteristic diurnal variations in discharge (e.g. Rainwater and Guy, 1961; Collins, [1978]) in April was probably due to cold weather and snow preventing the formation of supraglacial melt. Similarly, in September, snow would have prevented surface melt above 3 000 m a.s.l., whilst below this altitude the addition of rain would still further disrupt simple diurnal characteristics.

Carbonate equilibria

The hydrogen ions required for the weathering and dissolution of rock minerals in glacial systems are mainly derived from the solution of atmospheric CO₂ and the oxidative breakdown of sulphide minerals (Raiswell, 1984). The relative contributions of these two processes and the nature of the chemical pathway (whether open or closed) can be evaluated from analyses of cationic and anionic species, which allow the calculations of p(CO₂) values and Saturation Indices (SI) using the computer program WATSPEC (Raiswell and Thomas, 1984). Table I summarizes the p(CO₂) and (SI)_{calcite} values for all the sampled melt waters. The mean p(CO₂) values for the two surveys are similar (10^{-3.4} and 10^{-3.3}), and are both close to equilibrium with the atmosphere p(CO₂) = 10^{-3.5}), but there is a considerable difference in the (SI)_{calcite} values. Both sets of melt waters

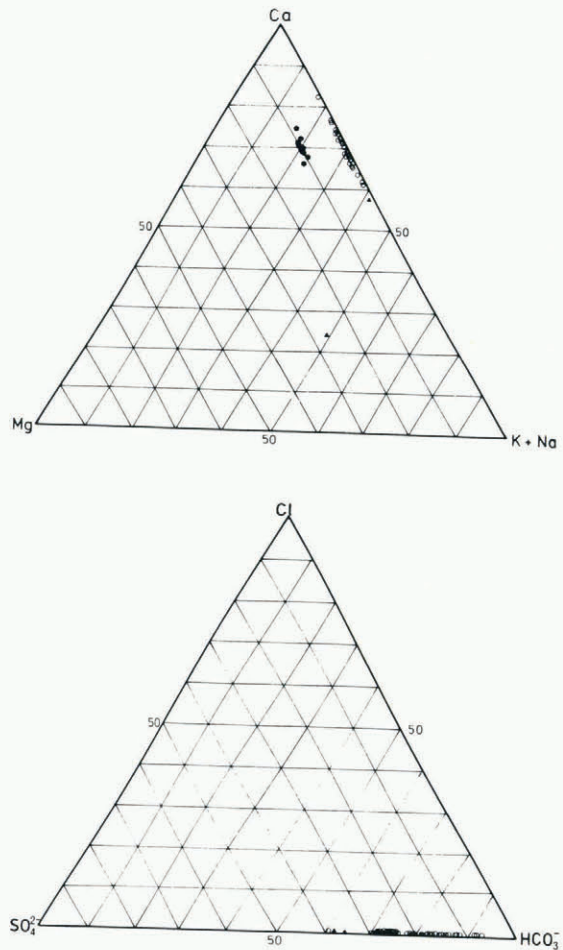


Fig. 1. Variations in the relative proportions of cation and anion equivalents in bulk melt waters from Argentière sampled in April 1975 (●) and September 1976 (○), also subglacial melt waters (▲) sampled in April 1975.

are clearly undersaturated with respect to calcite, but the degree of undersaturation is very much greater for the autumn survey (mean $(SI)_{\text{calcite}} = -3.6$) than for the Easter survey (mean $(SI)_{\text{calcite}} = -1.7$).

DISCUSSION

Influence of discharge on total dissolved load

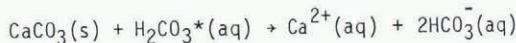
The difference in discharge between the April ($0.2 \text{ m}^3\text{s}^{-1}$) and September ($3.0 \text{ m}^3\text{s}^{-1}$) surveys almost certainly reflects meteorological conditions in the periods prior to, and during, sampling. Once supraglacial melt ceases, discharge is maintained for a period of one to several days by the draining of englacial reservoirs (Elliston, 1973; Collins, 1979). Thereafter, the discharge consists almost entirely of subglacial flow. During the April survey, the prolonged period of cold weather prior to the survey prevented supraglacial melting and allowed complete drainage of the englacial reservoirs. The observed flow should therefore mainly represent the subglacial component. In September, warm weather preceded sampling and, even if the addition by rain did not match the loss of supraglacial melt (from snow cover), the englacial reservoirs could still have maintained discharge over the sampling period. The flow rate of $3.0 \text{ m}^3\text{s}^{-1}$ therefore represents the sum of subglacial flow, englacial (originally supraglacial) melt, and precipitation. Observed dissolved loads are consistent with this explanation in being considerably larger for the mainly subglacial flow.

The variations in total dissolved load in either of the two surveys are not apparently dependent on discharge. This observation can be explained in two ways. First, the errors in discharge measurement are high enough to obscure any variation which could correspond to changes in dissolved load, at least in the April survey. Secondly, the variations in total dissolved load within either survey may reflect the complex nature of the subglacial hydrology: Vivian (1970) and Vivian and Zumstein (1973) have shown the existence of two major subglacial streams and a large subglacial reservoir in glacier d'Argentière. These different water components may exhibit some degree of variability in the duration and nature of their contacts with different rock minerals, and hence in their composition. Changes in the mixing ratios of these and other water components is a potential source of variation in bulk-melt water composition which is exacerbated by the abstraction of water for hydro-electric purposes.

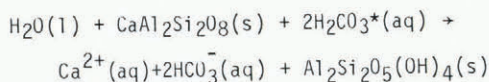
Chemical controls of melt-water composition

(i) *Ionic composition*

The large concentration of Ca^{2+} and HCO_3^- in the bulk melt waters are clearly compatible with CaCO_3 dissolution as the main mechanism of solute acquisition, since



where (g), (l), (aq), and (s) denote gaseous, liquid, aqueous and solid phases respectively. However, as Raiswell (1984) has shown, the processes of CaCO_3 dissolution and Ca^{2+} dissolution from silicates gives waters of similar composition;



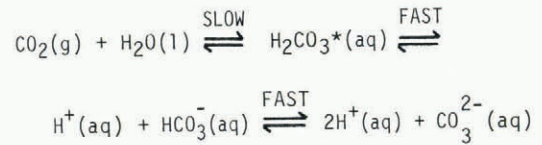
In an open system, the two processes cannot be distinguished. Nevertheless, CaCO_3 dissolution proceeds much more rapidly than silicate breakdown and is the more likely mechanism of solute acquisition, even taking into account the very low concentrations of CaCO_3 found in the analysed rock samples. Such a

situation occurred at Fjallsjökull (Raiswell and Thomas, 1984) where calcite only constituted 1-3% of the total rock but was probably the major source of solutes.

The common occurrence of Ca^{2+} -rich melt waters (Rainwater and Guy, 1961; Reynolds and Johnson, 1972; Slatt, 1972; Church, 1974; Collins, [1978], 1979) despite wide variations in glacial bedrock composition implies that CaCO_3 dissolution may often be the main process of solute acquisition even when calcite is only present in low concentrations.

(ii) *Values of $p(\text{CO}_2)$ and $(SI)_{\text{calcite}}$*

The process of chemical weathering involves the consumption of hydrogen ions from solution to release metal ions from rock minerals (shown above). The necessary hydrogen ions can be formed by the dissociation of dissolved atmospheric carbon dioxide



However the slowest step in this process is the rate of CO_2 transfer across the gas-liquid interface, which is frequently slower than the reactions (such as weathering and dissolution) which consume hydrogen ions (Stumm and Morgan, 1981). The relative rates of CO_2 -consuming reactions, as compared to CO_2 renewal, determine the nature of the chemical pathway followed by bulk melt waters; open-system conditions result where rates of consumption are less than rates of renewal, whereas rates of consumption in excess of renewal give closed-system conditions. The development of open- or closed-system conditions thus depends on those factors which affect rates of weathering (e.g. availability and reactivity of rock minerals) and the extent and rate of CO_2 transfer to the melt waters (e.g. time, turbulence). Open-system conditions can be recognized by $p(\text{CO}_2)$ values close to atmospheric levels ($10^{-3.5}$ bar) whereas considerably lower values may occur in closed systems (Raiswell and Thomas, 1984).

The $p(\text{CO}_2)$ values of the April and September bulk melt waters are generally close to equilibrium with atmospheric CO_2 and open-system weathering is indicated. The complex nature of the hydrological environment beneath the glacier is emphasized by the occurrence of low $p(\text{CO}_2)$ values (c. $10^{-5.5}$ bar) for the small subglacial streams. These streams have clearly evolved in a closed system, where rates of CO_2 consumption by weathering exceeded rates of CO_2 renewal by dissolution.

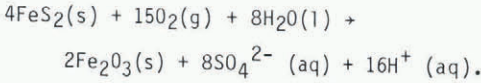
The saturation indices for the two sets of melt-water data (Table I) both indicate undersaturation with respect to calcite, although this is considerably greater for the September sample (-3.6) than for the recession-flow April samples (-1.7). This difference is consistent with a larger subglacial contribution to the latter, as the $(SI)_{\text{calcite}}$ values for the two subglacial samples (-0.44, -0.56) are comparatively high, in addition to being sufficiently close to saturation to indicate that regelation could induce supersaturation and subglacial precipitation of CaCO_3 (Hallet, 1976). The marked depletion of Ca^{2+} observed in the subglacial streams, in comparison with the bulk melt waters, is consistent with the occurrence of subglacial CaCO_3 precipitation but may also be due to ion exchange or chemical sorting effects, such as those observed by Souchez and Lorrain (1975).

(iii) *Dissolved load/pH relationships*

Under open-system weathering conditions, solutes are acquired according to the relationship (Raiswell, 1984)

$$\log[\Sigma^+] = \text{pH} - 11.2.$$

This relationship applies where dissolution of atmospheric carbon dioxide provides the major source of hydrogen ions consumed as a result of solute acquisition. In fact all the melt waters contain small concentrations of SO_4^{2-} , originating either from atmospheric precipitation or from the oxidative weathering of sulphides. Measurements of Cl^- were below $10^{-6} \text{ mol l}^{-1}$ and thus the molar ratio $\text{Cl}^-/\text{SO}_4^{2-}$ is much lower (c. 10^{-1} to 10^{-2}) than in average precipitation (c. 5.4; Garrels and Mackenzie, (1971). Sulphate has therefore been acquired as below:



Oxidative weathering of sulphides thus produces a 2:1 molar ratio of hydrogen ions to sulphate. Each mole of hydrogen ions generates one equivalent of cations in an open system, hence sulphate analyses can be used to correct values of Σ^+ for the extra solutes acquired using the hydrogen ions supplied by sulphate oxidation. This correction assumes that these hydrogen ions are used exclusively in weathering and is valid only as long as $[\text{HCO}_3^-] \gg [\text{SO}_4^{2-}]$ and concentrations of the latter are low. Sulphide oxidation without commensurate weathering may be recognized because the additional $[\text{H}^+]$ perturb the carbonate equilibrium to give $p(\text{CO}_2)$ values in excess of $10^{-3.5}$. Values of $p(\text{CO}_2)$ at Argentière are not significantly different from atmospheric, hence Σ^+ is corrected by using

$$\Sigma^+_{\text{plotted}} = \Sigma^+_{\text{measured}} - 2[\text{SO}_4^{2-}]$$

On this basis, sulphide breakdown supplies an average of 24% and 29% of the cation equivalents in the April and September melt waters respectively. Figure 2 shows $\log[\Sigma^+]$ plotted against pH, with the theoretical open system weathering line. Data from both Argentière surveys are plotted, together with literature data

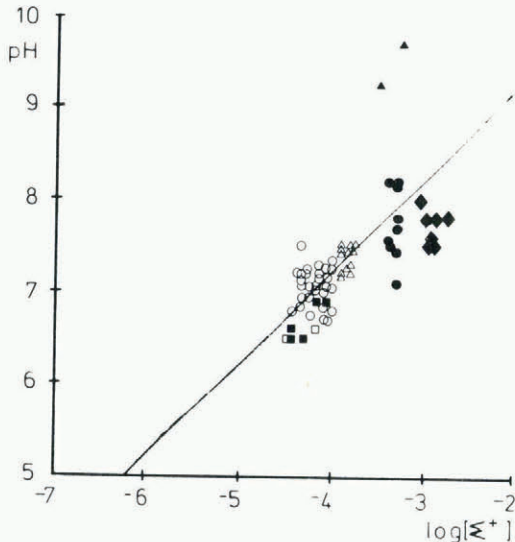


Fig. 2. Relationship between $\log(\Sigma^+)$ and pH. Theoretical open system relationship shown by straight line. Argentière data: bulk melt waters (●) and subglacial melt waters (▲) sampled in April 1975 and bulk melt waters (○) sampled in September 1976. Additional data from the Northern Cascades (■; Reynolds and Johnson, 1972), the Chamberlain Glacier (□; Rainwater and Guy, 1961), Les Bossons (◆; Thomas, unpublished) and Mer de Glace (△; Thomas, unpublished). All data corrected for the influence of solute acquisition by sulphide oxidation.

from Rainwater and Guy (1961), Reynolds and Johnson (1972) and Thomas (unpublished). Both sets of bulk melt waters from Argentière plot reasonably close to the open-system line, as also do the literature analyses. These data confirm that the Argentière melt waters evolved, at least latterly, in an open system and suggest that this mode of evolution may prove to be common.

(iv) Chemical controls of open- and closed-system behaviour

Two features of the Argentière data are worthy of closer examination, in relation to the chemical factors which determine whether open- or closed-system behaviour is exhibited.

1. Subglacial samples display closed-system characteristics and are close to saturation with $(SI)_{\text{calcite}} = -0.44$ to -0.56 , $(SI)_{\text{quartz}} \approx 0$ and other silicate-weathering products, such as kaolinite, gibbsite (Thomas, unpublished).

2. Recession-flow bulk melt waters (mean $\Sigma^+ = 0.61 \text{ meq l}^{-1}$) fall within the range of Σ^+ of subglacial samples taken in the same survey (0.46 to 0.68 meq l^{-1}) but display open-system characteristics. Recession flows are believed to consist largely of subglacial melt.

The first observation clearly demonstrates that the subglacial melt is close to equilibrium with typical weathering products and hence has little further capacity for solute acquisition. Any significant increase in dissolved load during transit through the glacier is unlikely, since rates of mineral dissolution decline to very low levels as equilibrium is approached. It seems likely that the uniform solute loads inferred for subglacial melt (Collins, 1979) represent similar, near-equilibrium values.

The second observation is apparently contradictory to the first, in suggesting that predominantly subglacial samples can display open-system characteristics. However the variations in subglacial composition and load are too large to exclude the possibility of deriving the recession-flow samples by mixing subglacial melt with water from another source. It is unlikely that this source could be englacial but a ground-water contribution could have the necessary characteristics of a relatively low pH and high $p(\text{CO}_2)$ (to produce an overall open-system mixture) and high solute load (to mix without dilution). An alternative explanation is that these mainly subglacial samples re-equilibrated with the atmosphere during their relatively slow transit through the glacier, but rates of CO_2 re-equilibration are slow enough for low $p(\text{CO}_2)$ values to persist for at least 30 min after first exposure to the atmosphere. The present data permit no distinction between the possibilities of ground-water mixing and CO_2 re-equilibration, although the former seems more probable.

These observations provide an indication of the processes which determine whether open- or closed-system behaviour occurs. Thus a general case can be made (Raiswell, 1984) to suggest that subglacial melt waters are close to equilibrium with their sediment load and hence reach a maximum solute load prior to mixing with englacial melt. By contrast, an englacial component similar in composition to supra-glacial melt is very undersaturated with respect to weathering products and can readily acquire solutes. Mixing to produce bulk melt waters can result in chemical reaction, provided reactive rock debris is available in the subglacial component. This results in closed-system conditions (Raiswell and Thomas, 1984) even at high discharge when the englacial component predominates. Open-system bulk melt waters will usually result if post-mixing reactions cannot occur (no reactive rock debris available). In this latter case, the subglacial contribution to the

carbonate equilibrium is often overwhelmed by mixing with large proportions of englacial water, so giving a bulk melt water with open-system characteristics (e.g. Argentière, September 1976 samples). However the recession-flow samples also suggest that open-system conditions may also arise after post-mixing reactions, if there is the possibility of mixing with ground-water or, less probably, sufficient time for re-equilibration with the atmosphere.

CONCLUSIONS

1. The nature of the dissolved load in bulk melt waters from glacier d'Argentière is essentially uniform under both autumn high-flow, and spring recession-flow, conditions. In both cases, Ca^{2+} (70-80%) and HCO_3^- (80-90%) are the major ions. The bulk melt waters are close to equilibrium with atmospheric CO_2 and display the characteristic open-system relationship between solute load and pH.

2. The magnitude of the dissolved load in bulk melt waters is considerably higher under recession-flow conditions, due to the large subglacial contribution, and the $(SI)_{\text{calcite}}$ is also larger than observed at high flow.

3. Subglacial melt waters are similar in their total dissolved load to the recession-flow samples, but are comparatively depleted in Ca^{2+} and enriched in Na^+ and K^+ . Subglacial samples have evolved in a closed system and are close to saturation with calcite. Recession-flow bulk melt waters could have evolved from subglacial melt by precipitation of CaCO_3 and either re-equilibration with the atmosphere plus continued weathering of Na- and K-bearing silicates or mixing with a Na^+ , K^+ -rich ground-water.

4. The development of open- or closed-system characteristics depends, in the first instance, on whether dissolution/weathering reactions occur when chemically aggressive englacial waters are mixed with solute and sediment-rich subglacial melt. Occurrence of dissolution/weathering reactions leads to closed-system conditions; otherwise open systems are observed. Such closed-system characteristics may be modified by re-equilibration with atmospheric CO_2 or ground-water mixing to give bulk melt waters with open-system characteristics.

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