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The characterization and role of aeolian deposition on water quality, McMurdo Dry Valleys, Antarctica

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ABSTRACT

The connection of ecosystems by wind-driven transport of material has become a topic of increasing interest and importance. Less than 1% of dust transported worldwide is exported to the Southern Ocean and Antarctic cryosphere; however, aeolian transport on the Antarctic continent is predominantly locally derived from the abrasion of bedrock. The deposition of the aeolian material is integral to nutrient and solute dispersal in the Antarctic ecosystem. This is particularly true in the ice-free areas of Antarctica, such as the McMurdo Dry Valleys (MDV), where aeolian material deposited in the aquatic system is solubilized during the melt season. The material is predominantly locally-derived from the abrasion of bed- rock. In this study, a two-step leaching experiment simulates the melt season and we quantify the flux of solutes and nutrients to the aquatic ecosystem. Soluble salts were removed from the aeolian material first during cold water leaching followed by an increase in carbonate and silicate dissolution during freeze– thaw. Major ion fluxes on glaciers and lakes are at least two orders of magnitude greater than nutrient fluxes. However, the fluxes derived from these experiments are less than the estimated flux from streams to lakes and probably represent minima. Aeolian redistribution of local soils is important because they are the only source of new solutes and nutrients to the aquatic ecosystem of the MDV.

Introduction

On the global scale the major source regions of aeolian material are concentrated in arid and semi-arid regions (e.g. the Sahara Desert of Africa and the Gobi and Takla Makan Deserts of China) characterized by low soil moisture and sparse vegetation that results in minimal particle cohesion and improved probability of deflation (Miller and Tegen, 1998). The major source regions emit 1–2 Pg of dust (up to ~50 μm in diameter) to the atmosphere each year (Harrison et al., 2001) that is transported through the atmosphere and integrated into the biogeochemical cycles of the ecosystems on which it is ultimately deposited (Jickells et al., 2005).

The ice-free areas of Antarctica, specifically the hyper-arid McMurdo Dry Valleys (MDV, Fig. 1), fit the broad description of an aeolian source area. The aeolian material locally transported within MDV is predominantly sand-sized (Lancaster, 2002; Speirs et al., 2008a), consistent with other cold climate regions (Sepälä, 2004). This larger average particle size is due to the fact that the locally-derived aeolian materials present in the MDV represent only a first-order physical weathering of parent material and regolith (Sepälä, 2004).

While aeolian emissions within the MDV region of Antarctica are small compared to other source regions (Li et al., 2008), the redistribution of new, reactive material by wind may be important to sustaining life in the ecosystem. The interface between the aeolian and fluvial sediment supply and its resulting geomorphological and ecological implications has only recently been investigated. Since the MDV receive very little precipitation, have very low soil moisture, and no overland flow, the interaction between the aqueous supply and aeolian system is more limited compared to more temperate climates. The MDV are considered a polar desert due to their extremely low mean annual temperatures and precipitation. Mean annual valley floor temperatures in the MDV range from −15 °C to −30 °C (Doran et al., 2002). Annual precipitation for the MDV is less than 5 cm water equivalent that falls as snow, most of which is subsequently sublimated before extensive melting can occur (Fountain et al., 1999, 2010). Liquid water only flows during a 4–12 week period of the austral summer when ephemeral streams connect the glaciers to the closed basin lakes through well-defined stream channels (Fig. 2) (Fountain et al.,
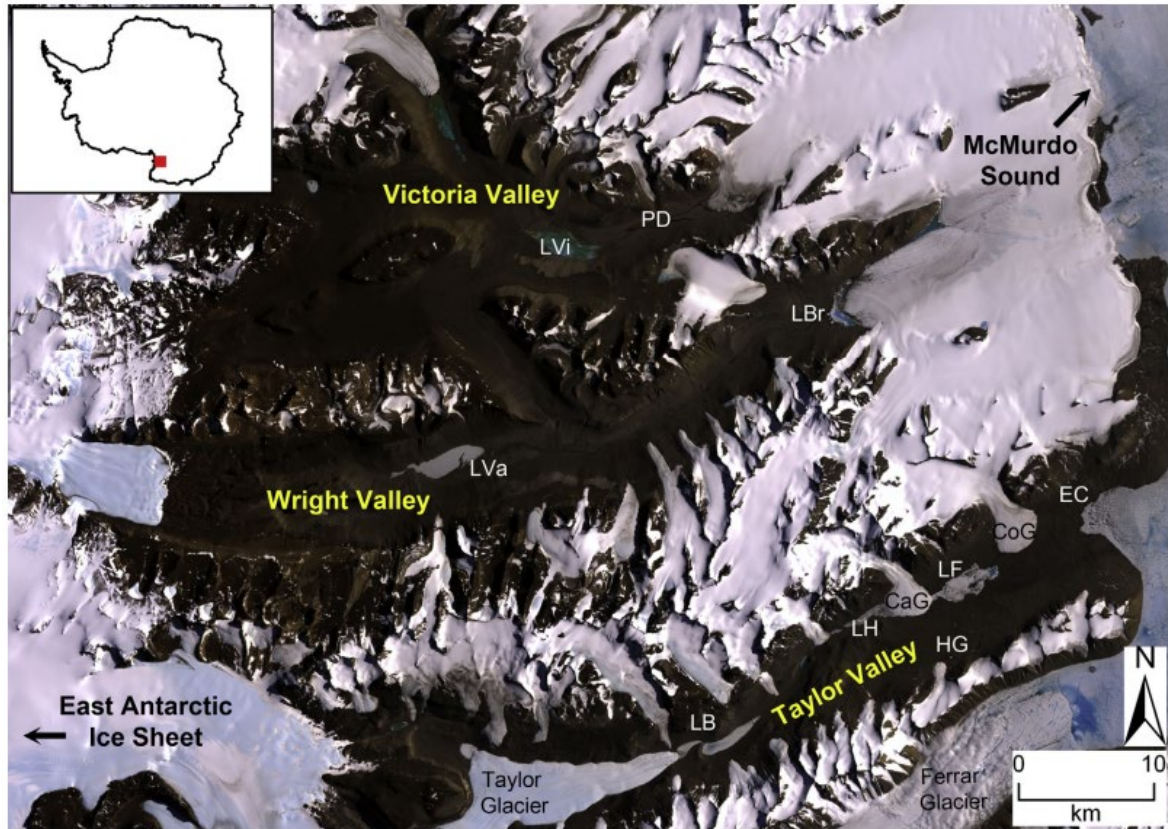
1999).

Fig. 1. Landsat (EMT+) image of the Dry Valleys region. The three major valleys are labeled in yellow. Locations of the collection of aeolian material mentioned in this study are abbreviated and labeled: Canada Glacier (CaG), Commonwealth Glacier (CoG), Explorer's Cove (EC), Howard Glacier (HG), Lake Brownworth (LBr), Lake Bonney (LB), Lake Fryxell (LF), Lake Hoare (LH), Lake Vanda (LVa), Lake Vida (LVi), Packard dune field (PD). Basemap image available from Landsat Image Mosaic of Antarctica (http:// lima.usgs.gov).

Sediment transport by two major wind regimes is an important physical process in this landscape and it is expressed in widespread aeolian landforms and deposits in the MDV (Fig. 1) (Lancaster, 2002). Topographically channeled, adiabatically warmed föhn winds that flow from the west off the ice sheet pre- dominate during the winter months and can reach speeds of greater than >37 m/s (Ayling and McGowan, 2006; Doran et al., 2002; Speirs et al., 2008b). These dry winds occur as strong, sporadic events that are responsible for the majority of sediment and organic matter redistribution within the MDV, especially near the valley floor (Šabacká et al., 2012). Easterly coastal breezes, prominent during the summer, advect into the valleys from Ross Sea and con- tribute salts and marine aerosols. These are generally cooler and moister than the föhn winds, allowing for increased precipitation (Fountain et al.,

1999). Additionally, the föhn winds are generally stronger than the coastal breezes accounting for the increased wind speeds closer to the polar plateau (average: 2.5 m/s near coast, 3.9 m/s near polar plateau) documented by Doran et al. (2002). These two wind regimes effectively ''recycle'' particles by transporting valley floor sediments onto glacial and lake ice sur- faces where they can then be solubilized and contribute solutes to the aquatic system (Lyons et al., 2003, 1998). These sediments can also change the albedo of the ice and eventually lead to the formation of cryoconite holes on glaciers that affect the composition of glacial melt (Fountain et al., 2004) as well as contribute to the surface topography of the permanent surface ice of the lakes (Squyres et al., 1991). Moreover, aeolian deposition in stream channels (Lyons et al., 1998) produces a new and changing source of material to be weathered during the melt season.
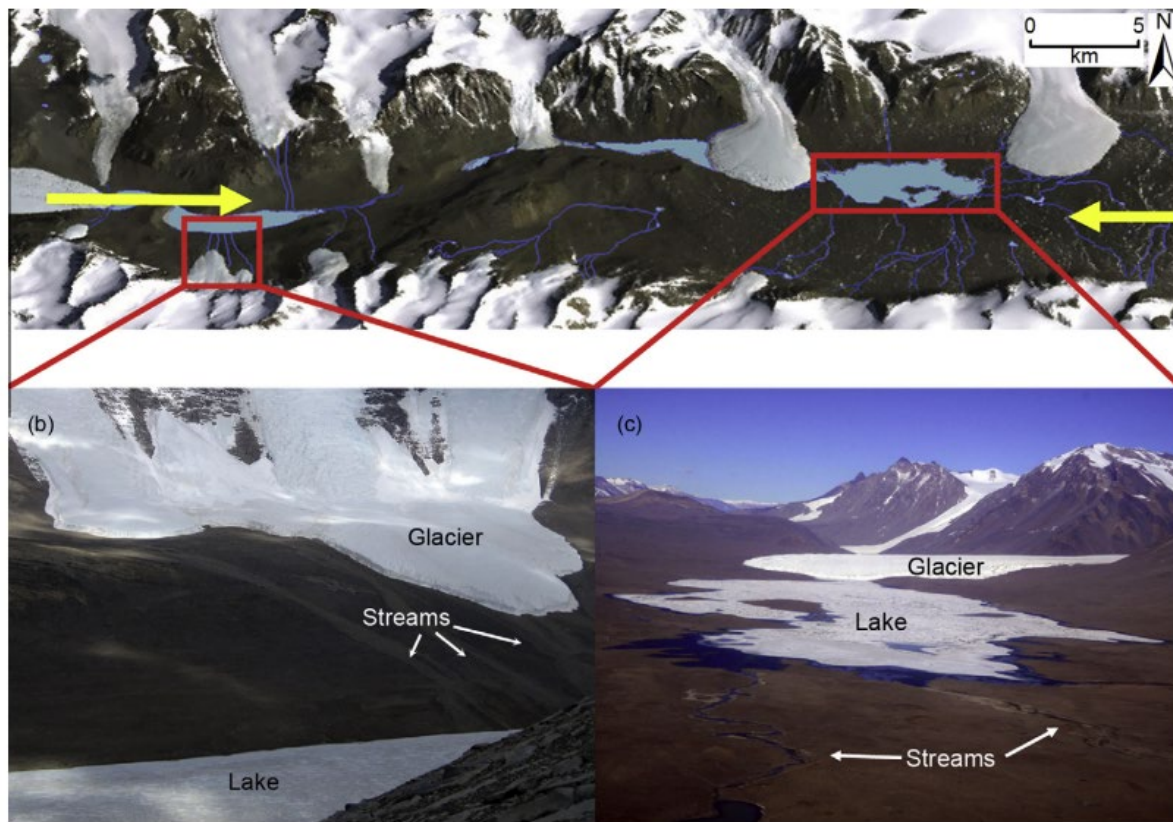

Fig. 2. The hydrological continuum present in the MDV. (a) The Taylor Dry Valley with streams hydrological features highlighted. Arrows represent foehn (westerly) and coastal (easterly) wind systems responsible for the redistribution of sediment and organic matter, as well as the geochemical evolution of meltwater. The continuum is illustrated at the highlighted sites at (b) the Hughes Glacier on the south side of the Bonney basin and (c) the Lake Fryxell basin. Hydrological system shapefiles, Landsat image, and 30 m digital elevation model available from http://www.mcmlter.org.

Previous studies have investigated the hydrochemistry of stream and lake waters, as well as the downstream solute patterns of the Taylor Valley aquatic systems (e.g. Green et al., 1988; Lyons et al., 2003, 1998; Nezat et al., 2001), but few have examined how the addition and dissolution of aeolian material could affect down-stream composition. The following study was designed to test whether the dissolution of aeolian material in the MDV ecosystem contributes a significant amount of solutes and/or nutrients to the aquatic environment.

Regional setting

Roughly 97% of Antarctica is covered by ice that may be greater than 3000 m thick. The remaining 3% of land area consists of ice-free regions, the largest of which is the MDV in the Transantarctic Mountains. The ice-free regions cover approximately 4500 km^2 (Levy, 2013) located at 76°30′–78°30′S, 160–164°E (Fig. 1). The MDV are situated perpendicular to the Ross Sea coast of southern Victoria Land and are characterized by hyper-arid landscapes made up of coarse soils derived from glacial tills reworked by aeolian processes, glaciers, and closed-basin lakes. These ice-free regions exist because the Transantarctic Mountains dam much of the eastward flow of the East Antarctic Ice Sheet into McMurdo Sound. The MDV have been the focus of the McMurdo Long-Term Ecological Research (MCM-LTER) program supported through the U.S. National Science Foundation since 1993 (Fountain et al., 1999).

Methods

Eighteen aeolian samples were collected from ice surfaces of lakes (Supplementary Fig. A.1) and glaciers (Fig. A.2), aeolian land- forms (Figs. A.3 and A.4), and elevated sediments traps (EST) arrays (Fig. A.5) in the MDV. Samples taken on 6 glacial surfaces and 5 lake ice surfaces were assumed to be aeolian in origin, if local, as the sample sites are out of range of any other likely source of sediment. Four landform samples were chosen based on previous work indicating aeolian origin (Packard Dune Field, Lake Hoare Beach, e.g. Nedell et al., 1987; Speirs et al., 2008a,b) or fine-grained sedimentation within ventifacts with visual mineralogical differences to host rock. The remaining three samples were collected from EST arrays located in each of

the major lake basins in Taylor Valley. As the EST sediments are completely removed from the aqueous system, these samples likely represent the actual mineral composition of MDV soils. Nitrile gloves were worn at all times when handling samples to minimize the risk of contamination. Samples were collected with a clean scoop and stored in clean Whirl-Pak bags.

A two-step leaching experiment was conducted on each of the 18 bulk, homogenized samples to ascertain the easily leachable pool of solutes from the aeolian material and its potential influence on the geochemistry of the aquatic system during the melt season. The experiment was designed to investigate the role of melt water and freeze–thaw mechanisms on the dissolution of material as originally described by Lyons and Welch (1997). Essentially, this experiment emulates the beginning of melt when both melt (day) and freeze–thaw (diurnal) mechanisms affect the flux of solutes/nutrients.

For the first leach ("Leach 1"), 25 g of sample sediment was placed in a clean HDPE sediment storage container and spread in an even layer. 50 mL of 18 Ω deionized water chilled to 3°C (pH assumed to be ~5.7) was carefully poured over the sediment, the container capped, and the slurry agitated for 5 min. The leachate was collected by filtering through a filter tower with 0.4 µm Whatman® Nuclepore track-etch membrane filter into clean 125 mL Nalgene® low density polyethylene bottles. Care was taken to de- cant the leachate so that as little of the sediment as possible was transferred onto the filter. This leaching step should approximate what the initial flux of nutrients and solutes at the beginning of the melt season.

The second leach ("Leach 2") was performed on the same sediment used in the first leach. Another 50 mL of 3 °C 18 Ω deionized water was added; then the container capped and placed in a −20 °C freezer. After approximately 72 h, the containers were removed and placed in a laminar flow hood to melt. Once all visible ice had melted, the leachate was again filtered into clean Nalgene® LDPE bottles as described above. This leachate represents what occurs over the course of the melt season as air temperature changes. All leachate samples were stored at 4 °C and analyzed within 48 h of collection.

Major cations (Na^+, K^+, Ca^{2+}, Mg^{2+}) and anions (NO_3^-; Cl^-; SO_4^{2-}) were analyzed

on a Dionex DX-120 ion chromatograph (IC) with an AS40 automated autosampler as originally described by Welch et al. (2010). Precision was determined to be ±5% for cations and anions. Alkalinity (HCO_3^-) was calculated through charge balance. The average error between calculated and measured alkalinity values in over 900 MDV stream samples from multiple summer sea- sons has been calculated to be ±14%, and is the best estimate for alkalinity precision (Lyons et al., 2012). Samples were analyzed for $NO_2^- + NO_3^-$, PO_4^{2-}, total nitrogen (TN), total phosphorous (TP), and reactive Si (H_4SiO_4) on a Skalar San++ Continuous Flow Analyzer with SA 1050 Random Access Autosampler. Precision was found to be ±5%. Only leachates from Leach 1 were analyzed for iron content via a Ferrozine method (Stookey, 1970). Precision was calculated to be within ±5%.

Results and discussion

Major ion data are recorded in Tables A.1 and A.2, respectively. Fig. 3 presents these data in ternary cation and anion plots. Leachates from both experiments are enriched in Ca^{2+}, Na^+, and K^+ relative to Mg^{2+}. ESTs are more enriched in Ca^{2+} relative to the alkali cations, but there is no discernible dominant cation for the other sample categories. The concentration of Mg^{2+} increases from Leach 1 to Leach 2 for all samples indicating a new or more readily available source of Mg over the course of the freeze–thaw experiment. In contrast, anion concentrations show no apparent spatial pattern or relationship based on sample type. Chloride and HCO_3^- generally occur at higher concentrations than SO_4^{2-}, however. There is a shift away from Cl⁻ to HCO_3^- from Leach 1 to Leach 2 at all sites, though this does not necessarily indicate a change in the dominant anion at a single site. While these shifts are recognizable in the data, the total dissolved solid concentration in the leachate represents less than 1% dissolution of the mass of the aeolian material as a whole over the course of the two leaching experiments (Table 1).
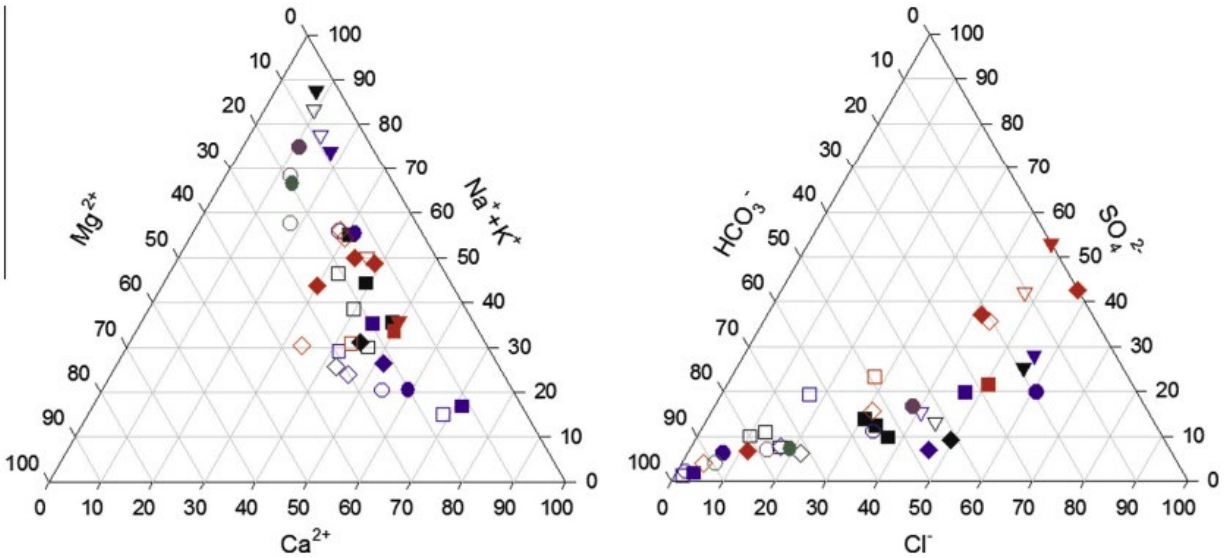
Fig. 3. Cation and anion variation diagrams. Basins/locations are color-coded (black = Fryxell, blue = Hoare, red = Bonney, green = Victoria Dunes, purple = Bull Pass). Sample types are distinguished by shape (square = glacier, circle = landform, diamond = lake, triangle = EST). Filled characters are from Leach 1, unfilled characters are from Leach 2.

To ascertain the primary solute source in the leachates, the influence of marine aerosols and salts was investigated through the comparison of Na:Cl concentrations to the seawater ratio (Fig. 4). The majority of the leachates lie close to the halite dissolution line except at the lowest concentrations where an excess of Na+ is noted, indicating another source of Na+ besides primary marine aerosol and/or salt dissolution. The majority of non-sea salt Mg2+, Ca2+, and Na+ in the leachates can be explained by the dissolution of salts containing SO_4^{2-} and HCO_3^{-} (Fig. 5). This is supported by comparison of Ca^{2+} + Mg^{2+} and HCO_3^{-} (Fig. 6) suggesting that leachates are near the stoichiometric composition of carbonate dissolution.

H_4SiO_4 increased in concentration from Leach 1 to Leach 2 (Tables A1 and A2), indicating that the weathering of a silicate minerals may be proceeding to a greater extent due to the freeze–thaw process. Alternatively, this may be due to the longer length of time of water–sediment reaction time and is corroborated by the leaching studies carried out by Toner et al. (2013) that show an increase in salt concentration in sequential water leaches on MDV soils.

Table 1
Percent dissolution[a] of aeolian material for leachates from both experimental methods. Take notice of the different magnitudes in the units.

Sample type	Sample ID	Na (% x 10^{-2})		K (% x 10^{-2})		Mg (% x 10^{-2})		Ca (% x 10^{-3})		P (%x 10^{-3})		Fe (% x10^{-5})	
		L1[b]	L2[c]	L1	L2	L1	L2	L1	L2	L1	L2	L1	L2
Aeolian landforms	Hoare Beach	1.6	1.2	1.7	1.6	0.4	0.4	1.8	1.4	3.0	7.0	2.4	-
	Defile	28.6	12.4	6.6	2.8	1.3	0.7	5.2	2.0	2.2	1.4	87.9	-
	Victoria Dunes	15.8	12.9	2.8	4.4	0.5	0.6	0.5	0.7	-	2.0	0.0	-
	Bull Pass	15.4	9.2	0.8	0.6	0.4	0.4	0.5	0.4	3.3	3.1	3.5	-
Elevated sediment traps	Fryxell EST	57.5	22.8	6.0	2.5	1.6	1.0	3.0	1.5	13.3	15.3	-	-
	Hoare EST	65.2	22.8	7.6	3.0	2.4	0.8	6.9	1.8	7.6	18.4	-	-
	W. Bonney EST	73.7	28.2	8.8	3.8	9.3	2.4	47.0	9.5	0.9	-	-	-
Glaciers	E. Commonwealth	11.4	6.3	2.5	1.5	1.4	1.3	3.0	2.1	24.4	11.4	25.6	-
	W. Commonwealth	10.7	4.9	1.6	2.1	2.1	1.5	5.1	2.8	5.9	6.8	17.2	-
	E. Canada	4.3	2.3	1.0	1.6	1.0	1.0	2.9	1.8	4.3	3.5	39.0	-
	W. Canada	2.7	1.3	1.7	1.0	1.0	0.9	1.8	1.0	1.0	0.8	3.5	-
	Howard	2.2	1.4	2.5	1.7	0.9	0.9	5.0	3.5	2.6	3.3	11.0	-
	Taylor	4.1	2.3	1.1	0.5	0.5	0.5	2.7	1.4	0.8	-	-	-
Lakes	Lake Fryxell	8.2	3.63	4.9	2.6	2.9	2.1	6.1	3.3	13.6	13.1	30.7	-
	Lake Hoare	1.5	0.9	1.2	0.8	0.6	0.6	2.1	1.3	1.4	1.7	3.6	-
	Mummy Pond	12.6	7.7	3.9	2.1	2.1	1.1	5.4	2.3	0.8	1.1	4.7	-
	E. Lake Bonney	3.4	1.9	1.6	1.0	0.5	0.6	0.8	0.8	3.5	2.8	3.4	-
	W. Lake Bonney	39.7	12.7	5.3	1.7	3.2	1.1	9.9	2.2	5.3	1.8	8.7	-

[a] Percent dissolution = (ion concentration in leachate)/(element concentration in sediment) *100.
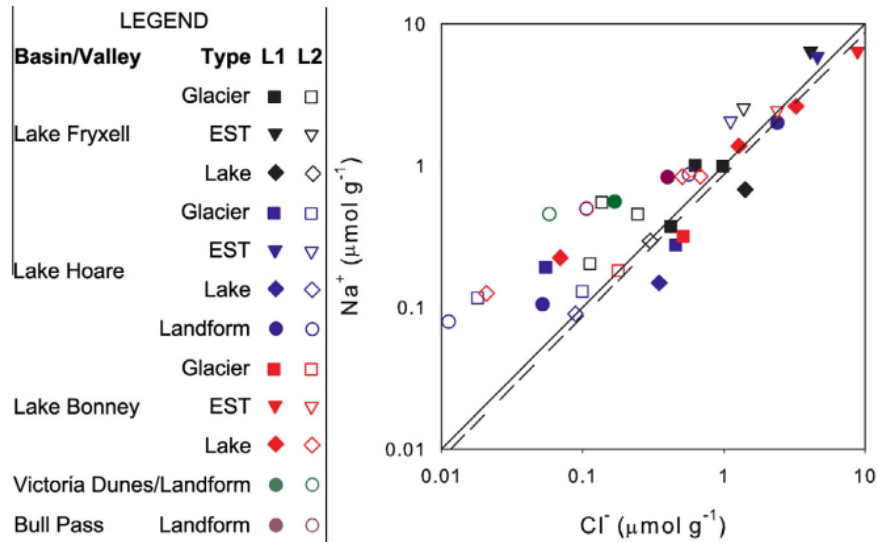[b] L1: Leach 1 (cold water).
[c] L2: Leach 2 (freeze–thaw).

Fig. 4. Na$^+$:Cl$^+$ ratios for both leach methods. Reference lines are the salt dissolution ratio (solid) and seawater ratio (dashed). Five percent error bars are smaller than size of marker.
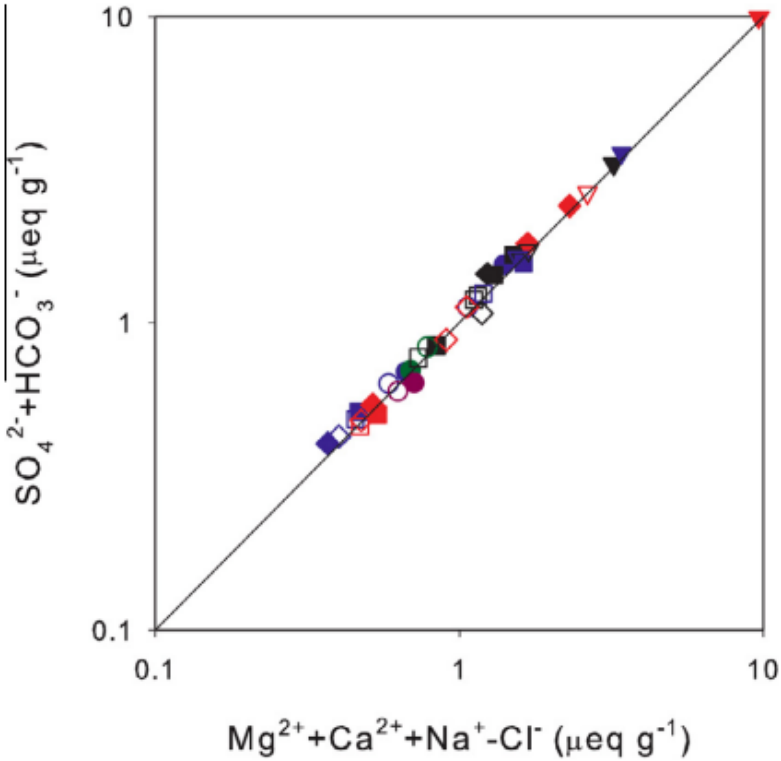
Fig. 5. $Mg^{2+} + Ca^{2+} + Na^+ - Cl^-$ vs $SO_4^{2-} + HCO_3^-$ for both leach methods. Reference line is the 1:1 line. See Fig. 4 for marker legend. Five percent error bars are smaller than size of marker.

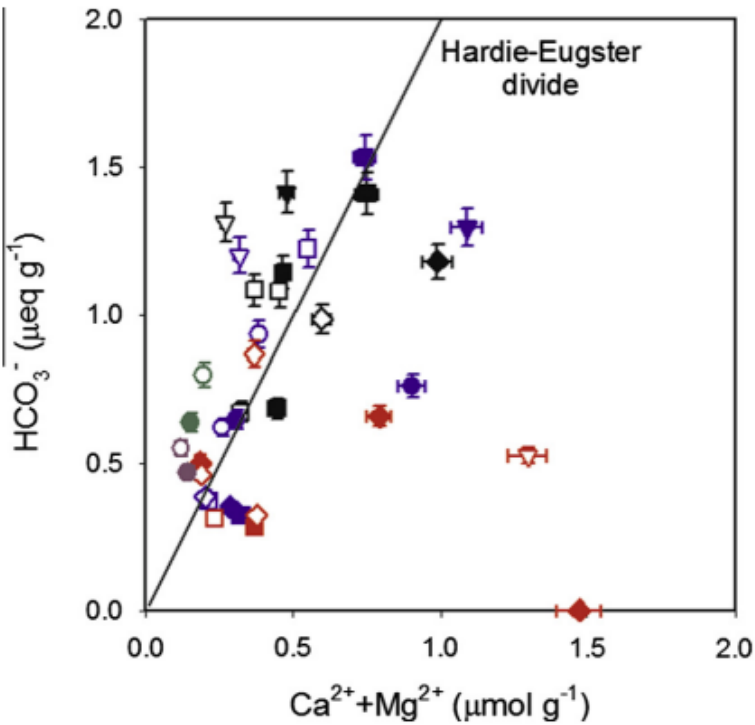

Fig. 6. $(Ca^{2+} + Mg^{2+})$: HCO_3^- ratios for both leach methods. Reference line is representative of the Hardie–Eugster divide for carbonate dissolution. See Fig. 4 for marker legend. Error bars represent ±5% instrument error.

Inorganic and total nutrient species concentrations are recorded in Tables A.1 and A.2 for Leach 1 and 2, respectively. Dissolved inorganic N and P species are at least an order of magnitude less than TN and TP. When plotted with the Redfield Ratio of balanced phytoplankton growth (16N:1P) for reference, a clear distinction is noted between total and inorganic nutrient species (Fig. 7). Inorganic N and P show dispersed patterns lying predominantly in the zone of P-limitation. Only the eastern lake basins of Taylor Valley (Fryxell and Hoare, Fig. 2) lie within the field of N-limitation. When TN and TP are considered, a different pattern occurs (Fig. 7b). There are two modes of TP concentration ($\sim$0.001 and $\sim$0.018 µmol g^{-1}) over a similar range of TN concentrations. In contrast to the inorganic N:P values, the majority of samples demonstrating N-limitation in TN:TP are from the western (Bonney) basin of Taylor Valley and the higher elevation Wright and Victoria Valleys. The majority of samples indicate that the Fryxell basin is predominantly P-limited, while Hoare basin samples can be either P- and N-limited. Soluble Fe^{3+} concentrations from Leach 1 are reported along with inorganic N and P values in Table A.1. Aqueous Fe follows no schematic pattern of enrichment in the leachates, but occurs in approximately the same concentration as inorganic P.

Chemical weathering of aeolian material

In the aquatic systems of the MDV, dissolved solute and nutrient redistribution occurs during the melt season, and our experiments suggest there is input from the dissolution of aeolian material as the concentration of solutes and nutrients in Leach 1, the initial cold water leach, is always greater than in Leach 2, the freeze–thaw leach (Table A.1 and A.2). Subsequent melt and/or freeze–thaw cycles allow for further dissolution of these materials in nature, and are supported by the continued dissolution in Leach 2 and leaching experiments in other studies discussed below (Gudding, 2003; Tegt, 2002). The values we report are a function of our leaching procedures and likely underrepresent the influence of aeolian deposition on the aquatic environment.
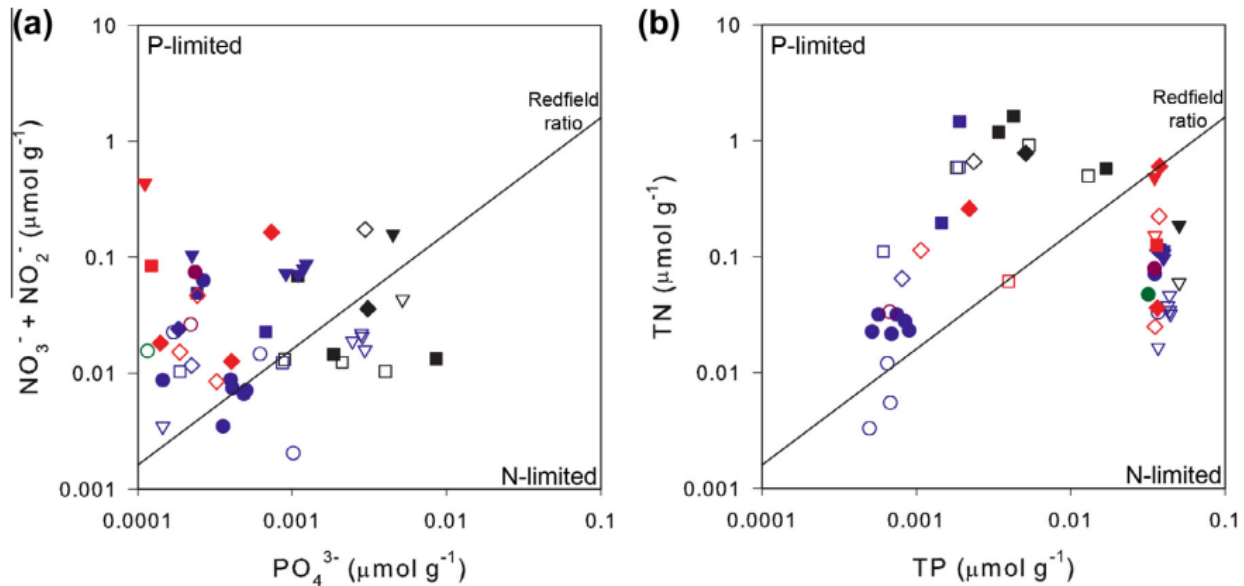
Fig. 7. (a) Inorganic and (b) total N vs P for both leach methods. Reference line is the Redfield molar ratio of 16N:1P with indications for P- and N-limited fields. See Fig. 4 for marker legend. Five percent error bars are smaller than size of marker.

The contribution due to aeolian deposition depends on the amount and type of material, particularly readily soluble salts, as well as the extent to which weathering can occur. The only source of solutes and nutrients to the leachate throughout the course of our experiment is from the minerals found in the aeolian sediments. The amount of Na, K, Mg, Ca, P, and Fe dissolved in the leachates is very small compared to the total mass of each element in the aeolian material available for leaching (Table 1), thus it is worth noting that not all bioavailable solutes and nutrients are being released during the two phases of leaching. For example, Na exhibits the greatest degree of dissolution regardless of leach method though this only is 0.7% of the mass of original material. In contrast, Fe demonstrates the smallest degree of dissolution (Table 1). Not surprisingly, the dissolution of major cations is 2–4 orders of magnitude greater than that of the more insoluble P and Fe. These values are significantly less than the values reported for soluble Fe (median 1.7%) and P (median 32%) in mineral dust collected in the Pacific Ocean (Baker and Jickells, 2006; Baker et al., 2006b). This decreased solubility of Fe and P in MDV aeolian material may be caused by the larger particle size (Baker and Jickells, 2006), lack of stratospheric processing (Baker et al., 2006a; Mahowald et al., 2005), and/or differences in the composition and reactivity of dust in the MDV compared to the aeolian material analyzed by Baker and others (e.g. Baker

and Jickells, 2006; Baker et al., 2006b).

There is no general geographic pattern within the MDV with regards to the extent of dissolution of aeolian material, but the data may suggest that landform elevation helps control the availability of solid material. Glacial surface samples all decrease in readily leachable solutes from east to west as the importance of marine aerosols becomes less important with distance from the coast (e.g. Fortner et al., 2005). In contrast, aeolian landforms and lake surfaces have different and alternating patterns of dissolution. This inconsistency appears to be at least qualitatively related to the grain size distribution, as finer particles make up a larger proportion of the bulk aeolian material at higher elevations (e.g. glacier surfaces).

The aeolian material collected from the ESTs generally demonstrate the greatest degree of dissolution and the greatest total dis- solved solids (Tables 1, A.1 and A.2), indicating an increased content of soluble salts or preferentially weathered minerals com- pared to the other sample types. The aeolian sediments collected in the ESTs are the only material studied that are completely removed from the aqueous system; both glacier and lake sediments are easily leached once ice melt begins effectively changing the overall mineral signature. Thus, as mentioned previously, the EST sediments may be a signature of the "true" mineral content of MDV soils.

The overall chemical composition of the snow and ice on the Commonwealth, Canada, Howard, and Taylor Glaciers of Taylor Valley (Fig. 1) differ and it has been speculated that at least part of this difference is due to the input and dissolution of aeolian material (Lyons et al., 2001). Marine aerosol Cl is more important in the east near the coast and stratosphere-derived NO_3 becomes more important further inland and with elevation (Keys and Williams, 1981). Because various glaciers within the MDV provide melt to the streams and lakes, the actual degree of dissolution as well as the amount of aeolian material that is deposited on each glacier is, therefore, important to the distribution of solutes and nutrients throughout the hydrologic continuum of the MDV. Fortner et al. (2005) found the influence of salts to be more important on the eastern (coastal) side and calcareous dust to be more important on the western (inland) side of the Canada Glacier. This was also reflected in the chemistry of streams, as streams originating on the western sides of glaciers are more impacted by aeolian in-

put by the strong down valley winds (Lyons et al., 2003). These findings are supported by the earlier work on the salt content in the MDV soils that noted an increase in Ca and Mg-bearing salts and a decrease in Na-bearing salts from the coast to inland areas (Keys and Williams, 1981).

The associations between major chemical species provide evidence that salt ($NaCl$, Na_2SO_4, $CaSO_4$) and carbonate dissolution are more important than silicate weathering in both sets of leaching experiments. Deviance from the 1:1 relationship between non-sea salt cations and $HCO_3^- + SO_4^{2-}$ in Fig. 5 indicates the presence of K^+ or NO_3-bearing salts and/or silicate mineral weathering (another source of K^+), as referenced by the very small overall concentrations of K^+; NO_3^-; and H_4SiO_4 compared to the other major ions that are generally an order of magnitude greater (Tables A.1 and A.2). Therefore, the solubilization of salts and carbonate mineral dissolution are likely the dominant processes occurring in these experiments compared to silicate weathering. Since silicate mineral weathering is known to occur in MDV streams (Gooseff et al., 2002; Nezat et al., 2001) and we see an increase in H_4SiO_4 concentrations from Leach 1 to Leach 2, the importance of silicate weathering may become more important over the course of the melt season as the preferentially weathered salts/carbonates are removed and numerous freeze/thaw cycles occur.

The occurrence of Na-bearing salts decreases with distance from the coast and occurs on all rock types (Keys and Williams, 1981). This is also reflected in the leachates from glacial and EST samples of this study that show increasing Na with proximity to the coast. One of the most common Na-salts in the MDV is halite originally derived from sea-salt (Campbell and Claridge, 1987; Claridge and Campbell, 1977). Mirabilite ($Na_2SO_4 \cdots 10H_2O$) is also common in some areas (Black and Bowser, 1968). At higher concentrations of Na^+ and Cl^- the stoichiometric dissolution relation- ship of $NaCl$ is strong (Fig. 4). However, the majority of samples from Leach 1 and all of the samples from Leach 2 have Na^+ concentrations greater than Cl^- indicating that there is a source of Na^+ ions other than sea salt aerosol or halite dissolution. Silicate, sulfate, and carbonate mineral dissolution are possible sources of this excess. In particular, sulfate and carbonate dissolution based on the relationship of non-chloride salts of Mg^{2+}, Ca^{2+}, and Na^+ account for the majority of

SO_4^{2-} and HCO^- in these leachates (Fig. 5).

Relatively high quantities of Ca^{2+} (Fig. 3) are attributed to carbonate and sulfate mineral dissolution, which has also been documented in the MDV streams (Lyons et al., 2002; Nezat et al., 2001). It has been suggested that the equivalent ratio of SO_4^{2-} to HCO_3^- in proglacial streams reflects the dissolution of gypsum relative to carbonates (Fortner et al., 2005): values greater than 0.5 reflect a greater degree of sulfate mineral dissolution compared to carbon- ate. Ratios increase to the west in leachates of both methods (Table 2) and reflect the relationship of inland gypsum increase noted by Keys and Williams (1981) in the Taylor Valley.

Fig. 6 demonstrates the importance of carbonate dissolution during the course of the leaching experiments. All samples in Leach 2 indicate that HCO^- is more proportionally important when com- pared to Leach 1. This is corroborated by the evolution of leachate composition from $Na^+ + K^+$- and Ca^{2+}-rich solution in Leach 1 to one with an increased Mg^{2+} content in Leach 2 (Fig. 3). These relationships also suggest that much of the Mg^{2+} in solution is due to the dissolution of a Mg-bearing carbonate phase. Marble is present the Precambrian metamorphic complex that is largely exposed in the Victoria Valley and western Taylor Valley (Peterson and Marsh, 2008) in addition to the limestones of the Ross Supergroup (Hask- ell et al., 1965). However, dolomite ($CaMg(CO_3)_2$) and magnesite ($MgCO_3$) are not commonly found in the MDV soils (Keys and Williams, 1981) suggesting that CaMg carbonate dissolution relation- ship (Fig. 6) may be predominantly due to Ca^{2+}. The generally higher concentration of HCO_3^- in Leach 2 compared to Leach 1 supports the occurrence of carbonate weathering. Other potential sources of Mg^{2+} are the weathering of volcanic rock particles rich in Mg, which are abundant in certain areas of the MDV. Within the Taylor Valley, Mg-bearing salts are associated with mafic igneous rocks and increase with distance inland (Keys and Williams, 1981). Mg-bearing salt dissolution or silicate weathering, there- fore, would only occur where these mafic rocks closer in composition to the Ferrar Dolerite and McMurdo Volcanic basanite, the two major mafic rock types in the MDV, have been abraded and trans- ported. Given the association of Mg-salts with the mafic igneous rocks, silicate weathering is likely the major source of Mg^{2+} in the

leachates. This is supported by the increase in H_4SiO_4 and de- crease in SO_4^{2-} in Leach 2 after all of the easily soluble salts have been removed by Leach 1.

Table 2
Ratio of SO^{2-} to HCO^- for both experimental leach methods.

Sample type	Sample ID	$SO_4^{2-} : HCO_3^{-a}$	
		Leach 1	Leach 2
Aeolian landforms	Hoare Beach	0.1	0.0
	Defile	1.0	0.2
	Victoria Dunes	0.1	0.0
	Bull Pass	0.4	0.1
Elevated Sediment Traps	Fryxell EST	1.3	0.3
	Hoare EST	1.8	0.4
	W. Bonney EST	>10	4.1
Glaciers	E. Commonwealth	0.2	0.1
	W. Commonwealth	0.2	0.1
	E. Canada	0.2	0.1
	W. Canada	0.6	0.3
	Howard	0.0	0.0
	Taylor	0.8	0.5
Lakes	Lake Fryxell	0.2	0.1
	Lake Hoare	0.2	0.1
	Mummy Pond	1.7	0.3
	E. Lake Bonney	0.1	0.0
	W. Lake Bonney	>2.5	1.7

[a] Calculated from equivalents.

The presence of reactive Si in the leachates indicates that silicate mineral weathering is occurring. However, the concentration of H_4SiO_4 increases from Leach 1 to Leach 2, indicating that silicate weathering is proceeding to a greater extent after initial salt con- tent is removed in Leach 1. In experimental work, increasing cycles of freeze–thaw lead to greater precipitation or loss from solution of Si (Dietzel, 2005), however this is not what we have observed in our experiments. Conversely, H_4SiO_4 content increased with time over a 24-h period in a leaching experiment on sediments from Lake Chad and the Canada Glacier (Tegt, 2002). The increased amount of H_4SiO_4 in the leachates of Leach 2 compared to Leach 1 implies, again, that silicate mineral weathering is likely the source of the increased Mg^{2+} in Leach 2. This is supported by the fact that mafic minerals in general have a higher weathering potential compared to more felsic minerals.

It is possible that the increase Mg^{2+}; HCO_3^-; and H_4SiO_4 observed in Leach 2 could be due to an increase in surface area from freeze–thaw particle fracture thereby

allowing more water contact with fresh mineral surfaces, but this could not be demonstrated. Alternatively, the increase in these ion species may be due to the removal of more soluble salts in Leach 1 leaving less soluble species to be solubilized in Leach 2. This idea of solute separation is supported by the flux of nutrients and solutes seen in glacial melt, presumably from particulate matter accumulation on ice and in streambeds during the winter, at the beginning of the melt season (Fountain et al., 1999). To address what processes are actually producing these observations, further experiments with different leaching protocols must be carried out. Nevertheless, the dissolution of MDV aeolian material is a significant process that proceeds throughout the melt season.

Nutrient (N, P) abundance and limitation

The concentration of nutrients in both Leach 1 and 2 are of the same magnitude, though Leach 1 generally had slightly higher concentrations than Leach 2 (Tables A1 and A2). The ratio of inorganic to total N and P varies throughout the MDV landscape and is greater in the aeolian landforms and EST leachates compared to the glacial and lake surface leachates. The low inorganic nutrient values compared to total N and P indicate a greater abundance of soluble organic N and P present in these samples (Fig. 7). This observation is possible since the margins of the terminal lakes and ponds in the MDV, and low gradient streams can be inhabited by algal and moss communities (McKnight et al., 1998). Similarly, glaciers are inhabited by diverse microorganisms that are found both on the snow and ice, as well as within cryoconite holes melted into the glacier surface that may continue to grow for years (Bagshaw et al., 2007).

Clearly the ultimate source of inorganic N and P in the MDV ecosystem is different: N is supplied through aeolian input derived primarily from the stratosphere (Vincent and Howard-Williams, 1994; Witherow et al., 2006) or marine mammal and avian excrement (Laybourn-Parry and Pearce, 2007), while P is derived from weathering of P-bearing minerals in bedrock (Barrett et al., 2007; Gudding, 2003). The extent to which these nutrients are available is dependent on landscape age and the lithology of tills and bedrock present (Barrett et al., 2007). Older landscapes have higher amounts of atmospherically-derived inorganic N; younger, less weathered

landscapes have a greater available pool of inorganic P. In the MDV, NO_3^- concentration generally increases with elevation and surface age. The climate at elevation (>800 m) in the western MDV is characterized by mean annual temperatures (MAT) < -27 °C, relative humidity (RH) < 45%, and rarely receives precipitation or meltwater (Marchant and Denton, 1996). In these conditions and the absence of melt, NO_3^- is able to accumulate to greater concentrations in the soil compared to the more temperate, lower elevation coastal climates (MAT = -17 °C, RH = 75%) and intermediate climates (MAT = -25 °C, RH = 10-70%) (Keys and Williams, 1981). All of the samples collected for this study are within the coastal and intermediate zones as defined by Marchant and Denton (1996). This relationship is reflected in NO_3^- and Cl⁻ concentrations from our leachates and both high and low elevation soils plotted for reference shown in Fig. 8. The leachates of aeolian material have higher concentrations of NO_3^- relative to low elevation soils and depleted relative to high elevation soils, which strongly suggests that the aeolian materials are derived from a mixed source of valley floor and higher elevation.

The overall heterogeneity of nutrient availability can be attributed to the complex geology of the MDV (Campbell and Claridge, 1987), the spatial availability of solar radiation and water (e.g. Gooseff et al., 2003), and the relative age of the surface. Spatial trends of inorganic nutrient ratios in Taylor Valley aeolian sediments follow those that have been demonstrated by Priscu (1995) for the terminal lake nutrient limitation: Lakes Fryxell and Hoare are N-limited based on stoichiometry while Lake Bonney is P-limited. That the aeolian sediments and lakes show the same trends indicates that aeolian transport in the MDV is local in scale. The youngest, most recently glaciated area of the Taylor Valley is the Lake Fryxell basin. While the weathering of igneous complexes containing high-P minerals (monazite, apatite) provide inorganic P throughout the MDV, the younger Fryxell basin has a greater amount of P-species in its soils indicating that chemical weathering proceeds to a greater extent and/or the weathered P is retained in soils there (Blecker et al., 2006). In contrast, P-limited Lake Bonney lies in the older, western portion of the Taylor Valley. The older surfaces in this region have been able to accumulate

stratospheric N for a longer period time (Vincent and Howard-Williams, 1994; Witherow et al., 2006), but also do not demonstrate the same P-species weathering and retention noted in the eastern portion of Taylor Valley (Blecker et al., 2006). Younger surfaces that demonstrate N-limitation and old surfaces that demonstrate P- limitation are supported by current models of landscape evolution (Vitousek et al., 1997).
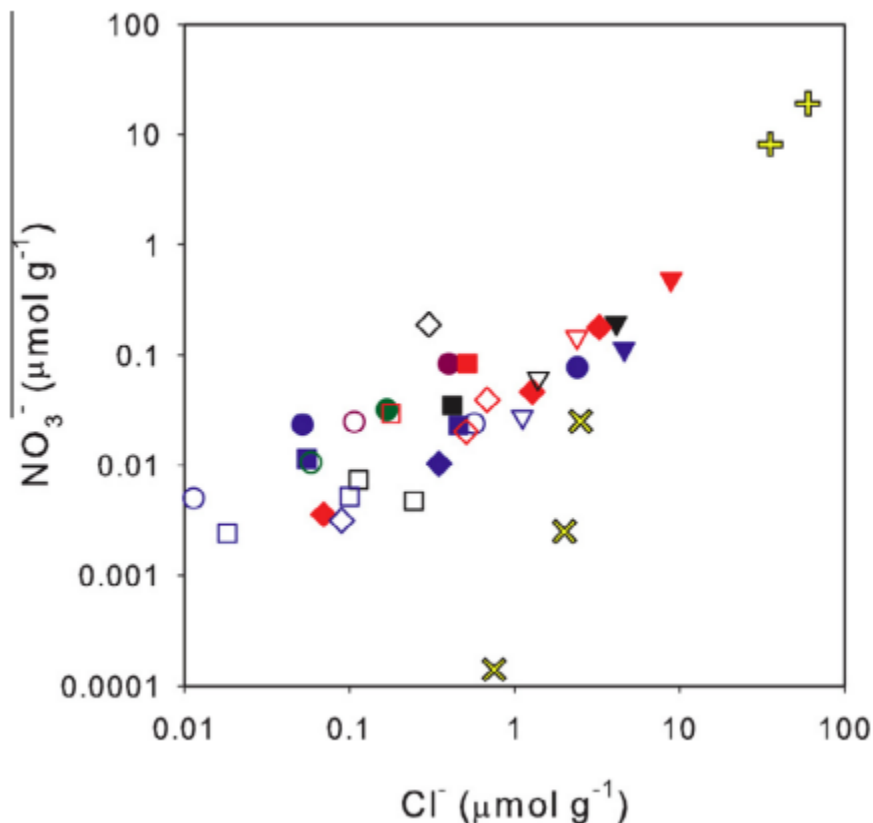

Fig. 8. NO_3^- : Cl^- ratios for both leach methods. See Fig. 4 for marker legend. Five percent error bars are smaller than size of marker.

Iron is a micronutrient important to photosynthesis and, there- fore, primary production (Martin et al., 1991). *In situ* iron fertilization experiments in the ocean have demonstrated that iron is the limiting nutrient, particularly in high nitrate low chlorophyll zones (e.g. Watson et al., 2000). However, only ~0.5% of dust-derived iron is bioavailable/dissolved upon direct deposition in seawater (Falkowski et al., 2000; Mahowald et al., 2005; Ridgwell, 2002). This can be increased up to 80% with atmospheric processing, but the mechanisms behind this increased solubility are unclear (Mahowald et al., 2005). Similarly, aeolian material from the MDV deposited on sea ice may stimulate growth in McMurdo Sound (Atkins and Dunbar, 2009). Within

the MDV, the total leachable Fe concentrations at each sample site are within one order of magnitude of the inorganic P concentration (Table A.1) indicating that Fe is not the limiting nutrient in these environments based on the Redfield Ratio of 16N:1P:~0.001Fe (Martin, 1990). Variations in concentration in the leachates may be due to the collection of varying amounts of material consisting of a relatively higher percentage of more mafic or Fe-rich source material.

Flux of solutes and nutrients to the MDV ecosystem

Annual fluxes of solutes and nutrients were calculated for the various sample types based on 1 m flux data from ESTs (Table A.3), the surface area of lakes (Lyons et al., 2000) and glacier ablation zones (Basagic, personal communication). These data are presented for solutes and nutrients in Tables 3 and 4, respectively. Taylor Glacier is currently not included in the flux calculations be- cause the ablation zone is extremely large and drains into several valleys (Basagic, personal communication). Aeolian landforms were also excluded from the flux calculation because their individual area-normalized sediment fluxes and surface areas are not known. More importantly, it is unclear if this material contributes to the solute/nutrient fluxes to the aquatic system except in unusual conditions.

The origin of solute and nutrient fluxes to the glaciers and lakes are most dependent on the aeolian sediment flux, which is highest at Lake Bonney. This inland increase in sediment flux is related to the föhn winds that are strongest in the Lake Bonney basin. Thus, solute and nutrient fluxes are greatest in the Bonney basin (Table 3 and 4). The higher solute flux at Lake Fryxell relative to Lake Hoare is likely related to its position and influenced by of the coastal wind system (Lancaster, 2002). Calculated glacier solute and nutrient fluxes are highest in the Fryxell basin at the Commonwealth Glacier. This relative increase is related to the ablation zone surface area that is five times larger than the Canada Glacier with the next largest ablation zone. However, the NO$^-$ flux is greatest on the eastern Canada Glacier that also drains into Lake Fryxell.

Table 3
Fluxes of solutes to the MDV ecosystem from glaciers and lakes based on sediment fluxes and feature surface areas.

Sample type	Sample ID	$Na^+(ga^{-1})$	K^+ (ga^{-1})	Mg^{2+} (ga^{-1})	Ca^{2+} (ga^{-1})	Cl^- (ga^{-1})	SO_4^{2-} (ga^{-1})	H_4SiO_4 (ga^{-1})
Glaciers	E. Commonwealth	190	46	37	100	140	110	48
	W. Commonwealth	170	67	48	170	230	92	57
	E. Canada	24	14	10	40	34	22	14
	W. Canada	17	9	8	26	36	27	5
	Howard	17	18	12	100	6	6	12
Lakes	Lake Fryxell	160	96	104	280	430	120	11
	Lake Hoare	10	7	7	23	28	8	4
	E. Lake Bonney	2900	1400	1600	2800	1200	1100	1100
	W. Lake Bonney	8600	1100	1300	6000	15000	16000	350

Table 4
Fluxes of inorganic nutrients to the MDV ecosystem from glaciers and lakes based on sediment fluxes and feature surface areas.

Sample type	Sample ID	N (ga^{-1})	P (ga^{-1})	Fe (ga^{-1})
Galciers	E. Commonwealth	-	2.0	0.77
	W. Commonwealth	0.34	0.64	0.51
	E. Canada	1.1	0.11	0.39
	W. Canada	0.72	0.02	0.03
	Howard	0.46	0.11	0.16
Lakes	Lake Fryxell	19	1.3	1.2
	Lake Hoare	0.34	0.02	0.03
	E. Lake Bonney	18	8.3	8.6
	W. Lake Bonney	330	3.3	5.8
Lake sediments[a]	Lake Fryxell		22	
	Lake Hoare		0.32	
	Lake Bonney		490	
Total flux from streams[b]	Lake Fryxell	2.6×10^4	9.7×10^3	
	E. Lake Bonney	7.3×10^3	880	
	W. Lake Bonney	1.3×10^5	1.1×10^4	

[a] 1:50 sediment to DI water extract (Gudding, 2003).
[b] Annual average, 1994–2002 (Foreman et al., 2004).

Aeolian nutrient flux has been discussed previously in regards to sediment composition in the lake basins of Taylor Valley and is calculated in the same manner as those reported here (Barrett et al., 2007). The soluble inorganic nutrient fluxes calculated here based on aeolian fluxes are ~6 orders of magnitude less than the total nutrient flux of MDV aeolian sediments (Barrett et al., 2007). Moreover, inorganic N and P values are two or more orders of magnitude less than the inorganic N and P load from streams to lake surface water (Table 4) (Foreman et al., 2004). The latter result suggests that on an annual basis, the solubilization of N and P from aeolian material is not a major nutrient source to the MDV lakes.

The total flux of solutes/nutrients from dust solubilization to any of the lake basins, but especially the Bonney basin, is an underestimation because not all the glaciers that could potentially con- tribute to the nutrient flux are included in the calculations. There are a larger number of glaciers that drain into the Bonney basin compared to the Fryxell and Hoare basins. Other geomorphological constraints, such as stream gradient and hyporheic zone interaction (Gooseff et al., 2002; Nezat et al., 2001), affect the degree of chemical weathering and thereby the flux of nutrients and solutes into the MDV ecosystem. Also, the presence or absence of biological communities within the streams can greatly affect nutrient concentration and fluxes (McKnight et al., 1999). Furthermore, the fluxes presented here represent only the result of our experimental design as opposed to the reality of aeolian material in contact with liquid water throughout the summer melt season.

Streamflow is primarily dependent on austral summer air temperature and can change tenfold in the course of a few hours (Ebnet et al., 2005), thus the actual flux of solutes and nutrients is also related to temporal climate variability as manifested in streamflow amounts and variations and these can change dramatically from year to year (Foreman et al., 2004). Previous studies have utilized experimental leaching to understand potential nutrient availability to the aquatic system from aeolian sediments deposited on the sur- faces of Lakes Fryxell, Hoare, and Bonney (Gudding, 2003). These experiments combined 1 g of lake surface sediment and 50 mL (1:50) DI water and allowed it to react for 16–24 h (Gudding, 2003), which resulted in fluxes of P at least 3 orders of magnitude less than calculated fluxes of P from streams into the MDV lakes (Table 4). The results of Gudding (2003) are also 3 orders of magnitude greater than those from this study (Table 4), and are likely a function of the length of reaction. That is, extracts from the 1:50 dilution left to react for longer yield greater fluxes than those at 1:2 dilution only reacted for 5 min. The actual flux due to the solubilization of aeolian material is probably greater than those measured here. Nevertheless, these calculations provide an initial estimate of solute and nutrient flux due to the dissolution of aeolian sediments. The calculated fluxes indicate that a small amount of solutes and nutrients were released during the course of our experiments. If these experimental results reflect the extent of aeolian dissolution, then the influence of dust dissolution to

the aquatic systems of the Taylor Valley is rather insignificant, except perhaps in the cryoconite holes on the surface of glaciers (Bagshaw et al., 2007). However, the greater fluxes achieved at a lower dust to DI water ratio (Gudding, 2003) and the increase in solute dissolution found in DI extracts over time (Tegt, 2002) indicate that the actual influence of dust dissolution could be much greater than indicated here.

Terrestrial system impact

While the dissolution of the aeolian sediment described here is relatively small in the overall mass flux of solutes to the lakes, the concentration of solutes and nutrients leached from the sediments is similar in both leach methods. This is significant because chemical weathering/dissolution over the flowpath of water is the only mechanism available to disperse solutes, and more importantly nutrients, to the MDV aquatic ecosystems. Total nutrient content of the leachates of both methods indicate an organic nutrient source to the glacier and lakes. Trends in inorganic (bioavailable) nutrients of Taylor Valley leachates indicate that nutrient limitation follows the pattern of limitation in the terminal lakes and their basins of the Taylor Valley; that is, Lake Bonney is P-limited while Lakes Fryxell and Hoare are co-limited by N and P (Priscu, 1995). The föhn winds from the ice sheet deposit aeolian material to ice surfaces and streams beds providing new, more reactive material for leaching by meltwater. These dissolution/chemical weathering processes proceed throughout the melt season, redistributing solutes and nutrients, and helping support the biota of the MDV eco- system. This transport of solutes, via wind deposition and later solubilization, to the closed-basin lakes was a heretofore described although unquantified process.

Conclusions

The amount of aeolian dust that is deposited on the Southern Ocean and Antarctica represents ~3% of the estimated Southern Hemisphere dust flux (Li et al., 2008). In the MDV, aeolian material is derived locally from the exposed bedrock abraded by wind. Aeolian deposition on ice surfaces and streambeds, occurring most extensively during the austral winter, is a source of nutrients and solutes to the aquatic

system during the melt season. Experimental leaching experiments have revealed that overall bioavailable nutrient and solute contribution to the aquatic system is important and changes depending on degree of leaching. Since the wind-driven redistribution of soils is the only source of new solutes and nutrients, it therefore follows that aeolian deposition is important in sustaining the aquatic ecosystem.

Acknowledgements

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.aeolia. 2014.01.002.

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