



# The generation and redistribution of soil cations in high elevation catenas in the Fraser Experimental Forest, Colorado, U.S.

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## ABSTRACT

Pedogenic processes imprint their signature on soils over the course of thousands to millions of years in most soil systems. Variation in soil forming processes – such as parent material weathering, organic material additions, hydrologic processes, and atmospheric additions – account for the distribution and sourcing of cations in ecosystems, and hence exert a strong influence on ecosystem productivity. Soil nutrient dynamics of cations also provide an indication of the dominant soil forming processes at work in a particular system. To gain insight into the generation and distribution of the soil cation pool in the Fraser Experimental Forest (FEF), we combined geochemical mass balance techniques and isotopic analyses of soil geochemical data to pedons across eight soil catenas in complex mountain terrain typical of the central Rocky Mountains. We found that mass gains in the FEF soils are primarily attributable to pedogenic additions of Ca to the soil mantle via atmospheric dust, and specifically that soil catenas on the summit landscapes were most enriched in Ca. Our data also show that atmospheric deposition contributions (calculated using Sr isotope ratios) to soils is as high as 82% ( $\pm 3\%$  SD), and that this isotopic signature in A-horizons and subsurface soil horizons diverges along a soil *catena*, due to both vertical and lateral hydrologic redistribution processes. Our results suggest that long term soil development and associated chemical signatures at the FEF are principally driven by the coupling of landscape scale cation supply processes, snow distribution, and snowmelt dynamics. Soil development models describing pedogenesis across catenas in montane ecosystems must pay special attention to atmospheric inputs and their redistribution. Any changes to these dynamics will affect productivity and soil/water chemistry in such ecosystems as investigated here.

## 1. Introduction

Two major sources of base cations exist in terrestrial ecosystems—cations derived from parent materials (usually bedrock) and cations added via both wet and dry atmospheric deposition (i.e. dust). In terrestrial ecosystems, bedrock weathering is an important process whereby essential plant nutrients like  $\text{Ca}^{2+}$  become biogeochemically available (Johnson et al., 1968; Walker and Syers, 1976). However, local weathering inputs alone may be inadequate to maintain soil fertility without the addition of exogenous cations (Capo and Chadwick, 1999; Zaccherio and Finzi, 2007). Indeed, long term additions of atmospherically-derived dust provide a key geochemical input for various terrestrial ecosystems (Stoorvogel et al., 1997; Capo and Chadwick,

1999; Okin et al., 2004). While an important factor in pedogenesis (Simonson, 1995; Porder et al., 2007; Lawrence et al., 2013), the incorporation of dust into soil systems is dynamic and poorly quantified. Herein, we examine the important contribution that dust inputs have on soils across the mountainous catenary sequences and disparate geomorphic surfaces of Fraser Experimental Forest (FEF).

Atmospheric dust has been found to contribute to soil nutrient pools in mountain ecosystems of the West and specifically, Colorado (Clow et al., 1997; Mladenov et al., 2012; Lawrence et al., 2013; Brahney et al., 2014). Dust may become trapped under conditions promoting surface roughness like in vegetated areas and in soil crusts which, exert a pronounced influence on the concentrations of Ca, Na, K, and N at and near the soil surface (Reynolds et al., 2001; Blank et al., 1999). Dust

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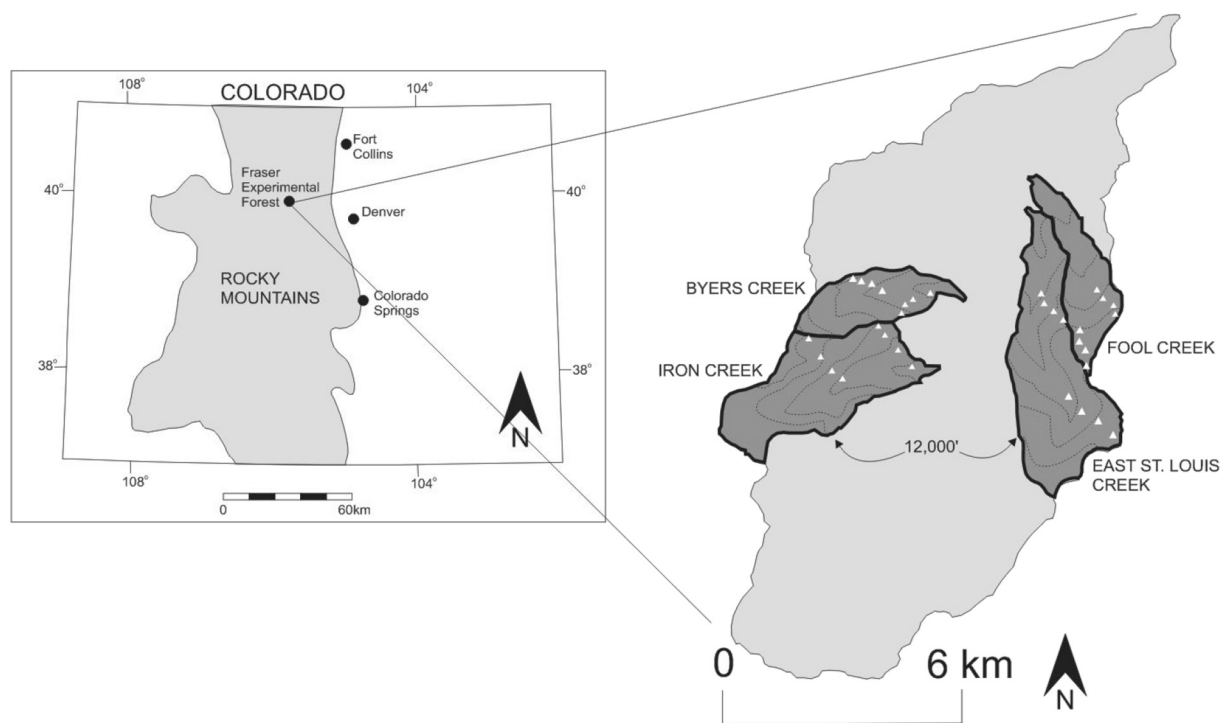


Fig. 1. The location of the Fraser Experimental Forest, Colorado. Soils were sampled along eight catenas in four catchments (dark gray) within the larger St. Louis Creek catchment (light gray). Sample points along the catenas are indicated by white triangles. Contour lines for elevation (countour interval = 500') are represented by hachured lines; the highest contour line shown (12,000') is labeled, for reference.

derived Ca, in particular, may regulate ecosystem function as soil Ca acts as a buffer to acid precipitation and surface waters, plays a main role in the base saturation of soils, and is an essential plant nutrient, exerting an important influence on the health of forest ecosystems (Richter et al., 1994; Schmitt and Stille, 2005; Groffman and Fisk, 2011). Human activities directly and indirectly impact dust production and, hence its potential influence in soil chemistry. It has been recognized for decades that drought conditions, in combination with agriculture and other land uses, markedly increase soil erosion and substantially contribute to airborne dust production (Middleton, 1985; Tegen et al., 1996). For instance, overgrazing by livestock has been shown to be a significant contributor to soil erosion and dust production (Niu et al., 2011; Su et al., 2005). Soil loss and production of airborne dust is also exacerbated by off-highway vehicles traveling on unpaved roads and trails (Padgett et al., 2008). It has also been suggested that wildfire may contribute to airborne dust production through the increase of wind erosion (Balfour et al., 2014; Santin et al., 2015).

Constituent mass balance techniques have been used to quantify soil weathering in a variety of ecosystems (Bern et al., 2011; Porder et al., 2007; Anderson et al., 2002) and the application of the constituent mass balance model allows for the identification of pedogenic gains that may indicate dust inputs (Chadwick et al., 1990; Egli et al., 2000). This approach has been used traditionally to quantify soil development by estimating soil strain, volumetric gains or losses within a pedon (Brimhall et al., 1992). Recently, supplemental analytical techniques, such as the utilization of XRD and application of Sr isotopes have been combined with the mass balance approach to quantify soil weathering (Bern et al., 2011; Porder et al., 2007; Anderson et al., 2002).

Strontium (Sr) isotope ratios are regularly employed to determine the relative contribution of soil nutrients from differing weathering pools in ecosystems, including the importance of atmospheric processes (dust) in soil across a variety of ecosystems (Capo and Chadwick, 1999; Blum et al., 2002; Drouet et al., 2005; Chadwick et al., 2009; Reynolds et al., 2012). In studies based in arid climatic zones, where rates of soil development are strongly controlled by eolian dust (Gile et al., 1966;

Gile, 1979; Chadwick and Davis, 1990; McFadden et al., 1991), Sr isotopes were used to determine the provenance of Sr, and therefore Ca, available in the soil environment (Graustein and Armstrong, 1983; Capo et al., 1995). Strontium is a powerful isotopic tracer in terrestrial ecosystems and is frequently used as a proxy for Ca, due to their chemical similarity (Dasch, 1969; Brass, 1975). Numerous studies have demonstrated the potential of using stable Sr isotopes in quantifying atmospheric deposition in ecosystems (Graustein and Armstrong, 1983; Aberg et al., 1989; Gosz and Moore, 1989; Graustein, 1989), weathering and chemical processes in soil environments (Miller et al., 1993), and paleoenvironmental applications (Quade et al., 1995; Capo et al., 1995). Use of isotopic tracers has shown that some terrestrial ecosystems rely more on soil cations received from atmospheric deposition than from bedrock weathering (Drouet et al., 2005; Vitousek et al., 1999; Lawrence et al., 2013).

To date, dust studies have been confined to alpine ecosystems, or to multiple, unconnected sites within various landscapes (Capo and Chadwick, 1999; Reynolds et al., 2006; Lawrence et al., 2010, 2013). The work presented here builds on these previous alpine studies and follows prior investigations conducted at the FEF that document the occurrence of dust deposition events (Retzer, 1962; Rhoades et al., 2010); importantly, dust's overall impact on soil functions at the FEF remains undetermined. As in many other studies, Sr isotopic techniques will be employed, though our innovation is using Sr isotopes to characterize the contributions of dust to soil chemistry along soil *catenas*.

The goals of this research were to 1) determine whether landscape position along catenas imparts a control on the distribution and sourcing of soil cations in the FEF and 2) evaluate the contribution of atmospherically-derived Ca to the soil cation pool in FEF soils. We focus on soil Ca here because of its chief role in ecosystem function. There is also recent interest in the effects of changing forest harvest practices on soil Ca stocks (Brandtbert and Olsson, 2012; Zetterberg et al., 2016). To our knowledge, this is the first study to offer a comparison between dust and weathering additions along a topographic continuum, and to describe how they contribute to pedogenesis along a soil catena. Mass

balance calculations are presented for soils along catenas in the FEF, and Sr isotopes were analyzed to determine estimates of dust accumulation and contributions to the soil chemistry at the FEF.

## 2. Methods

### 2.1. Study site description

The FEF, Grand County, Colorado, USA, is located in the central Rocky Mountains (Fig. 1). Research in the fields of hydrology and forest dynamics has taken place in the FEF since 1937. Daily minimum and maximum temperatures at the FEF range from  $-40\text{ }^{\circ}\text{C}$  and  $32\text{ }^{\circ}\text{C}$  annually; the mean annual temperature is  $1\text{ }^{\circ}\text{C}$ . Mean annual precipitation is 71–76 cm, two thirds of which is snowfall (Alexander and Watkins, 1977). Metamorphic rocks dominate the FEF landscape, with sedimentary rocks a minor component. FEF geology consists mostly of felsic to intermediate composition gneiss and schist, with small amounts of granitic and intermediate composition igneous rocks (Theobald, 1965). Alpine landscapes are dominated by grasses and shrubs. The forested landscapes consist of a mixture of lodgepole pine, Engelmann spruce, subalpine fir, and quaking aspen. The soils of the FEF are commonly young and poorly developed. Inceptisols and Entisols dominate the upper landscape positions; Alfisols with weakly developed illuvial horizons occur on the sideslope positions; and Histosols can be found along riparian corridors within slope and depression wetlands.

The topography of the FEF is dominated by steep, high mountain slopes. The only portion of the landscape that approaches a zero slope is either high atop alpine grasslands or small areas adjacent to surface water corridors. Summit landscapes are generally narrow and are somewhat convex, rather than having abrupt, sharp peaks. These landscapes are also relatively stable and gently sloping, while sideslope landscapes along *catenas* are relatively unstable and steep. Some of these areas are quite hummocky, most likely due to relic glacial features, while others have relatively smooth slopes. The slopes along the catena shoulder, backslope, and footslope landscapes sampled in this study approached  $20^{\circ}$ . Elevation of the sites within this study ranged from approximately 2900 m (9,500 ft) to 3550 m (11,600 ft) (Fig. 1).

### 2.2. Catena selection

The catenas selected for sampling were located in the FEF within four catchments: Byers Creek, East St. Louis Creek, Fool Creek, and Iron Creek (Fig. 1). These catchments were chosen primarily based on accessibility and parent material geology. Two catenas were sampled in each catchment, for a total of 8 catenas. Sites consisted of the summit, shoulder, backslope, and footslope landscape positions from each catena, for a total of 32 sites. The shoulder, backslope, and footslope, collectively, are often referred to as the sideslope. The summit positions do not receive upslope inputs hydrologically; they are the highest elevation sites along the catenas. All sites along the sideslopes are hydrologically connected to adjacent landscape positions.

Of the 40 sites, 35 were located under forest canopy. Five sites were located in alpine vegetation. The three highest-elevation catenas began in alpine vegetation in the summit landscape; two of these catenas contained one adjacent, lower elevation site in alpine vegetation.

A single type of parent material was isolated across all of the catenas to hold that influence constant in the study. Catenas and their individual sites were located on areas of mineralogically-similar granodiorite, biotite gneiss, and biotite schist. Relatedly, no lithologic discontinuities along the catenas were identified. The parent materials identified in this study are consistent with previous geologic mapping conducted at the FEF (Theobald, 1965; Eppinger et al., 1984; Shroba et al., 2010).

### 2.3. Soil sampling and analysis

At each of the 32 sites, a soil pit was excavated to the maximum depth allowable by hand, and that was “that portion of a C or R horizon which is easily obtainable but reasonably distant” below the solum (Buol et al., 1997). As such, parent material properties were obtained from the analysis of the deepest portion of the C horizons accessible in the soil pits or soil cores. Pedons were described and sampled by genetic horizon (Schoeneberger et al., 1998) and approximately 1–2 kg of soil was taken from each genetic horizon for laboratory and mineralogical characterization and analysis. All soil samples were bagged and sealed in the field, and transported to the Colorado State University (CSU) laboratory immediately.

Laboratory analyses included the determination of soil texture, total carbon, geochemistry, and bulk mineralogy. All laboratory work, except for X-ray diffraction (XRD), was performed at Colorado State University. Soil texture was determined using the hydrometer method (Gavlak et al., 2003). Total carbon was determined on a LECO Tru-Spec CN analyzer (Leco Corp., St. Joseph, MI, USA) by the method described by Kowalenko (2001). Soil bulk density was determined empirically by the method outlined by Rawls (1983). Major elements were measured on a Perkin Elmer Optima 7300 CV inductively coupled plasma-optical emission spectrometer (ICP) (Perkin Elmer, Waltham, MA, USA) following total digestion of samples in HCl, HNO<sub>3</sub>, and HF (Page et al., 1982). Strontium isotopes were measured on a Perkin Elmer Sciex Elan CRC II ICP-mass spectrometer. Bulk mineralogical analyses were performed on samples of unweathered parent material (C horizon rock). XRD spectra were obtained for randomly oriented aggregate mounts between  $2$  and  $65^{\circ} 2\theta$  on a Scintag GBC MMA Diffractometer (University of Northern Colorado, Department of Earth Sciences) configured at: 35 kV, 28.5 mA, a step size of 0.02 at  $2^{\circ}/\text{min}$ . Analysis of the obtained powder XRD patterns was performed using the RockJock program (Eberl, 2003) against the reference mineral, zincite. Mineralogy was determined by the RockJock mineral library.

### 2.4. Geochemical mass balance

Here, we employ the geochemical (constituent) mass balance approach to estimate weathering by calculating volume changes through a soil profile and parent material composition. Strain ( $\epsilon$ ), is a measure of soil volume change incurred during pedogenesis, and was calculated as follows: (Brimhall and Dietrich, 1987; Chadwick et al., 1990; Brimhall et al., 1992)

$$\epsilon_{i,w} = \frac{\rho_p C_{i,p}}{\rho_w C_{i,w}} - 1 \quad (1)$$

where  $\rho$  is soil bulk density and  $C_i$  is the concentration of an immobile reference element in w, the weathered soil horizon or p, the soil parent material. Titanium (Ti) and Zirconium (Zr) are typically used as immobile reference elements due to their stability within their host mineral phases (Milnes and Fitzpatrick, 1989) and resistance against dissolution (Brookins, 1988; White, 1995). Studies have found that, during the weathering process, Ti remains enriched in the fine fraction and Zr enriched in the coarse silt fraction of the particle size distribution (Stiles et al., 2003; Taboada et al., 2006). Herein, the mobility of Ti and Zr were evaluated by first regressing the mean strain for Ti against Zr to determine their relative pedogenic behavior, and secondly, regressing the relationship between their mass transfer function values against percent clay and sand, respectively. Regression analysis showed that the Ti mass transfer function value was not dependent upon percent clay, as the  $R^2$  of the fitted regression line between the two variables was 0.07%. Its concentration was more uniform with depth than that of Zr. Titanium was selected as the immobile element for this study as its influence over the volumetric changes is negligible relative to Zr and due to its conservative nature within the clay size fraction. These

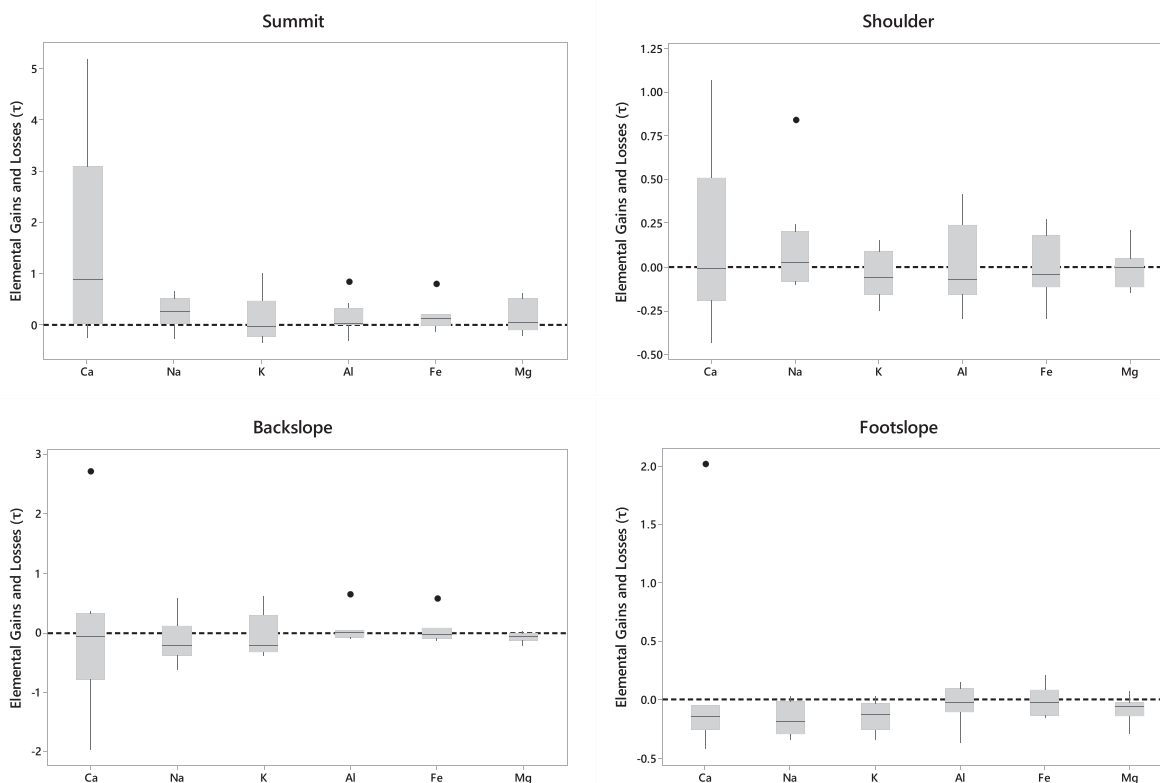


Fig. 2. Elemental gains and losses for whole soil profiles by landscape position the Fraser Experimental Forest, Colorado, by landscape position ( $n = 8$ ). Box plots show depth-weighted median mass transfer function values (horizontal line), middle 50% (box), upper and lower 25% (bars), and outliers (filled circles). Note the y-axis scale changes between panels. Values which fall on the dashed horizontal line displayed no change in median  $\tau$ .

results suggest a low degree of mobility in FEF soils.

Strain is a unitless index; positive and negative values indicate increasing or decreasing soil volume, respectively. Strain values calculated in this study are the sum of the depth-weighted contributions from each weathered soil horizon in its respective pedon.

The mass transfer coefficient,  $\tau_{j,w}$ , is used to evaluate element mobility within the soil (Brimhall and Dietrich, 1987; Chadwick et al., 1990; Brimhall et al., 1992) as such:

$$\tau_{j,w} = \frac{\rho_w C_{j,w}}{\rho_p C_{j,p}} (\epsilon_{i,w} + 1) - 1 \quad (2)$$

where  $C_j$  is the concentration of a chemical species and  $\epsilon_{i,w}$  is volumetric strain. The mass transfer coefficient is used to compute elemental flux for a given soil horizon, in relation to the element's mass in the parent material. The deepest horizon from each respective soil pit was used as the parent material for the purpose of these calculations.

The weathering mass flux from a soil profile was calculated using the following equation (Egli et al., 2000):

$$\text{mass}_{j,\text{flux}} = \rho_p \Delta z_w \frac{1}{\epsilon_{i,w} + 1} C_{j,p} \tau_{j,w} \quad (3)$$

where  $\rho$  is bulk density,  $w$  is the weathered soil horizon,  $p$  is parent material, and  $C_{j,p}$  is the concentration of element  $j$ , and  $z$  is the thickness of the soil horizons. The mass fluxes from horizons contributing to a soil profile were summed to obtain a weathering mass flux for the entire soil profile. Mass flux values estimate elemental gain or loss of a mobile element from the soil profile.

## 2.5. Strontium isotopes

Here, we express the isotopic composition of strontium as  $\delta^{87}\text{Sr}$ , which is calculated by:

$$\delta^{87}\text{Sr} = \left( \frac{87\text{Sr}/86\text{Sr}_{\text{SAMPLE}}}{87\text{Sr}/86\text{Sr}_{\text{SEAWATER}}} \right) \quad (4)$$

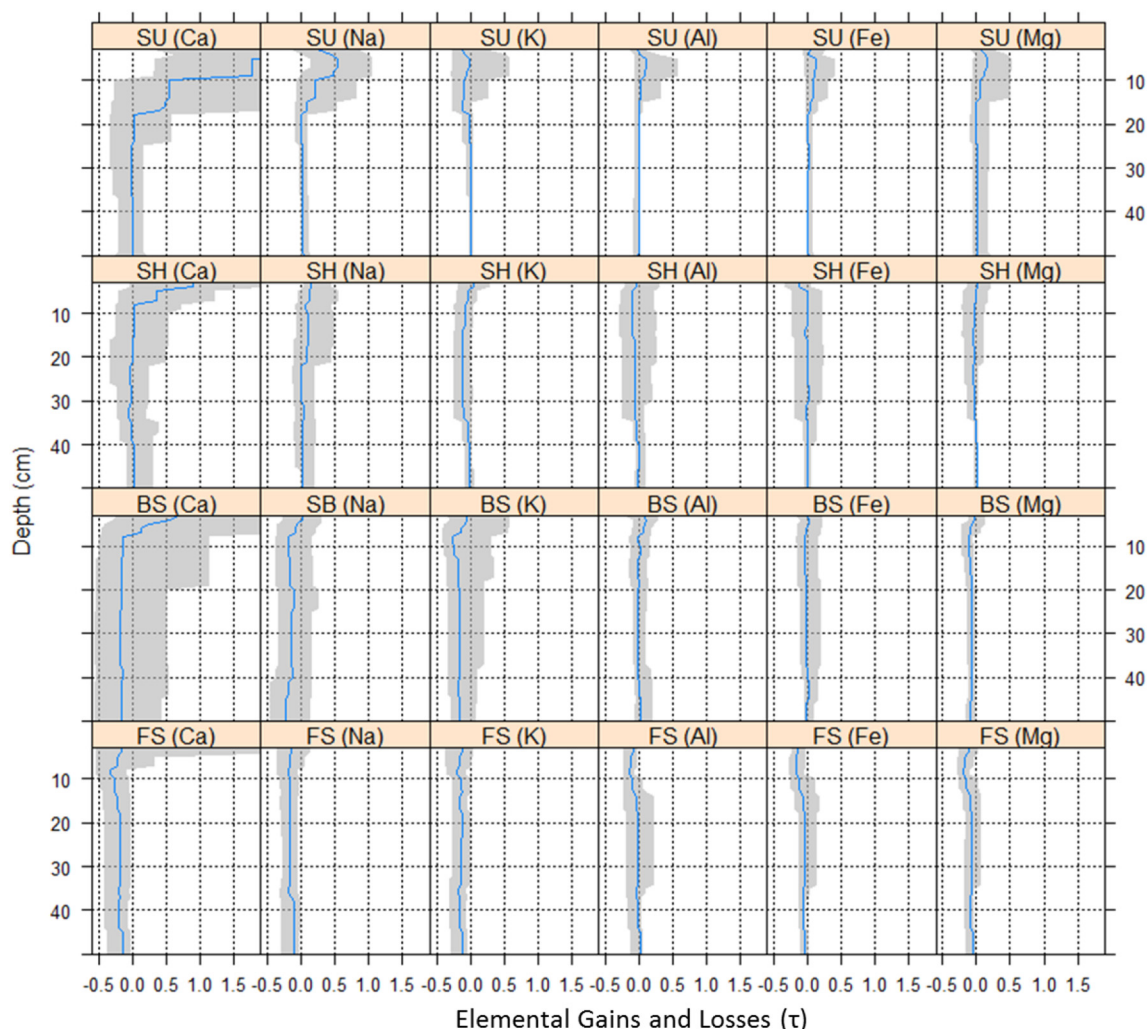
Likewise, a two end member mixing model can be used to determine the relative contributions of two sources (Capo et al., 1998). We use such a model to calculate the relative contributions of atmospheric dust and in situ weathered mineral rock to summit soils as follows:

$$X(\text{Sr})_1 = \frac{\left( \frac{87\text{Sr}}{86\text{Sr}} \right)_{\text{mix}} - \left( \frac{87\text{Sr}}{86\text{Sr}} \right)_2}{\left( \frac{87\text{Sr}}{86\text{Sr}} \right)_1 - \left( \frac{87\text{Sr}}{86\text{Sr}} \right)_2} \quad (5)$$

where  $X(\text{Sr})_1$  is the mass fraction of Sr derived from source 1. Subscripts 1 and 2 refer to the two sources, dust and rock. The mix subscript indicates the mixture (soil) component. The mean  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio from three rock (biotite gneiss) samples ( $\delta^{87}\text{Sr} = 41.66$ ) from soil pits along the Byers Creek lower catena was used for the rock source isotopic signature.  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios of dust ( $\delta^{87}\text{Sr} = 1.23$ ) sampled from the Central Rocky Mountains (Neff, personal communication, October 17, 2013; Clow et al., 1997) were used for the dust source isotopic signature, as it has been suggested that the background dust signature across the Rocky Mountains is largely homogeneous (Munroe, 2014).

## 2.6. Statistical calculations

The data were grouped by landscape position so that each catena was considered a replicate. Statistical tests were used to determine the effect of landscape position on weathering flux; results were calculated using Minitab 17 software (Minitab, Inc., State College, PA). We used Welch's ANOVA test ( $\alpha = 0.05$ ) to examine average values for mass transport coefficients and weathering mass flux along the catenas. When appropriate, average values are reported in the text as the data mean  $\pm$  standard deviation.



**Fig. 3.** Elemental gains and losses for soil profiles by landscape position the Fraser Experimental Forest, Colorado, by landscape position ( $n = 8$ ). Plots show the mean  $\tau$  for each respective cation (blue line) bounded by the 25th and 75th percentiles of each value at depth. SU = summit; SH = shoulder; BS = backslope; FS = footslope. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

### 3. Results

#### 3.1. Pedogenic gains and losses along catenas

Compared to their parent material, summit landscape soils were enriched in Ca, Na, K, Al, Fe, and Mg (Fig. 2), with the most substantial gains in Ca and Na (Fig. 2). Although the mean values for  $\tau$  for K, Al, Fe, and Mg were slightly positive, it could be argued that as much of the data indicated losses as gains for these four elements. Likewise, the average  $\tau$  for Ca in the summit landscape position was the highest gain among the landscapes, we found no statistical differences in  $\tau$ Ca along the catenas due to the high variability in the data (Fig. 2). Being the highest  $\tau$  value among elements analyzed for along the catenas, Ca enrichment dominates the calculated elemental enrichment values in the summit landscape and appears to exhibit the greatest mobility throughout the top 50 cm of profiles (Fig. 3).

Soils along the catena shoulders were pedogenically enriched (experienced gains) with respect to two-thirds of the elements. Average  $\tau$ Na and  $\tau$ Mg were positive; the median  $\tau$  value for the remaining four elements was negative (Fig. 2). The greatest elemental gains in the shoulder landscape were attributed to Ca; the  $\tau$  value of this element was the most variable, as well. Elemental gains with respect to Ca were highest towards the surface and showed great mobility moving down-profile (Fig. 3). The lowest degree of elemental enrichment were

observed with respect to Mg.

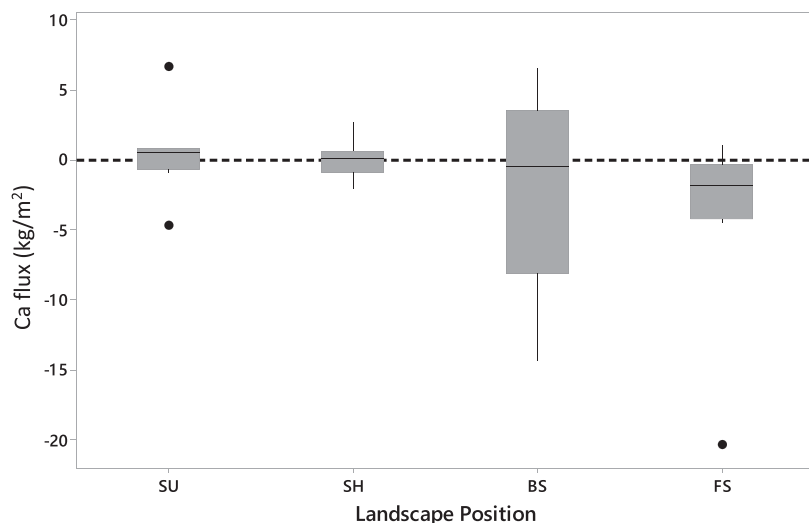
Backslope soils were pedogenically enriched with respect to only one element, Al. The data indicate that soils in this landscape position are depleted in Ca, Na, K, Fe, and Mg. The variability of the  $\tau$  data is lowest for Al, Fe, and Mg in this landscape.

Footslope soils were pedogenically depleted (experienced losses) with respect to all elements; the median  $\tau$  value for all elements is negative. Considering all the elements analyzed, elemental enrichment values in this landscape were less variable than in any other landscape position and the variability in  $\tau$  values decreased with decreasing elevations.

Generally, summit landscapes experienced the highest degree of elemental enrichment; landscapes along catena sideslopes experienced losses or minimal gains of all elements except for Ca and Na (shoulder; Fig. 2) during pedogenesis. Considering median  $\tau$  values along the sideslope, the data suggest that soils are losing all major elements during pedogenesis, with the greatest losses having occurred in the lowest elevation of these sites (Fig. 2).

#### 3.2. Weathering mass flux of ca along catenas

During preliminary data analysis it became clear that Ca enrichment may hold the most pedogenic importance to the FEF soils, among the elements analyzed. Subsequently, mass flux values were integrated over



the entire weathered profile for each study site; the average masses of Ca gained or lost from soils during pedogenesis are displayed in Fig. 4. Regardless of landscape position, individual Ca mass fluxes ranged from  $-20.4 \text{ kg m}^{-2}$  to  $6.6 \text{ kg m}^{-2}$ . No statistical differences are reported with regard to average Ca flux along catenas, though the median Ca flux values are more likely to be positive in the upper landscapes, and negative in the lowest landscapes (Fig. 4). Similarly, the average rate of Ca mass gain or loss during the period of soil formation was calculated using a maximum residence time of the soil. The period of pedogenesis assumed is conservatively based on the timing of most recent regional glacial retreat (12,000 years maximum). Considering all *catenas*, the average Ca mass flux for the four landscape positions moving from summit to footslope are: summit =  $0.4 \text{ kg ha}^{-1} \text{ yr}^{-1}$ , shoulder =  $0.1 \text{ kg ha}^{-1} \text{ yr}^{-1}$ , backslope =  $-1.6 \text{ kg ha}^{-1} \text{ yr}^{-1}$ , and footslope =  $-3.3 \text{ kg ha}^{-1} \text{ yr}^{-1}$ , respectively.

### 3.3. Strontium isotopes along a soil catena

To examine the influence of landscape position on atmospherically-derived soil Ca stocks,  $\delta^{87}\text{Sr}$  values were calculated along one catena with consistent forest species composition to minimize the effect of vegetation. Surface soil  $\delta^{87}\text{Sr}$  values along the soil *catena* ranged between 6.12‰ and 14.02‰ (Fig. 5). The average surface soil  $\delta^{87}\text{Sr}$  value was  $9.23 \pm 3.0\%$ . Moving along the catena, these values remain relatively close to the  $\delta^{87}\text{Sr}$  value of dust 1.23‰ collected at the Loch Vale Main Weather Station, Rocky Mountain National Park, elevation 3050 m (Clow et al., 1997). In subsurface soils  $\delta^{87}\text{Sr}$  values trend towards the  $\delta^{87}\text{Sr}$  of parent material (rock) moving down the catena. The  $\delta^{87}\text{Sr}$  value of subsurface horizons along the catena ranged between 4.42‰ and 27.88‰. The average  $\delta^{87}\text{Sr}$  value in subsurface soil horizons was  $14.73 \pm 9.3\%$ . There was less variation in surface than subsurface  $\delta^{87}\text{Sr}$  values. Selected soil properties of the pedons along this soil catena are presented in Table 1.

### 3.4. Atmospheric contributions to soils

Atmospheric deposition contributions (ADC) to summit landscapes were calculated for both A (surface) horizons and whole soil profiles in order to examine the influence of vegetative cover on atmospherically-derived soil Ca stocks (Fig. 6). The average ADC to soil Ca was higher in forested summit landscapes than in alpine summit landscapes. Mixing model calculations indicate that the ADC to alpine A horizon Ca was  $68 \pm 3\%$ ; the ADC to forest A horizon Ca was  $76 \pm 12\%$ . Whole-

Fig. 4. Average weathering mass flux of Ca across soil landscapes in the Fraser Experimental Forest, Colorado, over the period of pedogenesis (12,000 yrs). Data are the average of Ca mass flux integrated over entire soil profiles along the five landscape position ( $n = 8$ ). Box plots show depth-weighted median mass transfer function values (horizontal line), middle 50% (box), upper and lower 25% (bars), and outliers (filled circles). Values which fall on the dashed horizontal line displayed no change in median flux. SU = summit; SH = shoulder; BS = backslope; FS = footslope.

profile ADC to alpine soil Ca was  $74 \pm 7\%$ ; whole-profile ADC to forest soil Ca was  $82 \pm 3\%$ . Although the average ADC values are higher for forested vs. alpine landscapes, no statistical differences in ADC were found between the averages. Selected soil properties of the summit pedons are presented in Table 1.

## 4. Discussion

Studies have analyzed soil properties along soil *catenas* to gain insight into the dynamics of soil landscapes for over eighty years. Hillslopes were represented as a chain of soil profiles early in the application of the *catena* model by Nye (1954). The *catena* model has been used in studying soil connectivity (Young, 1976), soil topographic relationships (Huggett, 1975), and biogeochemical properties across soil landscapes (Schimel et al., 1985; Litaor, 1992). To this day, the soil *catena* model continues to be used in pedologic-based research to describe the influence of the soil forming factors on the variation in soil properties across landscapes (Evans and Hartemink, 2014; Badia et al., 2015). There is a great amount of evidence that demonstrates the importance of atmospheric deposition as a soil input; however, this is the first study to use the *catena* model in describing relative importance of atmospheric deposition (cation sourcing) in soil formation.

We evaluated the degree of elemental enrichment with respect to six major cations in order to identify “hot spots” of elemental accumulations (or losses) along soil catenas. We discovered gains of all elements in the summit landscapes, of similar magnitude as reported by Lawrence et al. (2013), and progressive elemental losses in each successive lower elevation position along the catenas. Our data suggest that along the catena transects that were sampled, higher landscape positions exhibit pedogenic gains of Ca and lower landscape positions exhibit pedogenic losses of Ca (Fig. 2). These Ca gains substantiate the importance of atmospheric deposition to the supply of base cations at the FEF, as was previously suggested by others as precipitation inputs (Retzer, 1962; Rhoades et al., 2010). Related studies have noted that the Colorado Plateau is a likely source for atmospheric dust in this region (Lawrence et al., 2010.) Indeed, the Ca ion is the most abundant element in the snowpack across Colorado (Stottleyer and Troendle, 1992). It is likely that the pedogenic gains of Na and Mg, especially in summit landscape positions, are largely influenced by atmospheric deposition as well. Our findings demonstrate how atmospheric deposition and elemental transfers have contributed to pedogenesis at the FEF.

The base cation reserve (stocks) contained in soil, including Ca, is

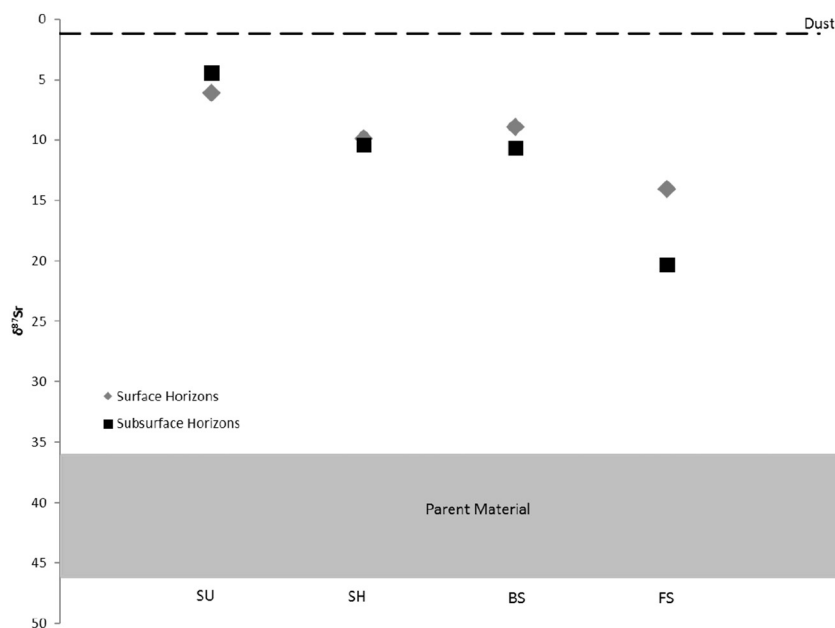


Fig. 5. Plot of  $\delta^{87}\text{Sr}$  for surface soil horizons and subsurface soil horizons along one soil catena from the Fraser Experimental Forest, Colorado. Moving down-catena from the summit to footslope landscape position, surface horizon values remain relatively close to  $\delta^{87}\text{Sr}$  dust values reported in similar ecosystems in Colorado, while subsurface horizon values trend towards the  $\delta^{87}\text{Sr}$  for the range ( $n = 3$ ) of parent material along this catena. SU = summit; SH = shoulder; BS = backslope; FS = footslope.

mostly dependent upon the flux of elements moving in and out the soil and controlled by weathering and element supply rates. The isotopic signature of a soil horizon or profile, specifically its  $\delta^{87}\text{Sr}$  value, is dependent upon the degree to which certain processes contribute to flux. In this paper, the contributions of parent material and atmospheric deposition to the Ca reserve held in the soil along a forested catena are presented. It is clear that the  $\delta^{87}\text{Sr}$  values of surface soil horizons and subsurface soil horizons diverged moving down this catena (Fig. 5), and there appears to be a geochemical divergence, below which the surface (A-horizons) and subsurface soils are most isotopically dissimilar. These data suggest that a chief ecosystem input of Ca at the FEF is being deposited via precipitation and/or dry dust deposition, and that this signature is strong in A horizons regardless of landscape position along catenas. Moving down-catena, this Ca signal becomes somewhat muted in subsurface soil horizons, as  $\delta^{87}\text{Sr}$  values approached values of the geological substrate on which these soils have formed. The wider range of  $\delta^{87}\text{Sr}$  values in subsurface horizons along this catena indicate that atmospherically-derived Ca have been variably incorporated into the soil mantle during pedogenesis through mixing processes such as bioturbation and other disturbances. Mammals, such as pocket gophers, contribute to the mixing of soil in mountain ecosystems (Zaitlin and Hayashi, 2012) and wind disturbances resulting in tip up mounds were observed along the catenas and have been documented in similar ecosystems (Kulakowski and Veblen, 2003).

Our statistical analysis suggests that the degree of mass flux for Ca is not dependent upon the position of soils in the landscape. This finding is not surprising, as the differences in soil temperature and precipitation across these catenas are likely not sufficient to impart differences in the chemical weathering rate of soils. It is interesting, however, that average  $\tau\text{Ca}$  and average Ca flux indicate additions in the summit landscape followed by progressive losses along the catenas. Relatedly, the magnitude of soil Ca mass flux presented here is comparable to Ca flux reported in streams ( $7.9\text{--}59.8 \text{ kg m}^{-2} \text{ yr}^{-1}$ ) by Barnes et al. (2014) and in soil ( $-7.1 \pm 2.1 \text{ kg m}^{-2}$ ) by Lawrence et al. (2013) in similar ecosystems in Colorado. In this study, when evaluating the mass flux (per year) for Ca along catenas, it can be argued that the magnitude of Ca flux is consistent, irrespective of landscape position, although there is a high degree of variability in the data. Nutrient budget modeling efforts may benefit from this finding that the location within or along upland soil landscapes may not affect the degree of Ca weathering—Ca is normally considered a nutrient most at risk of depletion through forest harvest or acid deposition, and thus, one of the base cations that

is often a focus of study in harvest intensity studies. It is likely that any real differences in soil Ca flux would be attributed to biomass uptake or atmospheric deposition.

Our ADC calculations are comparable with similar studies that have addressed soil element origins in regions where dust deposition is prevalent. Grausetin and Armstrong (1983) demonstrated that weathering of parent material contributes less than 20% of Sr to soil of the Sangre de Cristo Mountains in New Mexico; the remainder is supplied by atmospheric sources. More recently, Clow et al. (1997) found that ADC to A and B horizons of soils at Loch Vale, Colorado, ranged between 53% and 68%. Drouet et al. (2005) demonstrated that on average, throughout the profile, two forest soils in Belgium attribute 75%–78% of their soil Ca to atmospheric inputs. Data, based on Sr isotope ratios, indicating high (90%) eolian influence on gneiss-derived soils in New Mexico has been presented by Reynolds et al. (2012). Accounting for dust inputs is important when calculating predictive geochemical models and soil buffering capacities (especially in acid landscapes). The relative importance may be a function of both where soils exist in the landscape and the assumed thickness of the solum.

In addition to shedding light on the proportion of atmospheric contributions to FEF soils, Sr isotope data from summit landscapes demonstrate the preservation of atmospherically-derived Ca into the soil mantle. This variability is especially evident in the A-horizons of forested summit soils, although variability in the summit data is high, regardless of vegetative cover or soil section examined (A horizon vs. whole soil profile). Forest canopies intercept a large portion of snowfall in Colorado Mountains (Schmidt et al., 1998; Montesi et al., 2004; Troendle and King, 1985; Troendle and Reuss, 1997); dust that falls in these environments is also intercepted by the forest canopy. In fact, complementary research at the FEF found a lower dust signature under trees than in adjacent openings, demonstrating dust interception by the canopy (Rhoades et al., 2010). Data from our current study indicate that dust inputs have a greater impact on the geochemistry of forested summit soils as compared to alpine summit soils. Similarly, Clow et al. (1997) found that forest soil exhibited a higher atmospheric-deposition contribution in surface and subsurface mineral horizons as compared to alpine soil.

The distribution of atmospherically derived soil Ca in summit landscapes at the FEF is coupled to the dynamics of snowmelt runoff processes in these mountain ecosystems. The timing of snowmelt is more synchronous and rapid in the alpine ecosystems than in subalpine forests, as incoming solar radiation which drives snowmelt generation

**Table 1**

Selected soil properties for all sites along the Byers Lower Catena and all summit position sites in the study in the Fraser Experimental Forest, Colorado. Site names are coded as follows: Catena Name – Landscape Position – Vegetative Cover. BL = Byers Lower; BU = Byers Upper; EL = East St Louis Lower; EU = East St Louis Upper; FL = Fool Lower; FU = Fool Upper; IL = Iron Lower; IU = Iron Upper. SU = summit; SH = shoulder; BS = backslope; FS = footslope. F = Forest; A = Alpine.

Site	Horizon	Interval	Silt	Clay	Sr87/Sr86	Ca	Silt/clay
		(cm)	(%)	(%)			
BL-SU-F	A	0–5	26	25	0.7135	12,210	1.06
	Bw1	5–18	35	26	0.7157	20,210	1.33
	Bw2	18–58	20	20	0.7123	12,020	0.99
	C	58–85	18	10	0.7142	3180	1.83
BL-SH-F	A	0–10	31	29	0.7162	3748	1.06
	Bw	10–35	25	31	0.7165	3929	0.82
	BC	35–75	14	27	0.7134	4384	0.53
	C	75–100	25	24	0.7121	5196	1.06
BL-BS-F	A	0–10	26	30	0.7155	11,120	0.85
	E	10–20	28	27	0.7167	10,810	1.02
	Bw	20–82	10	24	0.7144	13,080	0.44
	BC	82–122	25	23	0.7150	11,920	1.08
BL-FS-F	A	0–13	34	33	0.7191	4665	1.04
	Bw	13–24	32	34	0.7161	6438	0.93
	C1	24–67	17	29	0.7171	7090	0.58
	C2	67–95	12	23	0.7236	9782	0.54
BU-SU-F	A	0–5	19	30	0.7143	6935	0.63
	Bw1	5–10	22	21	0.7175	878	1.04
	Bw2	10–50	16	16	0.7151	931	1.03
EL-SU-F	A	0–5	21	28	0.7151	4231	0.76
	Bw	5–18	22	20	0.7160	4557	1.09
	C	18–60	13	12	0.7141	894	1.08
EU-SU-A	A	0–10	20	37	0.7176	3455	0.54
	BC	10–25	18	20	0.7139	2050	0.92
	C	25–50	15	27	0.7138	786	0.56
FL-SU-F	A	0–4	27	24	0.7210	5404	1.13
	BC	4–15	38	17	0.7124	20,720	2.24
	C	15–60	26	28	0.7119	25,560	0.92
FU-SU-A	AB	0–10	33	33	0.7206	3871	1.01
	Bw	10–61	10	20	0.7176	1000	0.49
	BC	61–90	17	18	0.7180	751	0.95
	C	90–110	27	15	0.7239	745	1.80
IL-SU-F	A	0–3	41	29	0.7200	4979	1.42
	BC	3–17	36	24	0.7150	4407	1.51
	C	17–90	41	24	0.7141	3754	1.72
IU-SU-A	A	0–8	32	28	0.7189	2871	1.15
	Bw	8–31	23	31	0.7125	2789	0.73
	BC	31–37	16	25	0.7234	3834	0.62
	C1	37–60	22	30	0.7189	5485	0.74
	C2	60–100	11	15	0.7147	5916	0.75

is more uniform in alpine ecosystems. In contrast, the timing of snowmelt can be especially variable in forests of differing densities (Guan et al., 2013; Molotch et al., 2009). Sr isotope data indicate that atmospherically derived dust (and its chemical constituents) that falls in alpine summit landscapes in the FEF is incorporated into the soil and subsequently leached downslope into lower landscapes through a pulse of snowmelt-derived subsurface lateral flow. Baron (1992) describes that early season snowmelt flushes the accumulated by-products of “8 months of soil weathering and decomposition” as soil water infiltrates soil horizons. Contrary to alpine landscapes, snowmelt in the FEF forest landscapes infiltrates the soil at a more variable rate and in a more variable spatial pattern. Ca derived from both in-situ weathering and atmospheric deposition in forest landscapes is not subject to this “cationic flushing”. Our data provide evidence that this cationic flush influences the chemistry of the FEF summit soils. On a similar note, this mechanism is coupled to the occurrence of greater dust (snow) interception in forest canopies that is temporarily stored and washed down later via melt and/or throughfall, leading to a pedologically key chemical signature (higher ADC values) regarding the degree of

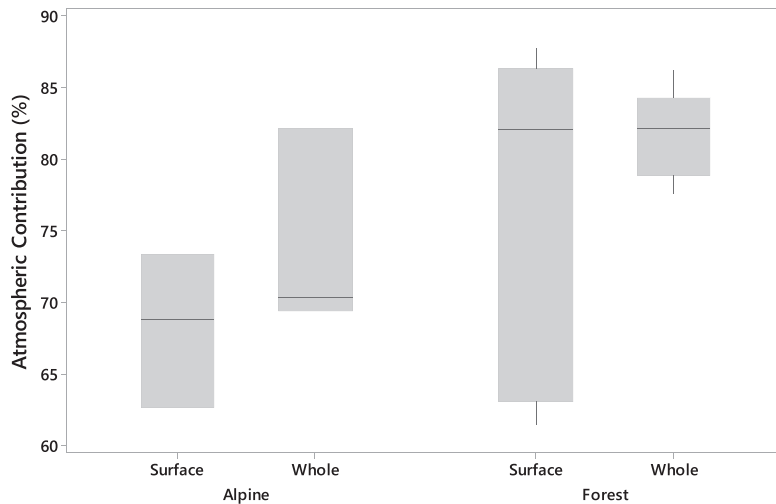
atmospheric dust contributions to these forested landscapes.

It was surprising to find that the contribution of dust to the whole soil profile was as great as or greater than the contribution of dust in the soil surface (A) horizons. Physical soil data revealed high silt size fractions in subsoil horizons, and we interpret this as evidence for the movement of atmospherically derived dust deep into the soil profiles via processes such as bioturbation and frost heaving. Silt is by far the dominant particle size fraction found in Colorado dust (Lawrence et al., 2010), and it follows that soils which are heavily influenced by dust deposition would contain high quantities of silt. Our data revealed a higher percentage of silt in our forested summit soil horizons as opposed to alpine summit soil horizons (Table 1). Similarly, the mean Sr87/Sr86 value for forested summit soil horizons is closer to the isotopic dust signature than their alpine counterparts, indicating that their chemistry is more heavily influenced by dust than by their underlying parent material. This chief input is traditionally understated when considering or defining parent material contributions to soil formation, especially when generalizing the development of soils along *catenas*. Historically, parent materials are highlighted as being sourced from, for example, bedrock, glacial till, alluvium, eolian sand, and loess deposits. Bedrock geology is usually the presumed parent material for a given soil, unless the soil formed on top of sediment. Typical soil development models may overestimate the relative importance of soil elements derived from bedrock geology, and underestimate the importance of elements derived from elsewhere (e.g. atmospheric sources). It has been shown that older landscapes depend almost entirely on exogenous sources for their soil base cation stocks (Kennedy et al., 1998; Chadwick et al., 1999) and it is well known that ecosystems in the Amazon rainforest depend on dust inputs. The base cation stocks of even relatively young soils can be highly dependent upon atmospheric inputs. We have shown how these inputs fit into the *catena* model of soil formation and how atmospheric influence with respect to soil Ca varies by topographic position.

## 5. Conclusions

Over time, the chemical signature of soils is shaped by the relative contributions of weatherable materials from sources such as bedrock and atmospheric dust. Most recent dust-related studies have focused on high-elevation alpine catchments; here, we broadened the focus to include multiple landscapes along soil *catenas*, essentially evaluating weathering along hillslopes. Evaluation of elemental gains and losses by landscape position pointed to noteworthy additions of Ca to summit landscapes, as hillslope processes enable progressive losses of soil Ca along the hillslopes that likely accumulate in wetland soils. Surface soil horizons in summit landscapes are more strongly coupled to the influence of atmospheric deposition than subsurface horizons—and this disparity increases with decreasing elevation. We display geochemical evidence that atmospherically-derived additions contribute to the Ca stocks in FEF soils and that these atmospheric contributions to the soil chemistry at the FEF are pedologically significant. This study also suggests that the chemical signature of soils in snowfall-dominated mountain ecosystems with significant dust inputs may be coupled to snowmelt runoff processes. Increasing rates of dust deposition due to land use change in the region and changing precipitation dynamics due to climate change (i.e. snow vs. rain) will continue to influence soil formation at FEF analogous environments; it remains to be seen how these montane ecosystems are to be affected by these changing dynamics. We conclude that atmospheric deposition plays an important role in soil development at the FEF, and its contributions to soil development at the FEF are entwined with landscape position, vegetation cover, and snowfall dynamics. We have shown how certain soil geochemical properties change along *catenas* in complex mountain *catenas*. Typical soil development models along *catenas* include the underlying parent material contributions but largely exclude this atmospheric input — we should always consider the importance this





**Fig. 6.** Atmospheric deposition contribution to Fraser Experimental Forest summit soils associated with alpine ( $n = 3$ ) and forest vegetation ( $n = 5$ ). Box plots show depth-weighted median mass transfer function values (horizontal line), middle 50% (box), and upper and lower 25% (bars). There were no outliers to report. Surface = surface mineral horizon; Whole = whole soil profiles.

additional input when describing pedogenesis along catenas. We have provided a foundational framework for how these inputs express themselves geochemically along soil catenas. Future work must focus on further unraveling these complex relationships.

#### Competing interests

The authors have no competing interests to declare.

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.17632/yj4jh8sv3y.1>.

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