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## Early Stage Transformation of 2 : 1 Layer Silicates in Pyroclastic Deposits from the 1980 Eruption of Mt. St. Helens

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### *Abstract*

Chemical weathering and pedogenesis are especially rapid in volcanic materials due to their glassy nature, fine particle size, and high porosity and permeability. Early stage mineralogical transformations (0–10 yr) in pyroclastic deposits from the 1980 eruptions of Mt. St. Helens were examined in a cryic–udic climatic regime of western Washington. Chemical weathering in pyroclastic flow deposits near the volcano was strongly affected by acidic precipitation (pH=3.6 to 5.2) originating from sulfuric acid emanating from the vent and by the microtopography that displayed an undulating surface with about 30 cm of relief. Weathering was more intense in depressions because they collected more water than adjacent mounds. Detrital 2 : 1 layer silicate minerals present in the original deposits of the depressions were degraded within five years to poorly crystalline kaolin and noncrystalline hydroxy–Al polymers and aluminosilicates. In the mound landscape position, there was no apparent alteration of the detrital 2 : 1 layer silicates. In forested areas receiving airfall tephra, weathering reactions were driven by carbonic acid originating from CO<sub>2</sub> diffusion from the buried soil. Aluminum released by weathering was preferentially retained as Al–humus complexes and hydroxy–Al interlayers of 2 : 1 layer silicates, which inhibited early formation of allophanic materials. We conclude that 2 : 1 layer silicates in pyroclastic deposits can be rapidly transformed with the resulting weathering products controlled by the dominant proton donor.

### *Introduction*

The 1980 eruptions of Mt. St. Helens in western Washington State, USA provided an excellent opportunity to examine early stage weathering

and mineralogical transformations in pyroclastic materials. Layer silicate minerals of the 2 : 1 type are often found in soils derived from pyroclastic materials. Their occurrence in young volcanic materials is variously ascribed to in situ pedogenic origin, eolian addition, or inheritance from hydrothermally altered materials in the parent material. The authigenic formation of 2 : 1 layer silicates in volcanic soils has long been a topic of debate (Shoji et al., 1993). Authigenic formation of 2 : 1 layer silicates has been identified as the initial crystalline weathering product in seasonally dry environments (Glassmann and Simonson, 1985; Quantin, 1992). Inheritance of 2 : 1 layer silicates has been documented in several pyroclastic materials (Kondo et al, 1979; Pevear et al., 1982; LaManna and Ugolini, 1987; Jongmans et al., 1994). Isotopic evidence has also demonstrated the importance of eolian transport as a major source of 2 : 1 layer silicates in areas such as Japan, Canary Islands, Tanzania and Hawaii (e.g., Dymond et al., 1974, Inoue, 1981; Inoue and Naruse, 1987, Mizota et al., 1988; Mizota and Matsuhisa, 1995). While formation of 2 : 1 layer silicates in volcanic materials in the udic soil moisture regime cannot be ruled out, current evidence suggests that eolian deposition is the major source of 2 : 1 layer silicates, with lesser amounts inherited from the pyroclastic deposits.

The primary objective of this study was to examine early stage (0 to 10 yr) weathering and mineralogical transformations of 2 : 1 layer silicates present in pyroclastic deposits from the 1980 eruptions of Mt. St. Helens. Smectite (saponite) and trioctahedral vermiculite were identified in pyroclastic flows near the volcano and in airfall tephra distributed across a wide area (Pevear et al., 1982; LaManna and Ugolini, 1987). These layer silicate minerals were

formed by hydrothermal alteration of the volcanic cone prior to the eruption and deposited with the pyroclastic materials. Mineralogical transformations are compared in two contrasting environments : (i) a barren pyroclastic flow deposit near the volcano and (ii) at a site that received simulated airfall tephra in a subalpine forest ecosystem.

### ***Study Areas and Methods***

#### ***Mt. St. Helens***

Several pedons were sampled on a pyroclastic flow located on the northeastern flank of Mt. St. Helens (see Nuhn, 1987 for details). The materials were about 5 yr old at the time of sampling. The elevation of sample sites ranged between 1060 and 1080 m. Mean annual air temperature is 5.6° C and annual precipitation averages about 225 cm with only 14 cm falling between June and August (Franklin and Dyreus, 1988). The depth of the pyroclastic deposits ranges from a few meters to greater than 40 m (Rowley et al., 1981). The study sites had no vegetation. Microtopography, consisting of mounds about 30 cm above the depressions and spaced between 2 and 3 m apart, has played an important role in the early stages of weathering.

#### ***Subalpine forest site***

Weathering and mineralogical transformations in airfall tephra were simulated at the Findley Lake Research Area located in the central Cascade Mountains east of Seattle, Washington. The site is located at an elevation of 1150 m. Mean annual air temperature is 5.5° C and annual precipitation averages about 230 cm with only 10% occurring during the summer (N.O.A.A. 1985-1986). Vegetation is dominated by a mature coniferous forest dominated by Pacific silver fir (*Abies amabilis*, Dougl.-Forbes) with mountain hemlock (*Tsuga mertensiana*, Bong.-Carr) as an associated species. The dominant soils at the study site are classified as Andic Cryohumods.

The Findley Lake Research Area did not receive any airfall tephra from the 1980 eruptions of Mt. St. Helens. Previously unleached tephra was collected from airport runways at Yakami, Washington, a distance (130 km) approximately equal to the distance between Mt. St. Helens and the Findley Lake Research Area. Tephra was applied to the soil surface in 2 x 2 m plots at depths of either 5 or 15 cm. Soil solutions were collected continuously for 4

yr to follow solute leaching from the added tephra. After 10 yr, solid-phase samples were collected from the tephra plots and changes in tephra properties were compared to a subsample of the unweathered tephra. Detailed descriptions of the study site, experimental design and methodologies have been previously published (Dahlgren and Ugolini, 1989; Dahlgren et al., 1997, 1999).

#### ***Pyroclastic materials***

The pyroclastic materials at both sites were of dacitic composition and consisted of newly crystallized materials along with lithic components derived from the hydrothermally altered cone (Korosec et al., 1980; Pevear et al., 1982). The magmatic materials were dominated by vesicular glass and plagioclase (andesine) with lower concentrations of hypersthene, hornblende, opaque minerals and biotite (Korosec et al., 1980; Fruchter et al., 1980). The older lithic fraction was dominated by plagioclase, but also contained minor amounts of smectite (saponite), trioctahedral vermiculite, cristobalite, tridymite and quartz (Ames, 1980; Pevear et al., 1982; LaManna and Ugolini, 1987). The pyroclastic materials at both sites had a loamy sand texture with 3 to 6% clay.

#### ***Analytical methods***

The air-dried, less than 2-mm fraction was used for the following analyses. Soil pH was determined in distilled-deionized water (1 : 1 soil : solution) following a 15-min equilibration period. Particle-size analysis was determined by dry sieving and the pipette method using no pretreatments other than dispersion with sodium hexametaphosphate (Gee and Bauder, 1986). Cation exchange capacity and exchangeable cations were determined by 1 M NH<sub>4</sub>Cl at ambient soil pH (Soil Survey Staff, 1984). Phosphate retention was determined using the method of Blakemore et al. (1981). Selective dissolution was performed by the following treatments : (i) sodium pyrophosphate (Al<sub>p</sub>), one 16-hr extraction (McKeague, 1967) and (ii) acid ammonium oxalate (Al<sub>ox</sub>, Fe<sub>ox</sub>, Si<sub>ox</sub>), one 4-hr extraction at pH=3 in the dark (McKeague, 1976). Iron, Al and Si in all extracts were measured by inductively coupled plasma (ICP) spectroscopy.

The clay-size fraction (<2 μm) was isolated by sedimentation following particle-size analysis

and desalted by dialysis against deionized water. Additionally, subsamples from the forested site were pretreated with citrate-dithionite prior to the separation of the clay-size fraction (Holmgren, 1967). X-ray diffraction was performed on oriented samples using standard methods (Whittig and Allardice, 1986).

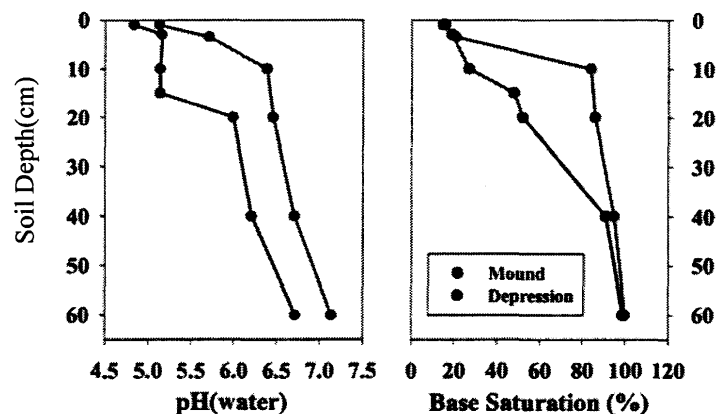
## Results and Discussion

### *Pyroclastic flow deposits near Mt. St. Helens*

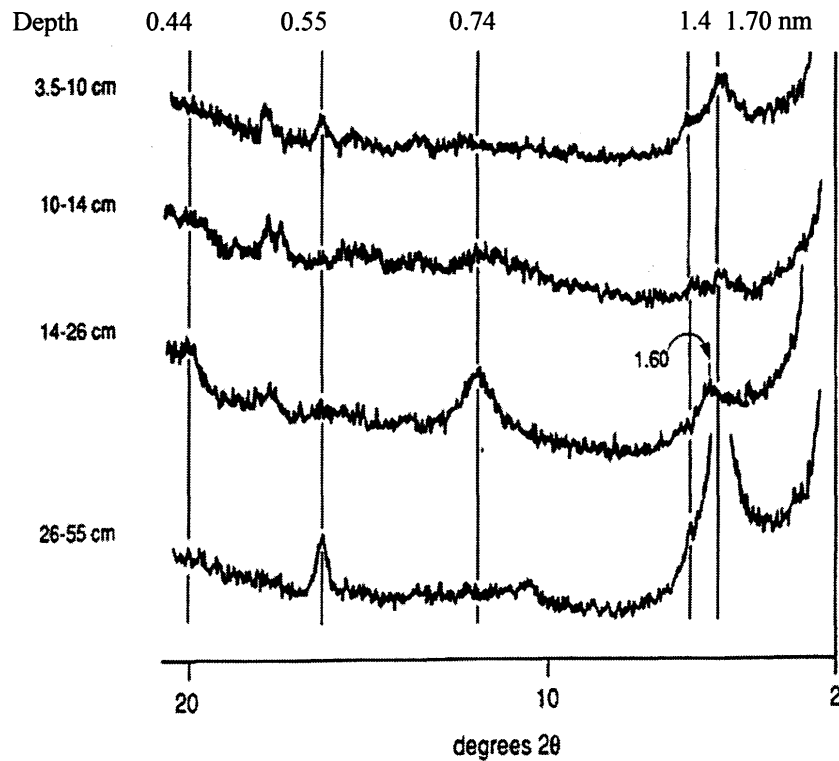
Soil acidification rapidly occurred (<5 yr) in the pyroclastic flow deposits near Mt. St. Helens (Fig. 1). Soils in the mound component of the landscape were acidified in the upper 10 cm with near neutral pH values throughout the lower portion of the profile (Nuhn, 1987). In contrast, soils in the depressions were more strongly acidified with pH values of 5 or less throughout the upper 15 cm. Base saturation showed a similar trend with lower values throughout the upper 40 cm of soils in depressions as compared to the mounds. In the immediate vicinity of Mt. St. Helens, the pH of precipitation ranged from 3.6 to 5.2 in the six years following the 1980 eruption with the acidity originating primarily from sulfuric acid (Nuhn, 1987). Emission of sulfur dioxide (SO<sub>2</sub>) averaged more than 1000 Mg d<sup>-1</sup> following the 1980 eruption (Casadevall et al., 1983) providing a considerable proton load from atmospheric deposition. The acidity originating from the precipitation was largely consumed by weathering reactions in the upper 5 cm of the mound soils (Nuhn, 1987). In contrast, acidification and weathering was more intense in the depressions because they collected more water than adjacent

mounds. Surface runoff from mounds to depressions was promoted by formation of a crust on the barren surfaces. Below the zone of intense soil acidification by strong acid (H<sub>2</sub>SO<sub>4</sub>), carbonic acid became the dominant proton donor (Nuhn, 1987).

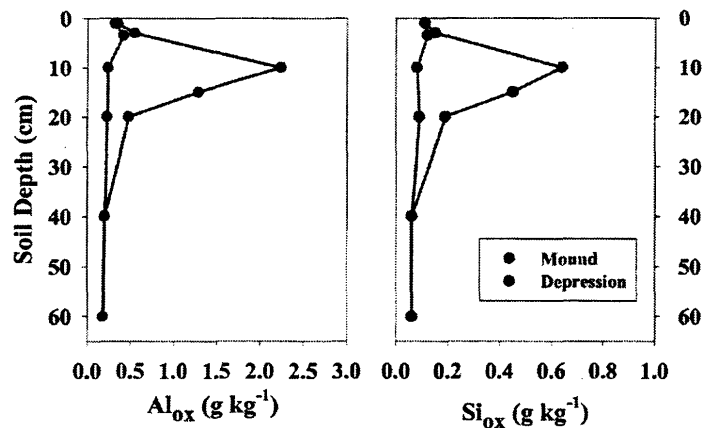
In the immediate surface layer (1–3 cm) of the mounds, there was an apparent depletion of 2 : 1 layer silicates in samples collected in 1983 and thereafter (LaManna and Ugolini, 1987). This layer corresponds with the zone of most intense acidification (pH=5) in the mound soils. Detrital 2 : 1 layer silicate minerals in the upper 26 cm of soils in the depressions were largely degraded within five years to poorly crystalline kaolin and noncrystalline materials (Fig. 2) (Nuhn, 1987; Ugolini et al., 1991). Below the surface layers (upper 10 cm) that have received recent eolian materials with 2 : 1 layer silicates, the soil layers display a weathering progression with depth. The 26–55 cm layer has experienced only weak acidification and the unaltered 2 : 1 layer silicates are the dominant clay mineral. The 14–26 cm layer represents a zone with moderate acidification and displays a large decrease in 2 : 1 layer silicates and the formation of a kaolin mineral (0.74 nm). The kaolin mineral did not expand with formamide treatment indicating that the phase was more similar to kaolinite than halloysite. The 10–14 cm layer represents the zone of most intense acidification and weathering within the profile. In this zone the 2 : 1 layer silicates are largely degraded and only a very weak and broad kaolin (0.74 nm) peak was observed. This suggests that crystalline clay minerals were largely unstable in this zone of intense soil acidification by sulfuric acid.



**Figure 1.** Soil pH(H<sub>2</sub>O) and base saturation of soil profiles from the mound and depression microtopographic positions five years after emplacement of the 1980 Mt. St. Helens pyroclastic flow.



**Figure 2.** X-ray diffractograms (Mg-glycol treatment) for the clay-size fraction from the depression microtopographic position five years after emplacement of the 1980 Mt. St. Helens pyroclastic flow.



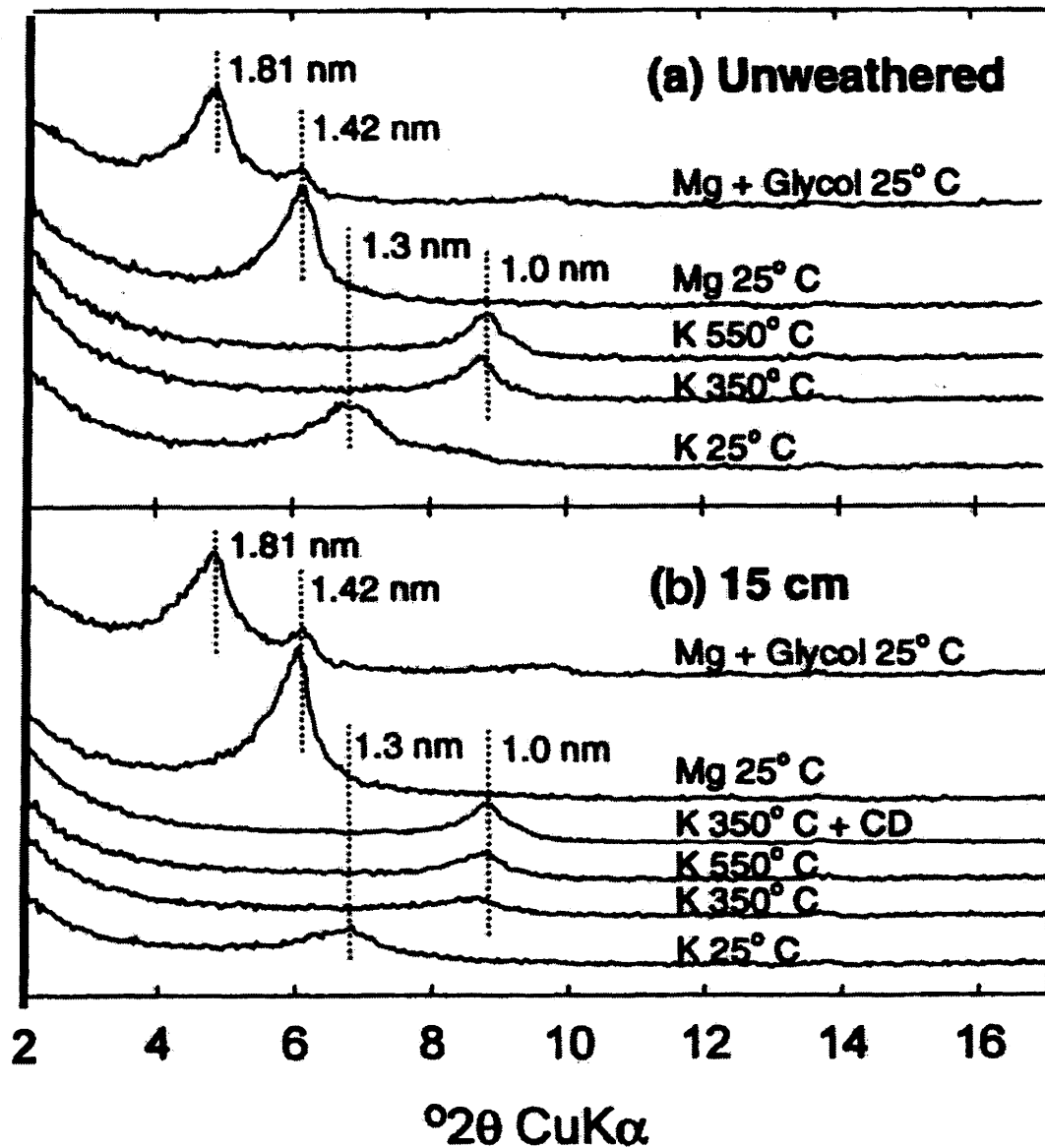
**Figure 3.** Oxalate extractable Al and Si concentrations of soil profiles from the mound and depression microtopographic positions five years after emplacement of the 1980 Mt. St. Helens pyroclastic flow.

Oxalate extractable Al and Si were distinctly elevated in the 10–14 cm layer suggesting that an allophanic material may be forming in the zone of strongest acidification (Fig. 3). The Al : Si atomic ratio of the oxalate extractable fraction ranged from 2.9 to 3.5, which is appreciably greater than maximum values of 2.0 for Al-rich allophane and imogolite. This suggests that in addition to Al extracted from allophanic materials, the oxalate extractable fraction may contain Al extracted from

a noncrystalline hydroxy-Al polymer phase or hydroxy-Al polymers from the interlayer position of 2 : 1 layer silicates (when present).

#### *Airfall tephra simulation in subalpine forest ecosystem*

The initial stages of weathering and mineralogical transformations in airfall tephra were simulated by adding previously unweathered tephra to the soil surface of Spodosols (Dahlgren and Ugolini, 1989).



**Figure 4.** X-ray diffractograms for the clay-size fraction of (a) unweathered and (b) weathered (10 yr) airfall tephra added to the surface of a Spodosol in a subalpine forest ecosystem. Treatment designations refer to temperature pretreatment, K and Mg saturation, glycol solvation, and pretreatment of clays with citrate-dithionite (CD).

**Table 1.** Selected solid-phase characterization for unweathered tephra and tephra weathered for 10 yr in a subalpine forest ecosystem.

	Unweathered tephra	Weathered tephra
pH(H <sub>2</sub> O)	6.3*	5.2*
Pyrophosphate extractable Al (g kg <sup>-1</sup> )	0.1*	0.3*
Oxalate extractable Al (g kg <sup>-1</sup> )	0.2*	0.5*
Oxalate extractable Fe (g kg <sup>-1</sup> )	2.0*	2.8*
Oxalate extractable Si (g kg <sup>-1</sup> )	0.1	0.1
Phosphate retention (%)	1*	7*

\*Means are significantly different at (P<0.05)

The primary proton donor in the tephra layer was carbonic acid with a minor contribution from organic acids originating from leaching of the tree canopy and the incipient litter layer accumulating at the surface of the tephra layer (Dahlgren et al., 1999). Solution leached from the tephra layer indicated incongruent dissolution resulting in formation of a cation-depleted, silica-rich leached layer on the glass and mineral surfaces. Due to the near neutral pH values and low concentrations of complexing organic ligands, Al and Fe were relatively insoluble and accumulated in the tephra layer. Field weathering rates during the first four years ranged between  $10^{-18}$  and  $10^{-17}$  mol  $\text{cm}^{-2}$   $\text{s}^{-1}$  for sodium, calcium and silicon (Dahlgren et al., 1999). While these rates are 1–3 orders of magnitude less than those determined for glass and plagioclase minerals in laboratory dissolution experiments, they are remarkably high given that soil temperatures were near 0° C for nearly six months of the year when a snowpack persists (cryic soil temperature regime).

The majority of the carbonic acid ( $\text{CO}_2$ ) promoting weathering reactions in the tephra layer originates from upward transport of  $\text{CO}_2$  from the organic-rich soil horizons of the buried soil. Elevated concentrations of  $\text{CO}_2$  beneath the tephra layer originate from biological respiration (roots and microbes) and from protonation of  $\text{HCO}_3^-$  leaching from the overlying tephra layer. Cations released by weathering in the tephra layer (pH=6–7) leach downward with bicarbonate to the acidic organic horizons (pH=4) where  $\text{H}_2\text{CO}_3$  reequilibrates with the high  $\text{pCO}_2$ . At pH 4,  $\text{H}_2\text{CO}_3$  decomposes to  $\text{CO}_2$  and  $\text{H}_2\text{O}$  and the gaseous  $\text{CO}_2$  diffuses upwards to take part in another cycle of weathering and transport. This example of weathering in surface deposits of airfall tephra demonstrates a unique weathering pathway in which the buried organic-rich soil pumps protons upward to the tephra layer that acts as an alkaline trap for  $\text{CO}_2$ . Thus, the overall process of tephra weathering appears to be controlled by solute/gas transport of the carbonate system ( $\text{CO}_2$ – $\text{H}_2\text{CO}_3$ – $\text{HCO}_3^-$ ).

Solid-phase samples were collected from the added tephra layer after 10 yr in the weathering environment. By this time, a litter layer (Oi) ranging from 2 to 3 cm had accumulated on the surface of the tephra. Tephra pH decreased from 6.3 to 5.2 after 10 yr (Table 1). There were small increases in  $\text{Al}_p$ ,  $\text{Al}_{\text{ox}}$  and  $\text{Fe}_{\text{ox}}$ . There was no detectable change in  $\text{Si}_{\text{ox}}$

concentrations. Phosphate retention increased from 1 to 7% and was probably related to the increase in active Al and Fe ( $\text{Al}_{\text{ox}}$  +  $\text{Fe}_{\text{ox}}$ ).

There was no apparent degradation of the inherited 2 : 1 layer silicates in the tephra layer following 10 yr of weathering in the subalpine forest ecosystem. The mineralogy was dominated by smectite as indicated by the shift of the 1.4 nm peak to about 1.8 nm following glycol treatment of the Mg-saturated sample (Fig. 4). The small 1.4 nm peak that remains following glycol treatment collapses upon K saturation indicating the presence of vermiculite. The K-saturated clays in the unweathered tephra collapsed to a relatively sharp and symmetrical peak centered around 1.0 nm on heating to 350° C. Further heating to 550° C resulted in no apparent change compared with the 350° C heat treatment. In contrast, clays that experienced 10 yr of weathering collapsed to a diminished peak near 1.0 nm that displays a broad shoulder toward high d-spacings after heating to 350° C. With further heating to 550° C, the peak became sharper and more symmetrical, similar to the clays in the unweathered tephra. Samples pretreated with citrate-dithionite to remove (or partially remove) hydroxy-Al interlayers (Barnhisel and Bertsch, 1989) collapsed to a sharp, symmetrical peak at 1.0 nm upon heating to 350° C, similar to the clays from the unweathered tephra. This response is consistent with the presence of hydroxy-Al polymers in the 2 : 1 layer silicate clays.

Aluminum released by weathering in the tephra layer is preferentially retained as Al-humus complexes ( $\text{Al}_p$ ) and hydroxy-Al interlayers in 2 : 1 layer silicates inhibiting the formation of allophane and imogolite (anti-allophanic effect). The lack of an increase in  $\text{Si}_{\text{ox}}$  concentrations and no detectable allophane or imogolite by transmission electron microscopy suggest that no appreciable formation of allophanic materials has occurred in the first 10 yr of weathering in this subalpine forest ecosystem.

Transformation of the inherited 2 : 1 layer silicates was shown to be a function of the weathering environment. In the pyroclastic flows surrounding Mt. St. Helens, strong acid deposition resulting from  $\text{SO}_2$  emission from the volcanic vent results in rapid soil acidification that destroys 2 : 1 layer silicate minerals. The weathering scenario indicated by mineralogical transformations with depth suggests the following weathering pathway with increasing

acidification and weathering intensity : 2 : 1 layer silicate → kaolin → noncrystalline materials (possibly allophanic materials and hydroxy-Al polymers). In contrast, the carbonic acid weathering regime dominating in the tephra layers applied to Spodosols created a less intensive weathering regime in which the Al released by weathering was incorporated into hydroxy-Al interlayers of 2 : 1 layer silicates and Al-humus complexes. As an organic layer accumulates on the surface of the tephra layer, we expect that the carbonic acid weathering regime will be replaced with an organic acid weathering regime that promotes leaching of Al from the tephra layer.

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