Quaternary alkalinity fluctuations recorded in rock varnish microlaminations on western U.S.A. volcanics

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Abstract

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Rock varnish contains an alternating micron-scale layering of manganese-rich and manganese-poor microchemical laminations. Holocene varnishes have lower Mn:Fe ratios when exposed to alkaline environments and higher Mn:Fe ratios when exposed to less-alkaline environments. This is consistent with abiotic and biotic mechanisms of varnish formation, as alkalinity inhibits the concentration of manganese in both models. Mn:Fe variations with depth in varnishes in the western U.S. are interpreted as fluctuations in alkalinity or local microenvironmental influences. If enough varnishes are sampled from different rocks in a region, local effects can be ruled out. K Ar dating of volcanics in the Coso, Cima, Lunar Crater and Crater Flat volcanic fields in the western Basin and Range provide numerical age control throughout the Quaternary, and allow the number of alkalinity fluctuations to be assessed for given periods of time. Alkalinity fluctuations in varnishes are likely tied to fluctuations in the abundance of alkaline aerosols, which are in turn related to fluctuations in the cover of water over playas and the cover of vegetation over soils. Microchemical laminations in varnish, therefore, have the potential to serve as an indicator of environmental fluctuations for the entire Quaternary.

Introduction

Alternating manganese-rich and manganese-poor microlaminae have been observed in marine (Margolis and Glasby, 1973), lacustrine (Moore, 1981), subsurface (Alhonen et al., 1975) and in "rock varnish" terrestrial (Perry and Adams, 1978; Potter, 1979; Dorn and Oberlander, 1982) ferromanganese deposits. Rock varnish occurs in virtually all terrestrial weathering environments but is noticeable in deserts, hence the common name of "desert varnish" (Dorn and Oberlander, 1982). Because of its biogeochemical stability in most arid and semiarid microenvironments, it has the potential to record long-term paleoenvironmental

fluctuations in regions where such records are often missing.

This paper reports on new studies of these micron-scale microchemical laminations in rock varnish, and indicate that a long-term record of alkalinity fluctuations is recorded. The rock varnishes studied here consist of about 15–20% manganese and iron oxides, with clay minerals being the dominant constituent composing up to 2/3rds of the varnish (Potter and Rossman, 1977, 1979). The remainder of the varnish is composed of over 40 minor and trace elements (Dorn and Oberlander, 1982).

Microchemical laminations in ferromanganese deposits have different rates of accretion,

different origins, and sometimes do not occur. The basic question of genesis in each type of Mn–Fe deposit, however, revolves around what factors control the deposition and erosion of manganese and iron.

In the case of rock varnish, three different mechanisms can theoretically produce alternating Mn-rich and Mn-poor laminations. (1) *Biogeochemical erosion* of varnish could occur by a change to more acidic conditions. The acidity would reduce and mobilize manganese, leaving an iron-clay lag. This is achieved typically on a scale of millimeters to centimeters by acid-producing organisms like lichens, cyanobacteria and microcolonial fungi. The result is usually a reduction of both manganese and iron, but it is possible to mobilize only manganese and not iron. (2) *Abiotic origin and alkalinity fluctuations.* Many investigators (Engel and Sharp, 1958; Hooke et al., 1969; Elvidge, 1979; Elvidge and Iverson, 1983; Whalley, 1983; Smith and Whalley, 1988) have argued that manganese can be concentrated by pH–Eh fluctuations. If there is a lack of periodic fluctuations to more acidic conditions, such as under hyper-alkalinity, manganese enhancement could not occur by pH–Eh fluctuations. Under conditions of greater alkalinity, a layer depleted in manganese would be deposited. (3) *Biotic origin and alkalinity fluctuations.* The other model of manganese enhancement in varnish is concentration by microorganisms. Different investigators have found that a microbial mechanism of manganese enhancement is consistent with field evidence and laboratory studies (Krumbein, 1969; Krumbein and Jens, 1981; Dorn and Oberlander, 1981, 1982; Taylor-George et al., 1983; Palmer et al., 1985). When environmental conditions become too alkaline, the microorganisms stop concentrating manganese (see experiments in Mulder, 1972; Bromfield and David, 1976; Schweisfurth et al., 1980; Dorn and Oberlander, 1981). Proponents of a biological origin reject a purely physical–chemical mechanism for concentrating manganese. Reasons have been presented by Krumbein and

Jens (1981) and Dorn and Oberlander (1982). As Dorn (1988a) has argued, there is no field or laboratory evidence to support an abiotic mechanism at this point. It is only reasonable speculation.

Alkalinity hypothesis

The evidence to be presented in the next section indicates a clear empirical association between Mn-depleted varnishes and arid, alkaline conditions. Although I favor a biological origin for the enrichment of manganese in varnish (Dorn, 1988a), this evidence is also consistent with an abiotic mode of varnish formation by Eh/pH fluctuations. Alkalinity would inhibit the concentration of manganese in both models. If manganese-concentrating microbes are inhibited by alkalinity, low-Mn varnish results. If slight pH–Eh fluctuations do not occur to mobilize Mn under highly alkaline conditions, low-Mn varnish results, according to the abiotic model.

The effect of chemical erosion in producing microchemical laminations is local in nature. Based on thin section analyses, Perry and Adams (1978), Dorn and Oberlander (1982) and Elvidge and Iverson (1983) feel the laminae are primary depositional features. Local pockets of acidity, even in a desert, are often generated by the biological activity of non-varnish forming agents like lichens and microcolonial fungi. These agents cause discontinuities in the laminations by eroding pits into the varnish (Fig.1A and B). However, to envision a major climatic change causing a uniform increase in acidity to chemically erode manganese and produce even microlaminations such as those observed here seems unlikely.

The hypothesis argued for here is that alkalinity controls the concentration of manganese in varnish, and that fluctuations in alkalinity over time have controlled past fluctuations in the concentration of manganese. The concentration of manganese at any given micron on the surface of a varnish

or within the varnish is affected by local influences. However, if enough samples are collected, a regional effect can be discerned, both at present and in the past. I should stress, again, that the control of alkalinity on the abundance of manganese in varnish is argued for theoretically by the abiotic and biotic models. The next section presents data consistent with both mechanisms of manganese enrichment in varnish.

Alkalinity and the surficial chemistry of rock varnish

Study sites and methods

The rate of varnish growth in desert subaerial environments is very slow, taking tens of thousands of years to form a complete coating on a rock surface (Dorn and Oberlander, 1982; Whalley, 1983). This makes the study of contemporary environmental controls on varnish chemistry very difficult to assess. This problem is made worse by anthropomorphic changes in the environment, such as changes in aeolian dust fallout (e.g. Baronne et al., 1981; Cahill and Gill, 1987). The approach used in this study, therefore, is to sample varnishes from deposits dated as Holocene using numerical-dating methods (terminology from Colman et al., 1987), or by examining only the upper <5 µm (checked by scanning electron microscopy) of varnishes that have formed over a longer period. Although climatic-environmental fluctuations have occurred in the Holocene (e.g. Baker, 1983; Davis, 1984), these variations are relatively minor compared to the major environmental change from the latest-Pleistocene to the Holocene (Smith and Street-Perrott, 1983; Spaulding et al., 1983; Wells, 1983). I assume the Holocene varnishes formed under vegetation, soil and climatic conditions roughly similar to those found today. The purpose of assessing the relationship between Holocene varnish and the present-day environment is to obtain a control to interpret changes in the microchemical variations in older varnishes.

Varnishes were sampled for this study mostly from outcrops that are influenced only by airborne fallout. Sites are avoided that are too close to the soil, where water runs over a rock face for a distance, adjacent to lichens or other local influences. Recent research has demonstrated that varnishes on such exposures are composed exclusively of dry fall and wet fall material accreted to the rock (Potter and Rossman, 1977; Perry and Adams, 1978; Allen, 1978; Elvidge, 1979; Dorn and Oberlander, 1982). In all, 177 separate sites and sites along 7 altitude-climatic transects were sampled in western North America for this study. However, not all types of measurements were made at each of these sites.

In addition to examining the varnishes that are exposed to only airborne fallout, a separate set of samples were collected from microenvironments that would be subject to the local effects, such as: subsurface; joints in rocks where dust collects; rock surfces exposed to runoff; proximate and distant from lichens, leaf litter, microcolonial fungi; near sources of alkaline deflation; close to the soil. Surficial soil samples were also collected within a meter of the varnished boulder.

pH determinations were made of the surficial soil, airborne fallout, rainwater and scrapings of Holocene varnish. The method for removing an entire Holocene varnish is described by Dorn (1983) and elaborated in Dorn et al. (1987). For sites not known to be Holocene in age by independent numerical dating methods, the upper <5 µm are removed with a tungsten-carbide needle under 45 × magnification. The depth of scraping is checked by scanning electron microscopy. Multiple measurements, multiple samples from a site, and different types of pH meters were used to insure reproducibility of results. Electrodes were calibrated with standard buffers. Varnish, soil, and collected airborne dust were measured separately in a suspension with deionized water in a 2:1 ratio (e.g. McLean, 1982). While measurements of oxidation–reduction potentials (Eh) were made, and would be beneficial in assessing the chemical and biological en-
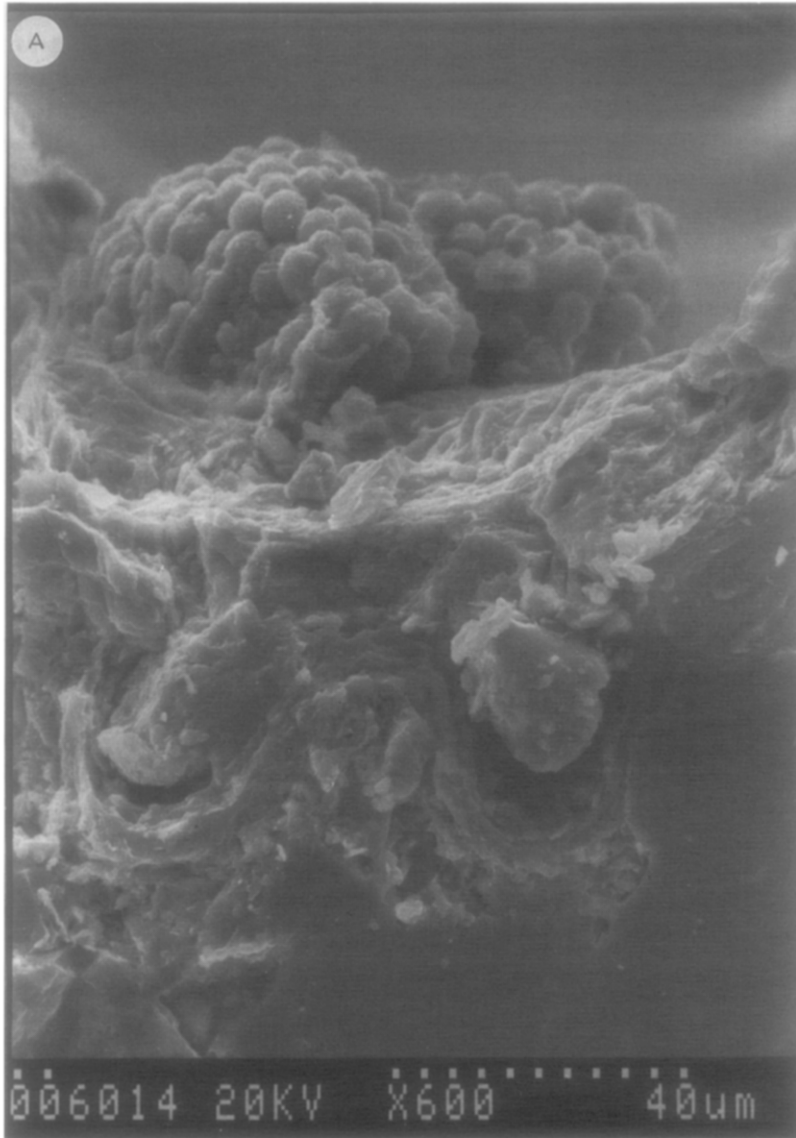
Fig.1. Local erosion of varnish. A. Varnish from Warm Springs Fan, Death Valley, CA, illustrating two paleoerosional pits that have been subsequently filled in with barium-rich detritus. Microcolonial fungi, now seen growing on the surface, are a likely cause of the biogeochemical erosion. Length of scale dots 40 μm.

vironment, recent research has shown that because of internal disequilibrium measured Eh values can be very misleading (Lindberg and Runnells, 1984).

The manganese-to-iron (Mn:Fe) ratios of varnish material, soils, and sampled airborne fallout were determined by proton-induced X-ray emission (PIXE) at U.C. Davis (Cahill, 1980; Cahill et al., 1984), energy-dispersive analysis of X-rays (EDAX) at U.C. Berkeley, U.C. Irvine and Texas Tech University, electron microprobe analyses at California Institute of Technology and U.C. Berkeley, and wet chemical methods were also used, but their results were not felt to be representative. EDAX and microprobe analyses are usually made on the scale of a few μm. PIXE determines Mn:Fe ratios for bulk samples, hence
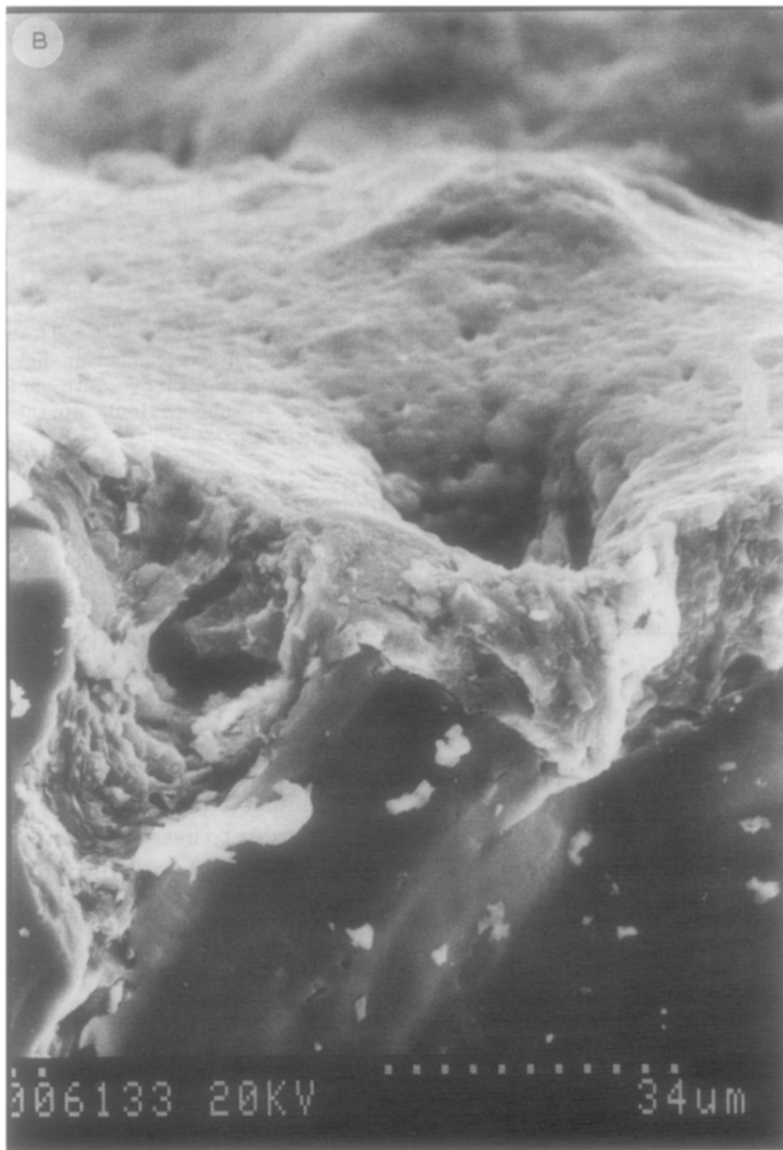
Fig.1.B. Bioerosion pits, in a SEM micrograph, probably made by adventitious microcolonial fungi. The pit on the right is visible by surface observations, but the one on the left would only be noticeable by cross-section analysis. These type of bioerosion pits produce discontinuous microlamination sequences. Varnish collected from Death Valley Canyon fan, CA. length of scale dots 34 μm.

can include truly representative volumes of varnish, several cubic millimeters.

Results

Soil and varnish pH appears to best explain the Mn:Fe ratio of surficial varnish. Figure 2A compares soil pH and varnish Mn:Fe ratios for 67 sites in western North America. Figure 2B is of varnish pH versus the Mn:Fe ratio of varnish. The high correlation of soil pH and varnish pH in Fig.2C does not permit a multiple regression analysis of soil pH and varnish pH with Mn:Fe due to multicollinearity. However, the relationships in Fig.2A and B are significant, to the 0.05 level. Similar results
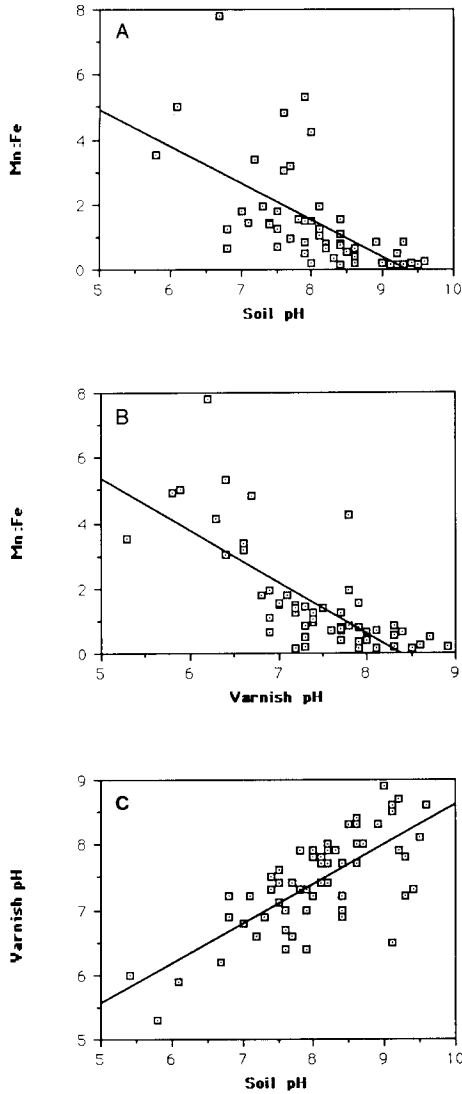
Fig.2. Relationship between varnish pH, soil pH and varnish Mn:Fe at over 50 sites in western North America. A. Mn:Fe = 10.65 − 1.14(soil pH) with $r = -0.61$, significant at 0.05). B. Mn:Fe = 13.33 − 1.59(varnish pH), $r = -0.73$, significant at 0.05. C. Varnish pH = 2.53 + 0.61(soil pH), $r = 0.74$, significant at 0.05.
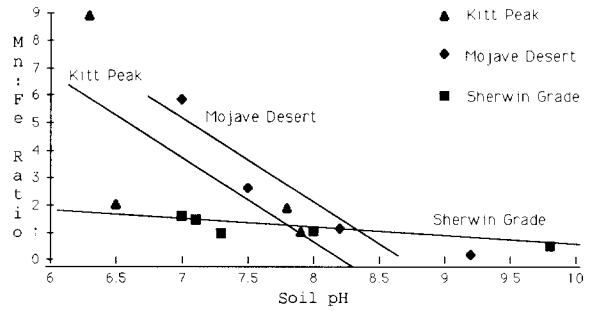

Fig.3. Least-squares regressions and data for 3 altitude-climatic transects comparing how soil pH varies with varnish Mn:Fe ratio over altitude for the central Mojave Desert, CA, Sherwin Grade into the Owens Valley, CA and at Kitt Peak, AZ.

were found for varnishes collected along 3 altitude-climatic transects (Fig.3), but the relationships were not significant at the 0.05 level, probably due to the limited number of sites along the transects. Cahill et al. (1981) found that airborne fallout in the western U.S. is largely composed of soil material. Since the varnishes for Fig.2 were from sites that experi-

ence only airborne fallout, it is reasonable that deflated soil material fell on the varnish. The pH of this material undoubtedly influences the pH of the varnish. In turn, the higher pH values are significantly correlated with Mn:Fe ratios. While correlation does not imply causation, the two mechanisms that could concentrate manganese are inhibited by alkalinity. This evidence is felt to support both the abiotic and biotic models.

In order to assess the influence of climate on Mn:Fe ratios, soil and varnish samples were collected near 18 meteorological stations where Oberlander (1979) had calculated Thornthwaite's index of aridity (Ia) and Oberlander's water balance (Ib). Both are better indicators of moisture effectiveness for vegetation and geomorphology than simple measures such as precipitation. Thornthwaite's (1948) aridity index is a measure of how annual potential evapotranspiration compares with monthly water surpluses and deficits. Higher Ia values indicate greater aridity. Oberlander's (1979) water balance is a measure of soil moisture storage, where higher Ib values indicate more storage. In addition, $\delta^{13}C$ were obtained for varnishes from these sites. $\delta^{13}C$ values in varnish indicate the relative abundance of plants with C_3 vs C_4 and CAM photosynthetic pathways (Dorn and DeNiro, 1985). Less negative $\delta^{13}C$ values are found in arid regions where C_4 and CAM plants are more abundant.
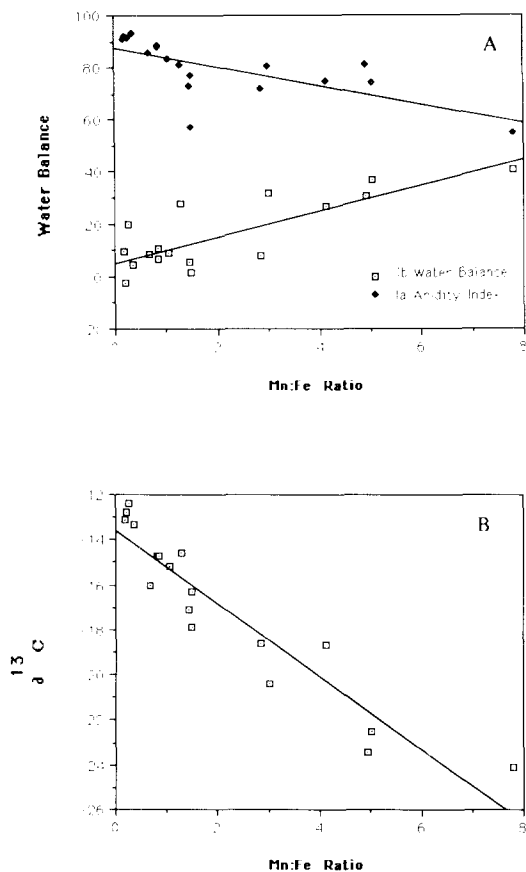
Fig.4. Comparison of Mn:Fe ratios with indicators of climate. A. Thornthwaite's index of aridity (Ia) and Oberlander's water balance (Ib) are correlated with Mn:Fe ratios of varnishes near the 18 meteorological stations where the water balance values were calculated by Oberlander (1979). Ia increases and Ib decreases with greater aridity. Both correlations are statistically significant at 0.05. This illustrates that Mn:Fe ratios are lower in more arid locales. B. $\delta^{13}C$ values of varnish organic matter are highly correlated with Mn:Fe ratios from the same varnishes, at the above 18 meteorological stations. The correlation is statistically significant at the 0.01 level. $\delta^{13}C$ values become more negative as the environment becomes less arid, as elaborated in the text. This illustrates that Mn:Fe ratios are lower where, over the same Holocene period of varnish growth, environmental conditions are more arid.

Figure 4 illustrates how Ia, Ib and $\delta^{13}C$ compare with Mn:Fe ratios. As the Ia and Ib water balances move towards more arid values, Mn:Fe ratios decrease (Fig.4A). Both correlations are significant at 0.05. As C_4 and CAM values move towards more arid values, Mn:Fe

ratios decrease (Fig.4B). The correlation of $\delta^{13}C$ and Mn:Fe is significant at 0.01. Also for these sites, soil pH, varnish pH and Mn:Fe ratios were also significant among each other at 0.05, like in Fig.2. The correlation of Holocene Mn:Fe ratios with Ia and Ib values for meteorological stations should not be indicative of causation. Arid precipitation is notoriously variable over time and space (cf. Bailey, 1979) and varnish sampling sites were sometimes a few kilometers away from the meteorological station. More importantly, a causal mechanism between climate and Mn:Fe ratios does not exist. The correlation of Mn:Fe ratios and $\delta^{13}C$ values does not indicate a causal relationship, but is interpreted here to indicate the same general environmental control on $\delta^{13}C$ values influence Mn:Fe ratios: the general degree of aridity. These data are presented here to illustrate the general association of drier climates and lower manganese concentrations. The cause of variations in Mn:Fe ratios must relate back to either the biotic or abiotic mechanisms of manganese concentration in varnish.

It could be argued that the determination of Mn:Fe ratios by removing only the top layer of varnish is a subjective assessment, and that this removal process includes more than just the surface layer. While the PIXE method analyzes bulk samples and provides more representative data, microprobe and EDAX analyses in cross section only the top layer of the varnish. In order to assess the reliability of the PIXE scraping method for assessing the chemistry of the $< 5\,\mu m$ surface layer of varnish, electron microprobe profiles were completed on varnishes collected along several of the altitude-climatic transects. Figure 5 presents examples of representative electron microprobe profiles of varnish from Kitt Peak, AZ. As the elevation of sites decreases from Fig.5a to 5b to 5c, the Mn:Fe ratio also declines in the surficial layer of the varnish, similar to PIXE analyses from these sites. Of note, this change in Mn:Fe ratios also matches the shift in varnish micromorphology observed from the same sites (Dorn, 1986). The same decrease in
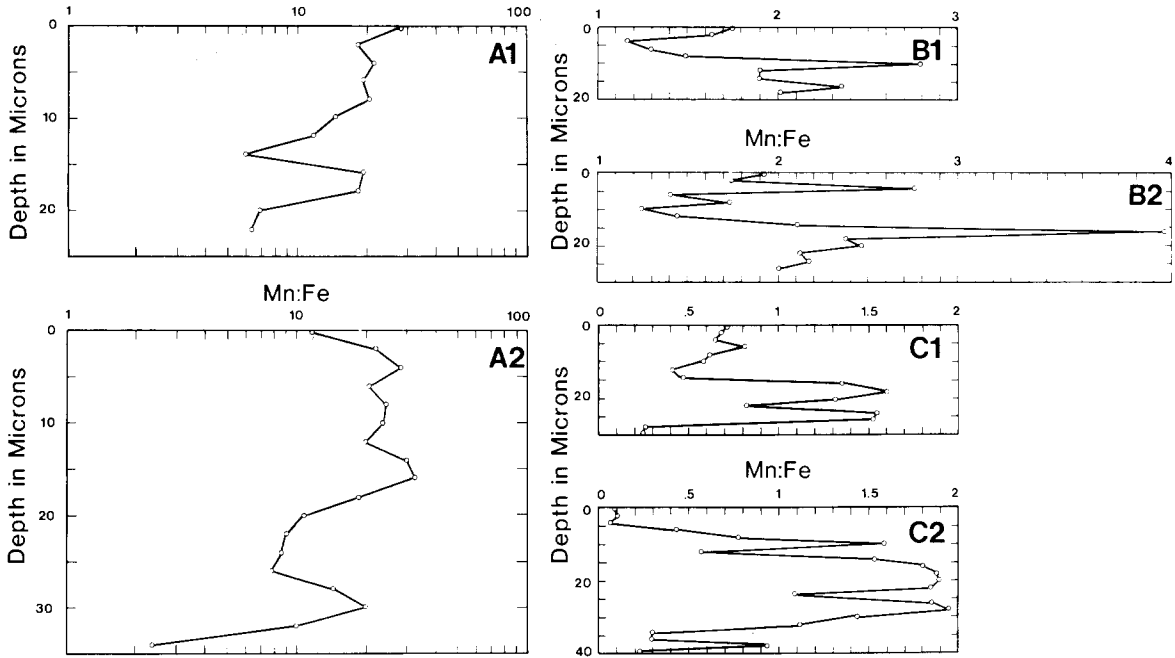
Fig.5. Representative electron microprobe profiles of varnish from Kitt Peak, AZ. *A1* and *A2* are from 1524 m, *B1* and *B2* from 1219 m, and *C1* and *C2* from 914 m.

surface Mn:Fe ratios with declining elevation is also found for elevation transects in the Mojave Desert, in the Owens Valley, and in the Panamint Range of eastern California.

Until now I have limited the results to varnishes collected from environments exposed to only atmospheric fallout, well above the surrounding soil surface. Another perspective is obtained by comparing the pH and Mn:Fe values of varnishes sampled from different microenvironments. Table I shows a negative relationship between varnish pH and Mn:Fe ratios for different types of varnishes from different settings. Where the microenvironment of the varnish surface is next to alkaline desert dust that has collected in rock crevices (crack orange varnish), or in contact with alkaline desert soils (bottom orange varnish), or in a subaerial position in an extremely alkaline area (surface orange varnish), the pH values are higher and the Mn:Fe ratio is lower. In contrast, where the microenvironmental setting is less alkaline, such as on rocks exposed only to atmospheric fallout,

TABLE I

pH values and Mn:Fe ratios of Holocene rock varnishes sampled from localities in western North America. Figure in parentheses represents the number of sites sampled

Varnish type	Varnish pH	Mn:Fe ratio
Surface black		
>1 m above soil (97)	7.3 ± 0.7	1.3 ± 1.7
near soil (15)	7.5 ± 0.5	0.9 ± 1.2
near lichens (8)	5.9 ± 1.0	1.0 ± 0.8
Surface dusky-red[a]		
>1 m above soil (35)	7.9 ± 0.7	0.3 ± 0.6
near soil (15)	8.1 ± 0.8	0.2 ± 0.6
Surface orange[b]		
>1 m above soil (27)	8.3 ± 0.5	0.1 ± 0.2
near soil (14	8.5 ± 0.4	0.0 ± 0.1
Crack black (30)	7.5 ± 0.7	1.0 ± 1.7
Crack orange (28)	8.8 ± 0.9	0.0 ± 0.2
Bottom dusky-red (2)	6.7 ± 0.3	0.4 ± 0.3
Bottom orange (24)	8.7 ± 0.7	0.0 ± 0.2

[a]Munsell color: 10R3/3 to 4/4 to 2.5YR3/2.
[b]Munsell color: 10R4/8, 2.5YR4/6 to 5/6 to 5YR7/6 to 7/8.

the pH is lower and the Mn:Fe ratio is proportionally higher. For the 177 sites represented in Table I, Mn:Fe is negatively correlated with pH with $r = -0.73$. The relationship is statistically significant at the 0.05 level.

Discussion

Desert soils and aeolian dust from arid regions characteristically have high pH values (Baas-Becking et al., 1960; Brady, 1974; Sidhu et al., 1976; Elvidge, 1979; Penkett et al., 1979). Data presented here on desert soils supports these generalizations. While only a small amount of regional data are available, what studies have been conducted on soil-aerosol relationships in the western U.S.A. (e.g. Gillette et al., 1980; Baronne et al., 1981; Cahill et al., 1981; Derrick et al., 1984; Young and Evans, 1986; McFadden et al., 1986; Reheis, 1988; Schlesinger and Peterjohn, 1988) show: (1) desert soils are a large source of aerosols in the western U.S.A.; (2) the desiccated surfaces of playas and playa margins are important contributors of aerosols to areas several tens of kilometers downwind; and (3) both playas and desert soils are rich in alkaline material.

The high correlations presented here between soil pH and varnish pH are consistent with varnishes being influenced by the surrounding surficial material, even though the varnish may be well above the soil surface on rock outcrops. The correlations of soil pH and Mn:Fe in varnish and of varnish pH and Mn:Fe in varnish are statistically significant. According to both the abiotic and biotic models of manganese enrichment in varnish, exposure to high pH values should inhibit the concentration of manganese in varnish. These results are consistent with both models. A confounding factor in this interpretation is that the pH measurement of the scrapings of varnish may not necessarily represent the pH of the varnish surface at the times of varnish accretion. It may not be possible to test this assumption with present technology.

The issue of a biotic vs. a purely physical–chemical mechanisms of manganese enhance-

ment has been discussed elsewhere (Krumbein and Jens, 1981; Dorn and Oberlander, 1982; Smith and Whalley, 1988; Dorn, 1988a), and is largely beyond the scope of this paper. The geographic distribution of different types of subaerial varnishes, however, is relevant to the issue of Mn:Fe microlaminations. It also lends support for the biotic model.

Manganese-rich (black) subaerial varnishes are common in both arid and humid areas. However, orange varnishes (Munsell color 10R4/8, 2.5YR4/6 to 5/6, 5YR7/6 to 7/8) occur almost exclusively in deserts. Orange varnishes have very little enrichment in manganese above the ambient levels in soils and airborne fallout, but are otherwise similar in structure and mineralogy to lamellate black varnishes (Potter and Rossman, 1977, 1979; Dorn and Oberlander, 1982). Orange varnishes tend to occur in subaerial positions where the environment is highly alkaline, for example next to saline playas. Orange varnishes are often interfingered with an amorphous silica glaze (e.g. Fisk, 1971; Oberlander, 1982; Curtiss et al., 1985), likely precipitated under conditions of high alkalinity. Orange varnishes are more common in the crevices of rocks, where accumulated alkaline dust fallout is in direct contact with the sides of the crevices, and orange varnish is common on the bottom sides of rocks in a desert pavement exposed to alkaline desert soils. Orange varnishes occur in environments outside of deserts in acid springs and acidic streams where chemolithotrophic bacteria and redox changes immobilize and mobilize iron (e.g. Crerar et al., 1978; MnKnight et al., 1988). These iron accumulations are distinct from orange rock varnishes in that clay minerals are not a major constituent; also, while microorganisms may play a role in the development of orange varnishes (Krumbein and Jens, 1981; Dorn and Oberlander, 1982), they are not required for the genesis of orange varnish. Microorganisms are required, in contrast, for the acid iron coatings (e.g. Crerar et al., 1978; McKnight et al., 1988).

I do not mean to underestimate the importance of local influences on the scale of

centimeters and μm. At any given varnish, this alkalinity — Mn:Fe relationship can be altered. Local sources of extreme alkalinity could inhibit Mn-oxidizing bacteria, as could local sources of acidity. For example, aeolian abrasion may remove a surface layer to a subsurface varnish. An orange crack varnish could have been recently exposed by spalling and the "surface" layer of varnish would inherit the lower Mn:Fe ratio developed in a crevice exposed to alkaline dust. Some microsites, such as places where water runs off or collects, generate their own microenvironments that are buffered against fluctuations in alkalinity and are slower to record environmental changes. Other reasons for discontinuities include different rates of varnish accretion over short distances and differential compression of subsurface varnish over time. The work of Dragovich (1984, 1988) highlights the need for care in interpreting microprobe data from varnishes sampled in different microenvironments. The presence of local effects and the fairly common occurrence of discontinuous laminations highlight the need for collecting multiple samples from a rock, from several rocks, and over a region. By analyzing only a few varnishes, highly misleading results might be obtained or alternatively, no clear pattern might be observed where a pattern actually does exist.

Concluding remarks on alkalinity and varnish chemistry

For the last century varnish researchers have concentrated on studying the black, manganese-rich coatings almost the exclusion of other types of varnishes (e.g. Merrill, 1898; White, 1924; Laudermilk, 1931; Blackwelder, 1948; Engel and Sharp, 1958). Potter and Rossman (1977) and Perry and Adams (1978) were the first to appreciate fully that varnish chemistry forms a continuum from those extremely enriched in manganese (where Mn:Fe ratio exceeds 1) to those reflecting the ambient crustal ratios (where Mn:Fe ratio is about 1:60).

Hunt and Mabey (1966), Butzer and Hansen (1968), Glazovskaya (1971), El-Baz and Prestal (1980), Oberlander (1982, 1988) and Dorn (1984) have previously recognized the association between the alkalinity of the environment and the degree of manganese enrichment in varnish. Data presented here from western North America demonstrate a clear relationship between the pH of the soil surrounding a varnish, the pH of a varnish, and the Mn:Fe ratio in the varnish itself. These empirical relationships support both the biological and physico-chemical explanations of manganese-enrichment in varnish, but the biological mechanism is strongly favored for many reasons including the geographic distribution of different types of varnishes.

Paleoalkalinity and varnish microlaminations

This section addresses the systematic nature of microchemical laminations in varnishes. If a paleoalkalinity signal is to be obtained, the consistency of varnish laminae requires assessment. The key to this issue is determining whether these layers repeat sequentially from place to place on a given rock (as first suggested by Perry and Adams, 1978), from exposure to exposure, from hill to hill in a single region, and from region to region.

Study sites and methods

The Coso volcanic field in eastern California (Duffield et al., 1980; Duffield and Bacon, 1981) was selected as the major region to determine whether changes in Mn:Fe ratios over depth yield a reproducible signal. Abundant K–Ar dated lava flows with stable constructional surfaces and excellent varnish development made the Coso region ideal. Another reason for selecting the Coso Range is the extensive work by G. I. Smith and collaborators on nearby Searles Lake (Smith, 1976, 1984; Smith et al., 1983; Phillips et al., 1988) provide a 3 million-year-plus record of sedimentation and lake-level in Searles Valley. Varnishes were also

collected from K–Ar basalt flows in the Cima (Turrin et al., 1985), Crater Flat (Vaniman et al., 1982; B.D. Turrin, pers. comm, 1988), and Lunar Crater (B.D. Turrin, pers. comm., 1988) volcanic fields.

Three methods were used to assess the microlaminations: electron microproble; energy-dispersive analysis of X-rays; and optical examination. Electron microprobe is superior to energy-dispersive analysis of X-rays in its quantitative precision and the automation needed to complete multiple transects. Optical examination is far inferior to both, because the optical record may not record just Mn and Fe concentrations. Color and opacity can be affected by mineralogy, clay content, as well as thin-section preparation.

Results and discussion

A total of 214 electron microprobe transects of varnishes were completed on varnish thin-sections from the Coso Range. Many of these probe profiles include data other than simply how Mn and Fe concentrations vary with depth. A full presentation of these data is beyond the scope of this paper. Most of these profiles are presented in Dorn (1985). Selected examples are presented here to illustrate certain key points.

The profiles in Fig.6 for the Coso Range represent long-term records of how Mn:Fe vary with depth in varnishes. Peaks are periods of high alkalinity; troughs are periods of low alkalinity. Approximate age is also given, based on the assumption of a constant rate of varnish deposition. The K–Ar dates from Duffield et al. (1980) provide the endpoint for the bottom of the varnish; the surface (present) provides the other end point.

The assumption of a linear rate of deposition with time is probably not valid, but it is a first approximation. Compression of the surface layer of varnish likely occurs upon burial. It is difficult to generalize under what conditions would promote rapid varnish growth. In the biogeochemical model of Dorn and Oberlander (1982), the limiting factor is the rate of manganese concentration by microorganisms. If the rate of manganese concentration is equal, then other factors such as the amount and mineralogy of dust fallout come into play. Still, normalizing all the transects to the K–Ar age assists in the visual presentation of the data. Smoothing functions such as running means were examined, but presenting the actual Mn:Fe ratios is desirable because each profile provides a unique "signature" that can be potentially matched to other profiles. Smoothing also obscures differences that may be important. Dorn (1984, 1985) presents other representative profiles in the Coso Range.

All microchemical transects of varnishes on Coso K–Ar volcanics about 1 m.y. and younger were averaged together and combined by normalizing Mn:Fe ratio and age. An "alkalinity index" from 0 to 1 was generated, where 0 represents the lowest Mn:Fe ratio in a given transect and 1 is the highest. This is to make comparable widely different Mn:Fe ranges, as seen in Fig.6. Age is normalized by the K–Ar age. Mn:Fe ratios were generated for every 5000 years by linear regression between two end points. By normalizing Mn:Fe and age different transects can be combined, as in Fig.7. The great standard errors generated from multiple microprobe transects and the smoothing effect are from: offsets between an assumption of a linear rate of growth and the reality of uneven growth rates; uneven compression of a surface layer that is too thick for its length of growth; some transects having a high enough rate of varnish accretion to record more fluctuations than less sensitive locations; and "noise" from previously discussed local factors affecting a varnish signal. Merged transects for each separate volcanic (like Coso 32 in Fig.7A) usually have a lower standard error on the alkalinity index for each 5000 yr regression and more amplitude than the average of all Coso transects (Fig.7B). This probably reflects the difficulty in assuming a constant rate of deposition and also that each individual transect reflects influences imposed on varnish on a small part of a single rock.
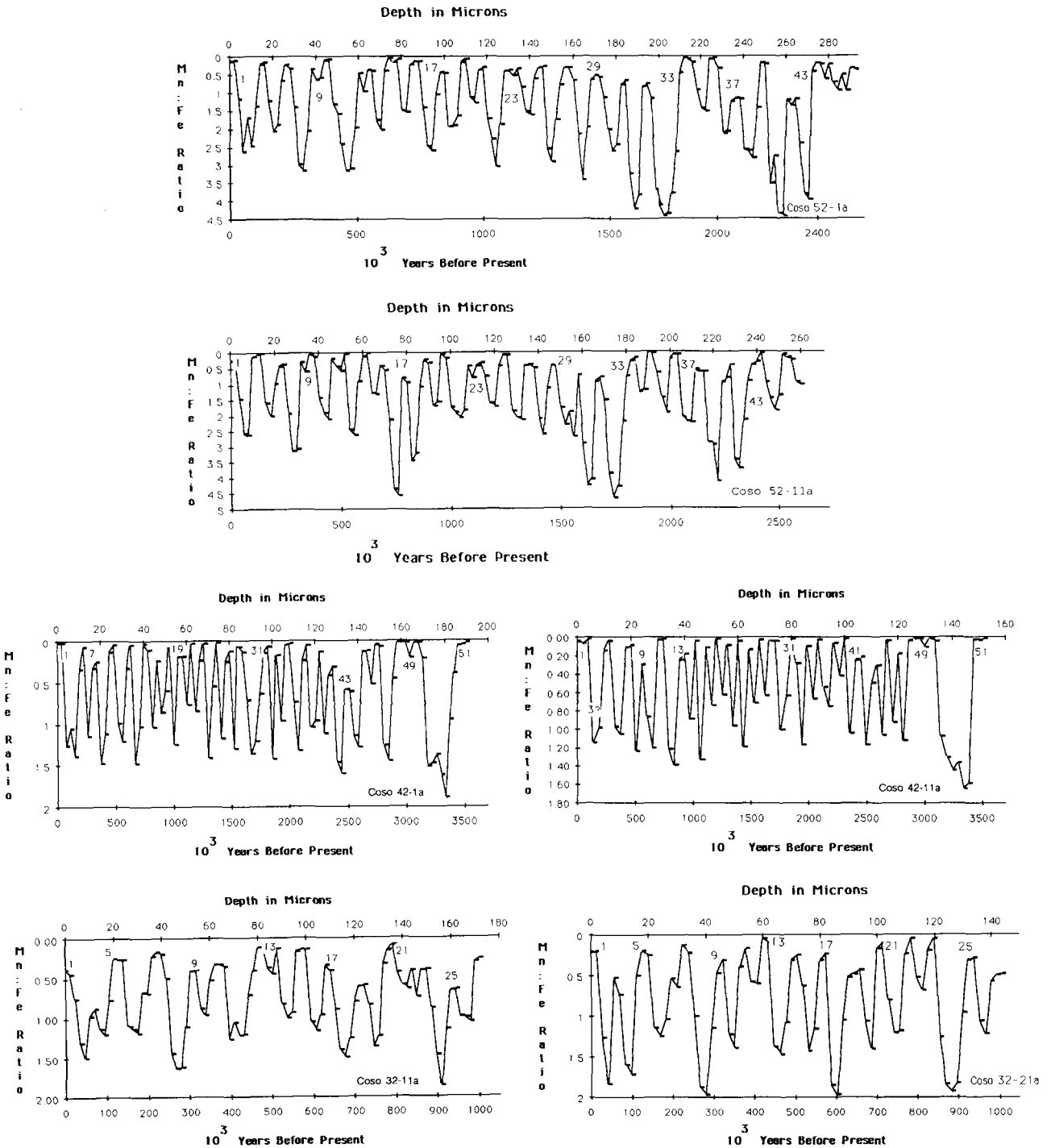
Microchemical laminations in rock var-

Fig.6. Representative microprobe profiles of varnish on constructional features of 3 K–Ar dated basalt flows older than 1 m.y. in the Coso Range (see Duffield et al., 1980 for K–Ar dating). Each transect has been normalized to the K–Ar age: Coso 32 (1.07 ± 0.12 m.y.); Coso 29 (2.06 ± 0.32 m.y.); and Coso 52 (2.51 ± 0.05 m.y.). The transects are identified by their K–Ar number (e.g. Coso 32) and their microprobe transect number (e.g. 11a) on the lower right corner of each profile. The peaks have been assigned an arbitrary notation of sequentially odd numbers to indicate periods of more alkaline conditions. The numbering provides the reader the subjective identification of peaks by the author. Note the greater sensitivity of the transects for Coso 52.
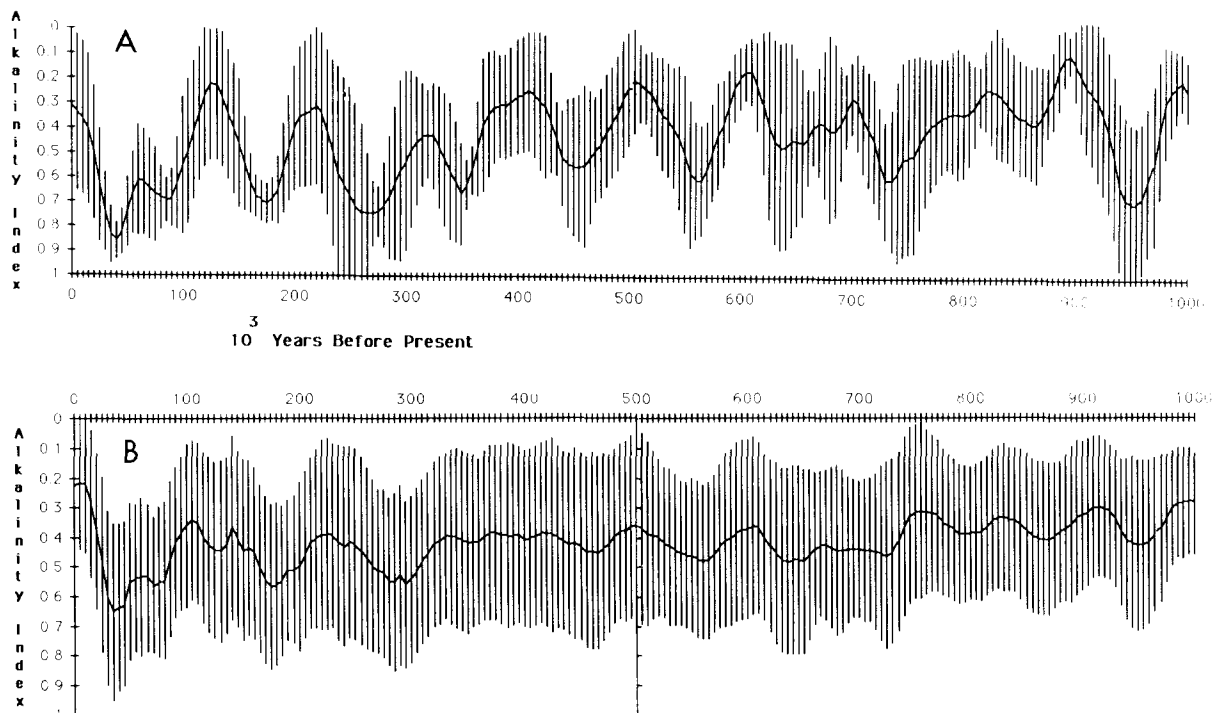
Fig.7. Merged Mn:Fe profiles in the Coso Range for the last million years. 17 transects have been averaged in A. for Coso 32 (1.07 ± 0.12 m.y.). B. average of all 214 transects completed for the Coso Range. The alkalinity index represents normalized Mn:Fe values. 0 is the lowest ratio and 1 is the highest ratio for each separate transect. Years before present (in k.y.) is derived by normalizing depth to K–Ar age. Alkalinity index values were then regressed for every 5000 yr. The line indicates the average alkalinity index value and the bars represent 2 standard errors. As in Fig.6, the peaks represent periods of greater alkalinity. Note that the surface layers of the varnish have not been compressed upon burial, and are unrepresentatively thick (and occupy proportionally too much "time").

nishes on subaerial exposures in the Coso Range provide a fairly consistent record of Mn:Fe fluctuations for more than a million years. The Mn:Fe ratio is an indicator of alkalinity, according to both the biological or abiotic model of manganese enrichment and results in the first section of this paper. An important concern is what level of resolution of environmental change can be obtained. The sensitivity of most of the Coso Range varnishes appears to be from 10,000 to 100,000 yr. The peaks (alkaline phases, dry) and troughs (less-alkaline, more moist) alternate regularly on this time scale. An analysis of the number of peaks and troughs recorded provides a useful comparison (Table II). The younger varnishes reveal a more detailed record of fluctuations; this is reflected in the interval between the peaks being less in Table II. This is probably

from younger volcanics having more preserved constructional surfaces with microsites that would accumulate varnish rapidly. This is an important reason why Fig.7B has a higher standard error than Fig.7A. Figure 7B combines records of different sensitivities. As the constructional surfaces of basalt flows spall off over time, they leave fewer microsites to choose from that would tend to record a detailed record. A 1 + m.y.-old volcanic, like Coso 32 in Fig.7A, is composed of few constructinal remnants to choose from.

The results of this study indicate that the Mn:Fe ratio in subaerial varnish is negatively correlated with alkalinity. This is related to either the biotic or abiotic mechanism of manganese enhancement; but what factor(s) might govern the fluctuations in alkalinity levels recorded in Coso varnishes? For any

TABLE II

Summary of varnish microchemical laminations on Coso Range volcanics. Coso volcanics are identified and dated by number in Duffield et al. (1980) and Duffield and Bacon (1981). When no standard error is listed, all the profiles had the same number of peaks for a given volcanic, excluding profiles that were discontinuous or lacked laminations due to local influences discussed in the text. A * indicates that the flow surface where the varnish was sampled may not be constructional, but the cation-ratio age indicates it is within 10% of the K–Ar age.

Number of Coso Flow	K–Ar date in ka	Mean (and standard error) of number of peaks (low Mn:Fe) in profiles	Mean interval between peaks in ka
2a*	39 ± 33	1	39
1*	81 ± 8	3	27
6*	101 ± 33	3	34
28	140 ± 89	3.4 ± 0.7	41
36	234 ± 22	5	47
15*	293 ± 35	5	59
32	1070 ± 120	13.5	79
33*	1080 ± 60	12.8 ± 1.3	84
52	2510 ± 50	24 ± 0.8	104
42	3670 ± 160	25.6 ± 0.9	143

given rock, the answer is a combination of local and regional factors. When a group of similar varnish signals over a region is considered, the answer could reasonable be the regional levels of aeolian alkalinity.

The level of regional aeolian alkalinity recorded in Coso rock varnishes is likely controlled by the past hydrology, pedology and biogeography of the region surrounding the Coso Range. These factors are in turn linked to past climates. Mayer and Anderson (1984) found the flux of aeolian carbonate is directly related to the proximity of local playas, dry lake bottoms containing abundant carbonates. They concluded that the "magnitude of variation in carbonate flux may be about a factor of eight" when environments near playas are compared to areas away from playas. Baronne et al. (1981) determined that the historic desiccation of Owens Lake generated significant amounts of alkaline aeolian material in the surronding region. In examining a chronosequence of soils on volcanic flows of the Cima field, CA, McFadden et al. (1986, p. 361) note: "loess events are attributed to past changes in climate, such as the Pleistocene-to-Holocene climatic change, that periodically caused regional desiccation of pluvial lakes, reduction

of vegetation density and exposure of loose, unconsolidated fine materials ... extensive saline playas which developed in the Mojave Desert during the Holocene are a likely source of much of the carbonate and soluble salts ..." Reheis (1988, p. 207) found that "$CaCO_3$ content is strongly influenced by local factors such as proximity to playas and carbonate-rich alluvial fans". As the vegetation cover declined in the southwest from the late-Pleistocene to the Holocene (Spaulding et al., 1983; Wells and Woodcock, 1985), the exposure of alkaline desert soils undoubtedly increased the abundance of alkaline aeolian material.

The Coso Range is surrounded by basins that have alternated between pluvial lakes and saline playas throughout the Quaternary (Smith et al., 1983; Smith and Street-Perrott, 1983; Smith, 1984). When these lakes were present, the vegetation cover over soils was probably more extensive. When the lakes were dry or nearly dry, alkaline playas were exposed and the vegetation cover over the upland soils likely waned, each providing a more abundant source of aerosols that likely fell on the neighboring Coso Range. While long distance airborne transport of desert dust is another aeolian influence (Muhs, 1983; Rabenhorst et

al., 1984), its alkalinity signal would probably be complementary (Derrick et al., 1984) and less important than more local influences such as playas (Mayer and Anderson, 1984). Thus, the regional climate, through influencing the past hydrology, pedology and biogeography has probably controlled fluctuations in the abundance of alkaline aeolian fallout. These fluctuations have, in turn, probably influenced the concentration of manganese in varnish, when viewed from a perspective of many, many different varnish exposures. It will be instructive to compare Mn:Fe curves of Coso varnishes with the new Searles Lake-level curve refined by ^{36}Cl dating techniques (Phillips et al., 1987).

Rock varnishes on volcanics K–Ar dated at about 1 m.y. in other areas of the Great Basin were analyzed to assess how similar the microlamination pattern is to the Coso Range. Figure 8 presents representative profiles from these areas. Typically, 12–13 events of reduced alkalinity are recorded from slowly accumulating varnishes over the last million years. This is similar to what was found for the Coso Range (Table II; Figs.6 and 7). The simplest interpretation is that the climatic changes controlled the alkalinity fluctuations by covering playas and increasing vegetation cover during moisture effective times. During more arid times, playas and soils were exposed to deflation. The fluctuations appear to be concordant in the western Great Basin, at least on the level of resolution recorded by varnish microlaminations.

An unresolved issue is the paleoenvironmental interpretation of peak heights and trough depths. It is unlikely that there is a linear relationship between Mn:Fe and lake level or other paleoclimatic proxy indicators. The results presented earlier suggest that relative Mn:Fe fluctuations should provide a general approximation of the relative alkalinity experienced. However, it would be highly questionable to compare the absolute Mn:Fe ratios to a specific climatic condition.

A highly speculative subject that should at least be touched upon is whether the microchemical laminations in varnish can be cor-related with other long-term records of environmental change. The most tested and detailed record of long-term Quaternary climatic changes is the deep-sea oxygen-isotope record. Figure 9 compares the orbitally-tuned record of marine oxygen-isotope stages (from Martinson et al., 1987) with the average of varnish Mn:Fe transects from the Coso Range. The two records appear to match roughly, but no causation is implied. The marine record is of global ice volume; the varnish is of alkalinity fluctuations in a specific area. Until independent dating of discrete varnish layers is developed or until varnish on glacial landforms or marine terraces are examined, a correlation between a local continental record in varnish (based on a variable time scale) and the well-documented marine record is nothing more than speculation.

Another long-term paleoclimatic record is the speleothem chronology of glacial and interglacial periods in western North America, based on uranium-series dating (Harmon, 1979). Like the deep-sea record, there appears to be a rough match in climatic trends (Fig.9). Unlike the marine-varnish comparison, this similarity may hve a more significant meaning. When the glacial periods occurred in western North America, the basins surrounding the Coso Range were occupied with pluvial lakes (Smith, 1984), even if there was some time-transgressive offset. The supply of aeolian alkaline aerosols is probably related to the glacial chronology. There is a tremendous difference, however, between the excellent time resolution of the speleothem record and the uncertain time-scale of the varnish based on a linear rate of accretion and K–Ar dates, some with substantial age-uncertainties.

Conclusion

Micron-scale alternations in Mn:Fe ratios in varnishes are interpreted here as a record of fluctuations in the levels of aeolian alkalinity. The concentration of alkaline aerosols is eventually governed by deflation in source regions. The vegetation cover over alkaline
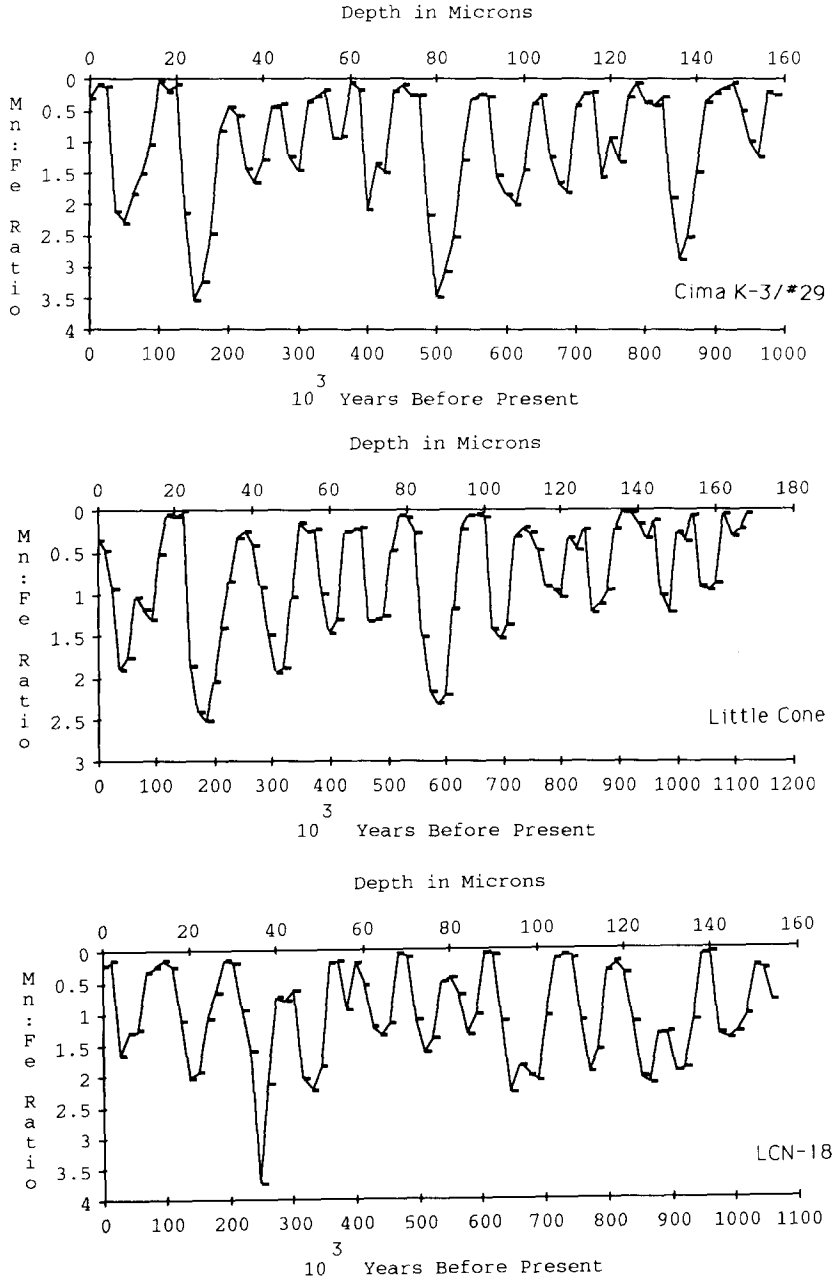
Depth in Microns

Cima K-3/#29

10^3 Years Before Present

Depth in Microns

Little Cone

10^3 Years Before Present

Depth in Microns

LCN-18

10^3 Years Before Present

Fig.8. Microprobe profiles of varnishes on constructional surfaces of K–Ar dated basalt flows in the western Great Basin from the Cima volcanic field in CA (Cima flow #29/K-3 at 0.99 ± 0.07 m.y. from Turrin et al., 1985), Crater Flat volcanic field in NV (Little Cone at 1.11 ± 0.30 from Vaniman et al., 1982), and Lunar Crater volcanic field in NV (LCN-18 at 1.09 ± 0.08 in Turrin and Dorn, in prep.).

desert soils and amount of water over saline playas have fluctuated widely in the southwestern United States throughout the Quaternary. Both vegetation cover and hydrology are controlled mostly by the regional climate. However, there are time-transgressive relationships between climate and vegetation changes (cf. Davis, 1984) and hydrologic
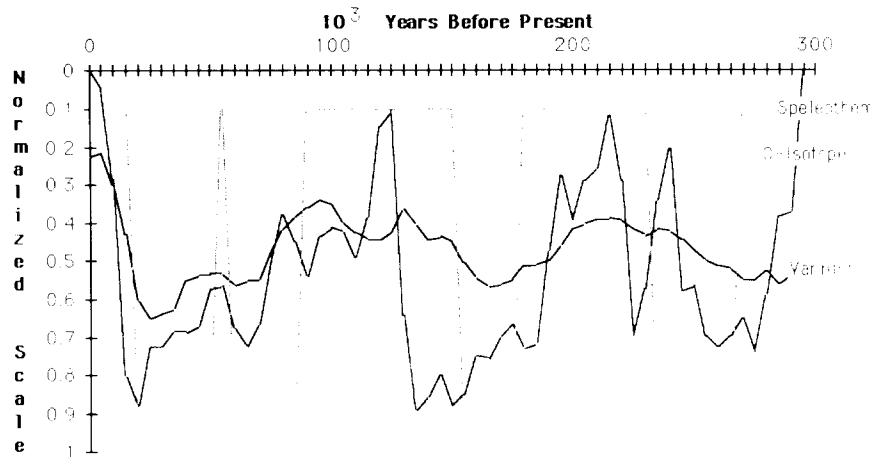
Fig.9. Comparison of different paleoenvironmental records. The varnish signal is the same as the alkalinity index in Fig.7B, modified by compressing the normalized period from 0–20 k.y. to 0–10 k.y. This is to adjust for the lack of compression in the upper layer of varnishes. The period from 0–20 k.y. has low Mn:Fe ratios that likely correspond to the Holocene starting about 10 k.y. (Smith and Street-Perrott, 1983; Spaulding et al., 1983; Wells, 1983; Smith, 1984). The O-isotope signal is the marine oxygen isotope record in Martinson et al. (1987). The normalized scale is based on the least-ice signal assigned a value of 0 and the most ice-signal assigned a value of 1; all other values were adjusted in a linear fashion. The time scale is orbitally-tuned. The speleothem signal is from Harmon (1979). The glacial phase is assigned a value of 0.9 and the interglacial phase is assigned 0.1. The speleothem time scale is from uranium-series dating.

changes (cf. Smith, 1984; Winograd et al., 1988). Still, given the 10^4 time scale over which the varnish laminations form, in more moist (or colder) climates the potential sources of alkaline dust probably decreased substantially. In drier (or warmer) climates, the abundance of alkaline dust probably increased.

On K–Ar dated volcanics of the Coso, Cima, Lunar Crater and Crater Flat volcanic fields of eastern California and western Nevada, systematic microlaminations in rock varnish have been documented. Despite the presence of some confounding problems, the consistency of this record is demonstrated over the Quaternary for multiple samples. These varnishes recorded fluctuations in the alkalinity level in aeolian fallout that have likely been caused by changes in the vegetation and hydrology in the western Great Basin.

The major limitations of the varnish record are in sorting out the many discontinuous or terminated Mn:Fe profiles caused by local influences and the inability to verify the age of any given layer in varnish profiles that record regional alkalinity fluctuations, other than through the assumption of a constant rate of accretion from the K–Ar age of the underlying rock. It may be possible in the future to date these layers, perhaps by volcanic ejecta incorporated into varnish (cf. Harrington, 1988), but for now interpretations must be based on sequences of alkalinity fluctuations.

There are many uses of this type of record. The microlaminations provide a record of changes in atmospheric aeolian conditions over time. It is one of the longer, continuous records of environmental changes in a continental setting, and a lengthy record that is not tied to a particular location like a lake basin. The laminations can be used to deduce general environmental fluctuations associated with a sequence of geomorphic events (cf. Dorn, 1988b). They may, with great care and abundant profiles, be used to correlate landforms in a local area or to assess relative ages.

Acknowledgements

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