ated magma in the roots of the system. Basaltic magma undoubtedly provides heat and mass to sustain the silicic magma reservoir at Mount Pinatubo, as indicated by the mingling and mixing of basalt and dacite in eruptive products from the volcano during the last 30,000 years (*21*). In this context, mantle-derived basaltic magma must be viewed as the ultimate source of both sulfur and CO_2 in the silicic magma reservoir (*23*). Vapor accumulation in the roof zone of the system is consistent with chemical and stable isotopic data for hydrothermal fluids and gases that indicate the introduction of a magmatic component into the overlying hydrothermal system (*8*). Upward concentrations of exsolved vapor could form through various processes such as (i) sidewall crystallization and buoyant rise of derivative silicic liquids with entrained vapor bubbles and (ii) diapiric rise of bubble-rich foams (*24*).

The vapor accumulation hypothesis can be tested by monitoring both CO_2 and SO_2 emissions from volcanoes during eruptive and noneruptive degassing. Many volcanoes in subduction-related arcs are similar to Mount Pinatubo in emitting quantities of SO_2 that greatly exceed preeruptive dissolved SO_2 contents (*3, 6, 7*), and there is only a weak correlation between the magnitude of an eruption (measured by tephra volume and eruption column height) and the mass of SO_2 emitted (*1*). In some cases, direct measurements of CO_2 in coerupted volcanic gases indicate that bulk magmatic CO_2 contents exceed the amounts that could be dissolved at crustal pressures (*7*). Magmas in crustal reservoirs probably range from those that are undersaturated, to those that are saturated with C-O-H-S vapor, to those that have accumulations of vapor. Within this continuum, magmatic systems with the highest fluxes of hydrous, SO_2-bearing vapor from underlying mafic magma have the greatest potential for injecting climate-altering quantities of SO_2 into the stratosphere during explosive volcanic eruptions.

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17. The vapor would also have contained ~1 mol% chlorine on the basis of the dissolved chlorine contents of glass inclusions (*8*). Only minor H_2S (~0.1%) would have been present because of the high oxygen fugacity of the Pinatubo magma (*5*).
18. This value is the unweighted mean of data reported by the following: G. J. S. Bluth, S. D. Doiron, C. C. Schnetzler, A. J. Krueger, L. S. Walter, *Geophys. Res. Lett.* **19**, 151 (1992); W. G. Read, L. Froidevaux, J. W. Waters, *ibid.* **20**, 1299 (1993); R. D. McPeters, *ibid.*, p. 1971. It is probably a minimum estimate of the total SO_2 emission because of the possible early removal of some SO_2 by scavenging on ash (*8*).
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20. We chose 260 MPa for all vapor saturation and degassing calculations on the basis of the glass inclusion data and experimental constraints (*11*). This value ignores the minor contribution from SO_2 (P_{SO_2} = 11 MPa) for simplicity. We calculated pressure-volume-temperature properties and fugacities of vapor species using the Redlich-Kwong equation of state.
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Evidence from Paleosols for the Geological Antiquity of Rain Forest

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Kaolinitic claystones in Paleozoic paleokarst underlying the Middle Pennsylvanian Fort Scott Limestone near Drake, Missouri, contain abundant fossil root traces. These include a surficial root mat as well as stout, woody, deeply penetrating root traces: a rooting pattern similar to that under rain forest. Also similar to soils of rain forest is the deeply weathered clay of the paleosol, in which minimal amounts of nutrient bases remain. Forest communities adapted to oligotrophic clayey substrates in humid climates existed at least 305 million years ago.

Fossil evidence of rain forest is sparse, because most rain forests colonize sites too well drained to favor the preservation of fossil plants and soils insufficiently calcareous to preserve bone and shell (*1*). Exceptional fossil biotas of well-drained wet forest as old as the Eocene are known from paleokarst (*2*). Fossil fruits, seeds, and leaves similar to those of Indo-Malesian rain forest also are known no further back in time than the Eocene (*3*). Fossil plants of Carboniferous coal measures have been interpreted as the remains of rain forest, but despite the discovery of a great diversity of Carboniferous plants including vines (*4*) and epiphytes (*5*), no Carboniferous plant assemblages are physiognomically comparable to Guineo-Congolian, Amazonian, or Indo-Malesian rain forest (*6*). Given this sparse fossil record and preservational biases, rain forests of the past may be better recognized from fossil soils than from fossil plants. Rain forests have high productivity and stature despite their oxidized oligotrophic clayey soils (*7*).

An ancient example of a deeply weathered and oxidized paleosol containing large woody root traces is in the Farnberg pit, 5 km southwest of Drake, Missouri (*8*). The pit is an excavation for refractory clay in the Cheltenham Formation. These clays and sandstones fill paleokarst with 46 m of relief into the Ordovician Jefferson City Dolomite (*9*). Elsewhere in this region this paleokarst is incised into Burlington Limestone, and so is younger than the Mississippian (*9*). The Farnberg clay paleosol is only 15 cm below the top of the Cheltenham Formation, which is overlain by the Fort Scott Limestone of Middle Pennsylvanian age [middle Desmoinesian (*10*)], correlative with the European Moscovian and Westphalian D some 305 million years old (*11*).

The Farnberg clay paleosol is overlain abruptly by gray clayey siltstone containing

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fossil logs, one of which is at least 179 cm long, 4 cm thick, and 28 cm wide. The width of compressed logs is thought to equal their original diameter (*12*). The clayey surface horizon includes a dense mat of root traces up to 3 mm wide that define a crude platy ped structure (Fig. 1B). Stout root traces up to 6 mm in diameter with the silky surface texture characteristic of wood were seen penetrating the paleosol to depths of up to 210 cm (Fig. 1, A and C).

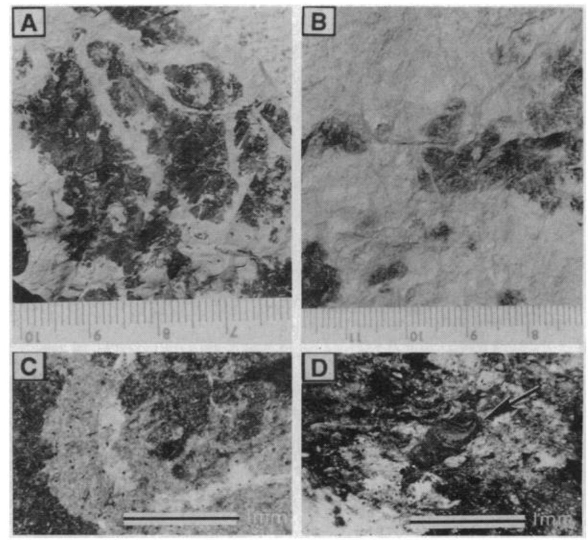

Fig. 1. Fossil root traces and associated features in the Middle Pennsylvanian, Farnberg clay paleosol, near Drake, Missouri. (**A**) Vertically penetrating drab-haloed root traces in the red (dark) matrix of the upper B horizon (specimen R981). (**B**) Subhorizontal root traces in the A horizon (R980). (**C**) Petrographic thin section of root trace (white) with drab-halo (dark gray) in oxidized soil matrix (dark gray) at a depth in profile of 210 cm (R989). (**D**) Colloform clay fill of root trace at depth in a profile of 130 cm (R987). Scales are graduated in millimeters for hand specimens (A and B) and are 1 mm for photomicrographs (C and D).

Fossil root traces in the paleosol are surrounded by gray to green colored clay (Fig. 1, A and C). Such drab halos are commonly produced in paleosols as a result of the anaerobic decomposition of remnant organic matter shortly after burial (*13*). This paleosol also may have been reddened by the dehydration of iron hydroxides to hematite during burial (*13*). Also likely is lithostatic compaction from overlying Paleozoic rocks 1.2 km thick according to estimates from regional isopachs and 1.5 km thick according to the regional variation in moisture of coals (*14*). Compatible with such a depth of burial is the high volatile bituminous rank of the Mulky Coal at the stratigraphic level of the Farnberg paleosol in Missouri (*15*) and the white to cream color of Pennsylvanian conodonts in this region (*16*). On the basis of standard compaction curves (*17*), the clayey paleosol is probably 76% of its former thickness. These alterations after burial do not alter the conclusion that large woody plants grew here in a thick clayey soil during Middle Pennsylvanian time.

Also little altered during burial was the chemical and mineral composition of the Farnberg paleosol, which is remarkably low in weatherable bases, represented by lime, magnesia, soda, and potash (Fig. 2 and Table 1). The analysis of clays of the Farnberg paleosol by x-ray diffraction revealed mostly well-ordered kaolinite. All of its characteristic peaks from 0° to 30° 2θ were sharp, and there was little response to glycolation. Small amounts of submicroscopic hematite were also identified from x-ray peaks. No halloysite or diaspore could be identified by differential thermal analysis, but small amounts of dickite and metahalloysite could not be ruled out (Table 2). The absence of diaspore and illite is surprising, because diaspore is common in underlying claystones and illite is common in the capping clayey siltstone (*9*). In modern rain forest soils, kaolinite is maintained by biological activity against chemical equilibrium (*18*).

Some refractory clays in Missouri may have been created by hydrothermal fluids responsible for Pennsylvanian to Permian mineralization of Mississippi Valley type (*19*). Local high-temperature geothermal anomalies apparent from conodont (*16*) and coal maturation studies (*15*) or fluid inclusions of sphalerite mineralization (*20*) are all remote from the Farnberg pit, in southeastern Missouri and southern Illinois and Indiana. Even more distant is the likely source of mineralizing brines in overthickened Alleghenian sediments in Arkansas and Tennessee (*21, 22*). The Farnberg clay paleosol has much lower values of Co, Cr, Cu, Li, Ni, and Zn than clayey alteration in the mineral districts (*21*), and low Ba/Sr ratios indicate only modest leaching (*23*). The eastern end of the Farnberg paleosol is both overlain and underlain by sandstone (Fig. 3), including rare unaltered amphibole, in a much broader and shallower paleokarst depression (9) than the cherty, sulfide-bearing, narrow, cylindrical, breccia pipes thought to have been hydrothermally altered (*19, 21*). Hydrothermal alteration is difficult to envisage from the distribution of the Farnberg paleosol sandwiched by illitic shale and diaspore clays and for correlative kaolinitic claystones sandwiched by illitic shaly sequences far to the north, east, and west (*9, 10*). This is not to say that the Farnberg paleosol has been unaffected by geothermal heating attendant on burial. The Mulky Coal of Missouri, which also underlies the Fort Scott Limestone, has a moisture-free calorific value of 6.4 kcal/g (12,795 Btu), a moisture of 10.5%, and a

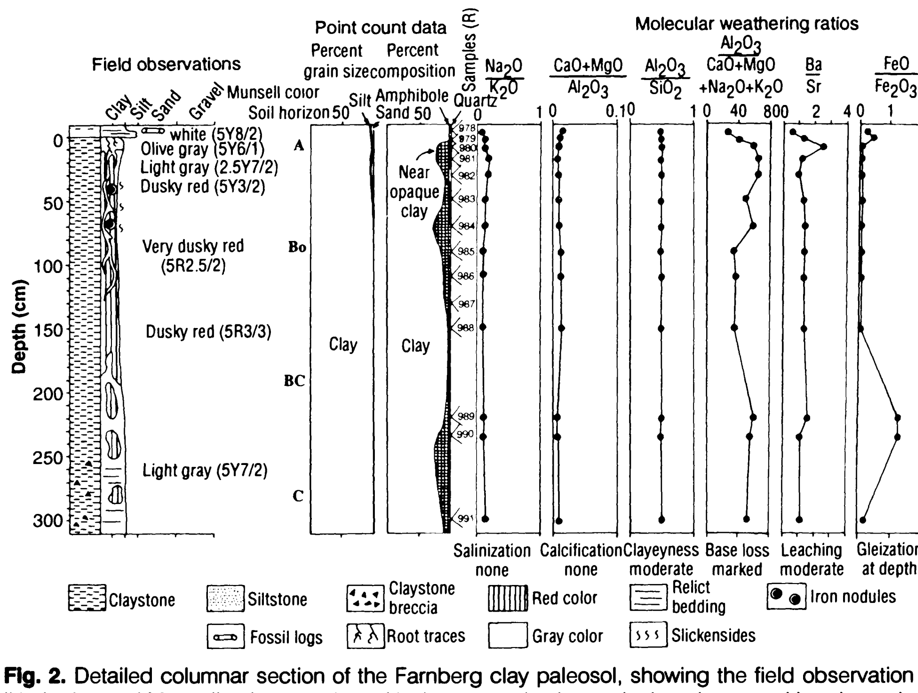

Fig. 2. Detailed columnar section of the Farnberg clay paleosol, showing the field observation of lithologies and Munsell color, together with data on grain size and mineral composition determined by the point counting of petrographic thin sections and selected molecular weathering ratios calculated from chemical analyses by atomic absorption.

Table 1. Major (weight %) and trace element (parts per million) chemical analysis of the Farnberg clay paleosol (*37*), with depths in centimeters given in parentheses.

Element	Sample													
	R978 (+5)	R979 (−2)	R980 (−7)	R981 (−17)	R982 (−30)	R983 (−50)	R984 (−70)	R985 (−90)	R986 (−110)	R988 (−150)	R989 (−220)	R990 (−235)	R991 (−300)	error (±2σ)
SiO_2	44.94	44.74	42.45	42.26	43.61	42.90	43.22	43.83	42.75	42.25	43.77	44.70	44.39	0.50
TiO_2	1.11	0.75	1.02	0.86	0.95	1.06	0.68	0.78	0.81	1.11	1.00	0.86	0.81	0.04
Al_2O_3	37.82	37.85	37.86	35.88	37.21	36.59	36.82	36.08	36.65	35.98	37.18	37.36	37.66	0.34
Fe_2O_3	0.62	0.60	3.61	6.15	3.34	4.89	4.95	4.78	4.94	6.07	2.15	0.23	1.55	0.30
FeO	0.08	0.13	0.24	0.24	0.13	0.29	0.24	0.18	0.18	0.18	1.22	0.13	0.13	0.16
MnO	0.01	0	0	0	0	0	0	0	0	0	0	0	0	0.01
MgO	0.16	0.13	0.08	0.07	0.07	0.09	0.09	0.13	0.12	0.13	0.08	0.08	0.10	0.10
CaO	0.10	0.08	0.08	0.09	0.09	0.08	0.07	0.08	0.09	0.09	0.08	0.09	0.10	0.20
Na_2O	0.03	0.04	0.02	0.02	0.02	0.03	0.02	0.03	0.03	0.03	0.02	0.02	0.02	0.10
K_2O	0.63	0.47	0.24	0.15	0.17	0.30	0.25	0.48	0.42	0.45	0.21	0.24	0.22	0.02
P_2O_5	0.24	0.27	0.13	0.10	0.07	0.09	0.07	0.09	0.07	0.10	0.07	0.13	0.13	0.01
LOI	13.99	14.36	13.88	13.82	13.91	13.51	13.56	13.29	13.52	13.18	13.73	14.51	14.50	—
Total	99.73	99.42	99.61	99.64	99.57	99.83	99.97	99.75	99.58	99.57	99.51	98.35	99.61	—
Ba	84	72	84	84	72	96	96	120	108	120	84	84	96	24
Co	31	30	26	24	26	25	27	26	28	27	26	27	26	2
Cr	248	666	157	224	207	203	218	210	198	270	234	211	257	4
Cu	24	34	13	13	13	12	13	13	13	13	12	14	13	4
Li	119	200	116	92	92	123	124	121	136	143	137	136	143	2
Ni	46	56	38	46	43	61	61	70	83	85	51	45	46	2
Rb	14	10	5	3	2	7	6	12	10	11	3	3	3	4
Sr	92	34	20	48	44	46	43	57	53	56	34	48	57	8
Zn	15	16	12	13	12	16	16	18	23	21	13	11	14	6

volatile matter of 39.5%, averaged from four analyses (*15*). On the basis of standard coalification curves (*24*), these values correspond to a maximum burial temperature of about 40°C. Regional low-color alteration of conodonts [conodont alteration index (CAI) of 1] indicates temperatures less than 50° to 80°C (*16*).

The Farnberg clay paleosol presents an example of an extremely oligotrophic clayey soil that supported forest and that is similar to modern soils of rain forest in Brazil, Zaire, and Malaysia (*25*). Such low-nutrient tropical soils are generally classified as Oxisols and Ultisols (*26*) or as Acrisols, Ferralsols, and Nitosols (*27*). We could not discern any significant subsurface clay enrichment like that of Acrisols, Nitosols, and Ultisols from point counting petrographic thin sections (Fig. 2) or clay mineralogical work (Table 2), although there are a few spectacular examples of clay skins (Fig. 1D). Also lacking are the spherical micropeds, high iron content, and abundant social insect burrows seen in many modern Ferralsols and Oxisols (*28*). The most similar modern soils are Acric Ferralsols (*27*) and Acrudox or Acroperox (*26*).

The deep weathering of bases and abundant kaolinite is characteristic of soils of very humid climates, with more than 2000 mm of mean annual rainfall (*29*). Ferruginous concretions (*9*) are evidence of climatic seasonality, but the Farnberg paleosol has no organized vertic structures of the sort found in soils of highly seasonal climates (*21*).

Vegetation of the Farnberg paleosol was mainly gymnosperm trees, on the basis of the stout, copiously branched, woody root traces. Nothing like the stout fleshy roots of tree lycopsids was seen, although a likely specimen has been reported in diaspore clay of the Cheltenham Formation only 16 km north of the Farnberg pit (*9*). Tree lycopsids, calamites, and ferns were widespread in Pennsylvanian lowland vegetation (*30*) and in the swampy bottoms of Early Pennsylvanian paleokarst in northeastern Illinois (*31*). The paleokarst fossil floras also include probable upland taxa seldom seen in coal measures, including the seed ferns *Megalopteris* and *Lesleya* (*31*). Broadleaf seed ferns are also found in association with Early Permian noncalcareous red paleosols in Texas (*32*) and Arizona (*33*). Conifers are another plant group of likely upland habitats, rare in lowland fossil plant assemblages of Middle Pennsylvanian age as fusinized fragments (*34*). This preservation

Table 2. Differential thermal analysis of clays in the Farnberg clay paleosol (*38*).

Parameter	Sample					
	R980	R982	R983	R986	R988	R989
Percent kaolinite	84	89	83	86	83	86
OH/H_2O ratio	49.0	37.5	48.3	36.2	38.6	46.1
Corrected temperature of thermal decomposition (°C)	−2.0	−9.0	−8.7	−11.6	−10.5	−5.1
Activation energy (kJ/mol)	132.6	137.5	138.4	141.8	132.7	140.5
Temperature exothermic peak (°C)	>999.6	995.9	>999.5	991.4	989.4	>998.95
Intensity exothermic peak (°C)	0.6	0.98	0.58	1.0	0.81	0.65
Width exothermic peak (°C)	38.8	39.2	36.4	55.8	72.8	34.1

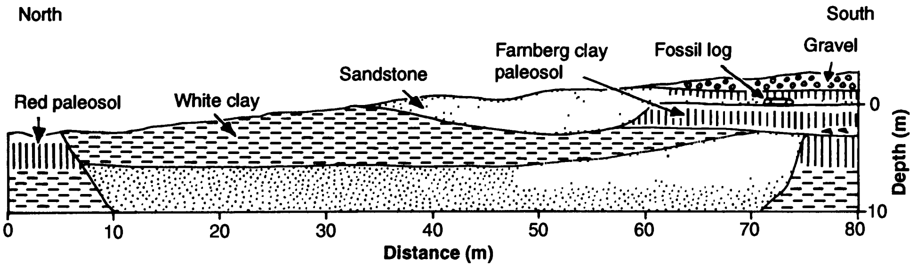

Fig. 3. A north-south cross section, true to scale of lithologies associated with the Farnberg clay paleosol in the eastern wall of the Farnberg pit.

may indicate that conifers preferred seasonally dry soils more prone to forest fire than is likely for the Farnberg paleosol.

Thick cross-bedded sandstone interbedded with the Farnberg paleosol (Fig. 3) is a paleochannel and indicates a connected drainage through this upper portion of the partly filled paleokarst. The Farnberg paleosol formed on well-drained valley slopes of this polje between hills of karst and chert of the Ozark highlands to the south and extensive seas and swamps of lowlands to the north (*9*). Parent material of the Farnberg paleosol included flint clays remaining from paleokarst dissolution, slope wash, and a small amount of fresh fluvial deposits and air-fall dust. Relict bedding can be discerned in the basal part of the profile, but none remains in the rooted horizons (Fig. 2). By comparison with modern soils (*35*), this observation indicates at least tens of thousands of years of soil formation. Soil formation could have extended for millions of years to create such deeply weathered material, but it is likely that a substantial component of this chemical weathering occurred in preexisting soils that were eroded to provide the parent material of the Farnberg paleosol (*9*).

Fossil trees and forested paleosols are known to be as ancient as the Middle Devonian (*22*), but none of these Devonian paleosols are nearly as base-depleted as the Farnberg paleosol. Quartz sandstones with large fossil root traces represent other kinds of oligotrophic soils (Spodosols, Dystrochrepts, and Quartzipsamments) and, like the Farnberg paleosol, are also no older than the Carboniferous (*22, 36*). No Devonian or older orthoquartzites or bauxites are known to contain large woody root traces. These are difficult substrates for plants, and the coevolution of nutrient-conserving ecosystems capable of sustained growth on such soils may have taken some time. Whether it took as long as the Late Devonian and Mississippian for trees to adapt to such infertile soils remains to be seen from the continuing search for fossil root traces and paleosols.

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$$C = -0.5/\{[0.49/e^{(D/3.7)}]-1\}$$

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Fabrication of Atomic-Scale Structures on Si(001) Surfaces

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The scanning tunneling microscope has been used to define regular crystalline structures at room temperature by removing atoms from the silicon (001) surface. A single atomic layer can be removed to define features one atom deep and create trenches with ordered floors. Segments of individual dimer rows can be removed to create structures with atomically straight edges and with lateral features as small as one dimer wide. Conditions under which such removal is possible are defined, and a mechanism is proposed.

The ability of the scanning tunneling microscope (STM) to fabricate ordered atomic-scale structures of any desired geometry (*1–3*) and to probe their properties opens new frontiers for investigation, including studies of spatially resolved electron interference in model systems (*4*) and current modulation mechanisms for electronic devices with atomic-scale active areas (*5*). The four main categories of fabrication with the STM are (i) arranging adatoms into a pattern (*1, 4*), (ii) writing patterns by depositing atoms onto the surface from the tip (*6, 7*) or by decomposing precursor molecules (*8, 9*), (iii) using the STM to create an etch mask (*10, 11*), and (iv) defining patterns by transferring unwanted atoms from the surface to the tip (*2, 3*). For the investigation of nanoelectronic devices and model systems, an important aspect of any of these fabrication methods is the ability to produce atomically ordered structures of any desired geometry. Removing material from an already well-ordered surface is the most straightforward method for creating crystalline atomic-scale structures with well-defined and regular features that are stable at ordinary temperatures.

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