1967

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Abstract. Many soils have clay-enriched horizons that are thought to result from pedogenic processes. Various hypotheses are proposed for expected clay size-distributions in soil profiles. The average size of clay crystallites determined by the amount of broadening of x-ray diffraction peaks ranged from 160 to $>2000A°$, but the lack of trends in four loess-derived soils did not permit the hypotheses to be evaluated. An unexpected relationship in a well expressed illuvial horizon was noted: as the percent of accumulated clay increased to the observed maximum the average crystallite size also increased rather than decreased as proposed. This suggests that small-sized illuvial clay particles undergo reorganization to form larger stable crystallites. Knowledge of clay crystallite size is expected to supplement other data in interpreting soil genesis.

Soil genesis is the study of the development of horizons in a soil. It involves, in part, estimating the original properties of the unconsolidated material and attempting to reconstruct the possible conditions and events which have given rise to the present-day observable features.

The ideal situation exists where initially a soil material is isotropic in all measurable properties and the soil horizons result from a rearrangement or removal of various components, such as carbonate and clay. Concepts to explain changes have classically assumed the initial materials to be homogenous, thereby focusing attention on the kinds of processes that would differentiate the parent material into recognizable soil horizons.

Loess approaches the concept of an ideal soil parent material in that it (a) has a mixed mineralogy capable of responding to a weathering environment, (b) has a medium texture which slows up percolating water but does not unduly restrict the removal of weathering end products, (c) has been exposed long enough on stable landscapes to reflect the influence of the climate and vegetation characteristic of a region, and (d) has less variability of other physical and chemical attributes than many materials in which soils have developed.

Starting with such a material, a disparity of clay content between upper and lower horizons in a profile is commonly thought to result from a translocation of the inherited clay from the upper zone to the underlying zone. Processes of clay formation and clay destruction may also account for some of the observed differences in clay content throughout a profile. The weight of several

size fractions or of total clay is usually measured, however, very little is known about the size of clay packets or crystallites as they occur in soils. The technique of estimating average crystallite size based on x-ray peak broadening (Rosauer and Handy, 1961) can be applied to soil clay to provide additional information on horizon development.

HYPOTHESIS CONCERNING CLAY SIZE-DISTRIBUTION

Phyllosilicates are reported to range widely in size (Grim, 1953; van Olphen, 1963; Marshall, 1964). For example, kaolinite may have single plates 40A° thick and 400-900A° wide but more commonly they are about 500 A° thick and 3000 A° or more wide; some mica (illite) particles are 30 A° thick and 1000-3000 A° wide; and some montmorillinite particles are 20-80 A° thick and 200-300 A° wide. A general relationship is that montmorillinite crystallites are smaller than micas (illite) and chlorites which in turn are smaller than kaolinite crystallites.

Previous investigations (Hutton, 1951; Hanway *et al.,* 1960; Glen *et al.,* 1960; Frye *et al.,* 1962) indicate that the inherited clay mineralogy of loess in and adjacent to Iowa consists of montmorillinite, chlorite, hydrous mica (illite), interlayer variations of the above, and small amounts of kaolin with montmorillinite usually comprising 60-80% of the mixture.

In their study of the clay mineralogy of a Tama silt loam from southwestern Wisconsin, Glen *et al* (1960) fractionated the -2μ material into 3 classes: coarse clay, 2-0.2μ; medium clay, 0.2-0.08μ; and fine clay, -0.08μ. The 2:1 layer lattice minerals comprised between 65 and 80% of the clay-sized material with the remainder including amorphous material, kaolinite, and primary minerals of clay size. The fine clay fraction contained 60-70% montmorillinite; the medium clay contained 40-50% montmorillinite and 15-30% mica, and the coarse clay contained 10-20% montmorillinite and 30-40% mica.

It is assumed that the original loess deposits contained a mixed clay mineralogy dominated by 2:1 layer lattice minerals and that a broad distribution of sizes of these clays also existed.

Changes of a uniform distribution of clay content in an initial homogenous material may arise by (a) translocation of the clay from one zone to another, (b) formation of new clay-sized material, (c) destruction of inherited or formed clay, or (d) some combination of formation, destruction, and translocation (Arnold, 1965).

Clay is commonly reported as a weight percent, thus if equal weights of different size fractions are analyzed one would measure many more fine clay particles than medium or coarse clay

particles, consequently the average size would tend to be weighted toward the smaller crytallites. An average crystallite-size of the dominant clay present in these soil horizons may be estimated by measuring the boadening of the 060 peak of the 2:1 lattice clays.

Several postulates concerning the expected average size of clay particles may be made for the above kinds of transformations. If destruction is taken to mean the chemical dissolution and break-down of clay it would be expected that the small size particles with large surface areas per unit volume would be destroyed first, thus the average size of the remaining material would increase. Formation of additional clay may result from the physical break-down of silt or silt-sized aggregates of clay deposited in the original material, in which case the initial breakdown would yield relatively large-size clay particles; thus the average size of crystallites would increase. Formation may also result by the synthesis of amorphous gels which crystallize into detectable particles. The initial development of crystallinity is thought to give rise to small-size particles (Hénin and Pedro, 1965), thus the average size of the bulk would decrease. Dispersion and movement in suspension of small-size clays, whether inherited or formed, would be expected to occur before movement of large particles, thus in the zone of depletion the average crystallite size should increase, whereas in the zone of accumulation the average size should decrease.

Assuming a homogeneous parent material with respect to kinds, amounts, sizes and distribution of clay minerals increasing values of the average crystallite size may be due to (1) physical removal of smaller-size particles, (2) chemical destruction of the smaller-size particles, or (3) increased breakdown of silt or silt-size aggregates of clay. Decreasing values of the average crystallite size may be due to (1) the formation of new crystalline material or (2) an accumulation of translocated clays.

MATERIALS AND METHODS

Four loess-derived soils, differing in their expression of a clay enriched subsoil, were selected for examination. A profile of Monona silt loam from Crawford County, Iowa, has an ill-defined textural horizon extending from 8 to 32 inches; two profiles of Tama silt loam from Grundy County have weakly expressed textural horizons between 8 and 32 inches; and a profile of Tama silt loam, subsoil variant from Cedar County has a well expressed textural horizon between 32 and 42 inches. Monoliths obtained at each site were kept moist to reduce any effect of drying on clay aggregation.

A small matrix sample of soil taken from each 2-inch depth

increment of a profile was placed in a glass vial and shaken with distilled water. As soon as particles coarser than about 30μ had settled to the bottom, several ml of the remaining suspension were removed with a pipette. Aliquots of the fine silt and clay suspension were placed on an unglazed ceramic tile, to which a slight suction was applied to withdraw the excess water, until a relatively thick layer of clay was deposited. The prepared tile was then air dried. The procedure was adopted to reduce any dis-aggregating affects of vigorous physical or chemical dispersion, and to obtain a sample representative of the soil clays in their natural state. The inclusion of some fine silt insured that enough quartz would be present to provide a reference peak in the x-ray dif-fraction pattern.

In the three Tama profiles additional samples were taken from the exterior portion of subsoil structural units for comparison with the matrix samples.

The air-dried samples were examined with a General Electric XRD-5 x-ray diffraction unit. Diffractions were obtained using Ni filtered $CuK\alpha$ radiation produced at 50 kv and 16 ma, and counting was done with a scintillation tube. Other operating conditions included: a 3° beam slit, medium resolution Soller silt, 0.2° detector slit, 3 second time constant, reverter was in, linear range of 2000cps, scanning speed of 2°/min, and a chart speed of 1 in/min.

In large crystals there are numerous parallel atomic planes diffracting an incident x-ray beam at a very precise angle which gives rise to a sharply defined diffraction peak. In small crystals the number of atomic planes is reduced, small deviations of the ideal angle do not cancel the diffraction of the x-ray beam, and the diffraction no longer produces a sharp maximum peak.

The width of a diffraction peak is measured at half-maximum peak height, graphically corrected for broadening due to the $K\alpha$ radiation and the instrument geometry, and the mean dimension, D, normal to the diffracting planes of the crystal is calculated using the Scherrer equation.

This equation,

$$D = \frac{K\lambda}{\beta \cos \theta} \, ,$$

expresses the average crystallite size, D, as an inverse function of the corrected diffraction broadening, β, where K is a constant equal to unity, λ is the x-ray wavelength, and θ is the angle between an atomic plane in the crystal and the incident x-ray beam at which a diffraction maximum will occur.

Line broadening of the 060 diffraction peak for the 2:1 layer lattice clays occurs at or near 62° 2θ corresponding to a "d" spac-

ing of approximately 1.50 A° (Figure 1). The calculation proced-
ure was that of Rosauer and Handy (1960) except that in each
case the correction for instrument broadening of the diffraction
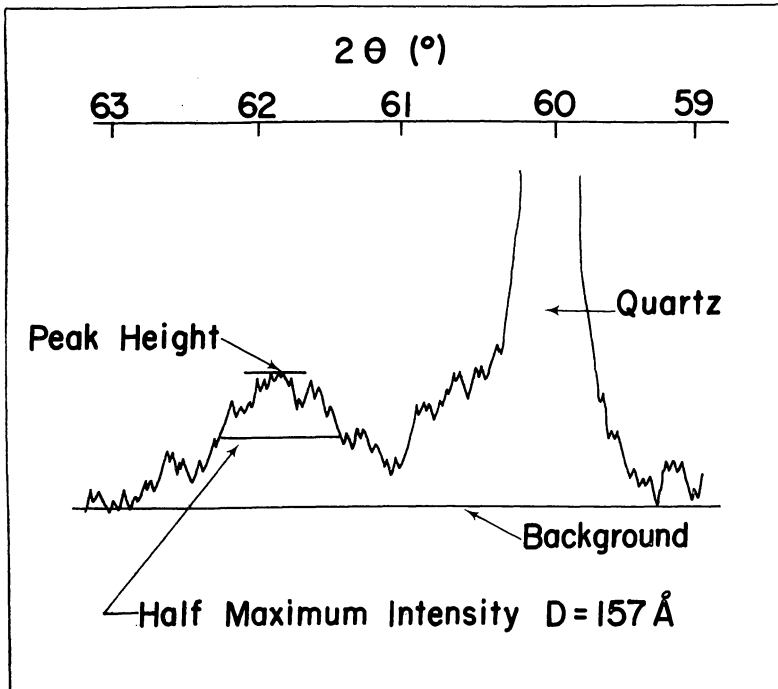peak was obtained from a reference quartz peak at 60° 2θ.

Figure 1. Broadening of 060 diffraction peak of 2:1 phyllosilicates in loess-derived soil.

RESULTS AND DISCUSSION

Most of the calculated crystallite sizes (Table 1) are from
single determinations. Very little variation was noted if a sample
was scanned twice in succession or if the low, broad peaks were
remeasured at a later date, however, if the tile was shifted slight-
ly or the sample was rerun a week later, larger variations were
observed. The error of estimating size increases as the size of the
crystallite increases (Rau, 1965) and based on seven sets of trip-
licate determinations the average coefficient of variation was
20.3%.

The crystallite b-axis dimensions of these clays are commonly
between 200 and 350 A°, and assuming an average unit cell to be
about 9 A° wide these particles have 20 to 40 unit cells connected
edge-to-edge. Several measurements of the c-axis of the mont-

Table 1. Average crystallite size of 2:1 layer lattice clays in four loess-derived soils.

Depth (in)	Monona Matrix	Tama I		Tama II		Tama-Var.	
		Matrix	Exter.	Matrix	Exter.	Matrix	Exter.
1	271[a]	201	...[b]	650	...	...	...
3	207	226	...	218	...	203	...
5	198	...	...	291	...	194	...
7	199	...	...	209	...	203	...
9	520	217	...	358	...	206	...
11	408	459	...	297	...	276	...
13	373	472	496	194	218	201	...
15	297	260	936	291	242	187	...
17	203	>2000	501	271	631	226	...
19	258	221	166	172	269	203	...
21	271	255	643	410	286	225	...
23	303	431	259	326	207	270	304
25	254	180	196	302	189	194	252
27	245	537	182	343	1704	218	610
29	250	230	...	>2000	>2000	263	230
31	286	157	726	218	1008	221	520
33	217	219	...	326	236	276	376
35	203	147	...	247	206	244	244
37	239	266	245	286	338	524	267
39	257	>2000	175	271	225	225	185
41	289	263	...	284	520	178	271
43	230	...	...	263	466	242	285
45	217	283	333	271	203	199	...
47	239	938	938	171	173	327	...
49	178	...	...	...	...	...	...

[a] Size reported in angstroms (A°) for the b axis.
[b] Dash indicates that the size was not determined for that depth.

morillinite component indicated that about 5 unit cells are stacked face-to-face, thus the crystallites are assumed to be tabular.

Profile distributions of crystallite sizes reveal wide fluctuations over small vertical distances, consequently no overall trends of increasing or decreasing sizes are noted. In all four profiles, one or more samples from depths of 8 to 14 inches have larger sizes than adjacent samples which may reflect increased rates of weathering or translocation in the relatively undisturbed soil immediately below the plow layer.

The clays on the exterior of subsoil peds are commonly larger than those in the matrix in the Tama II and the Tama variant profiles but the reverse was noted for the Tama I profile.

It is apparent that no general interpretations can be made regarding the proposed hypotheses for expected crystallite size in soil profiles without additional information. Calculations of theoretical clay changes for the Tama II and Tama variant profiles (Arnold, 1963) provide values for the amount of clay formed from non-clay components, and the amount of clay lost or gained

by translocation. No consistent trend was found when crystallite size was compared with the ratio formed: illuvial clay or the ratio formed: eluvial clay, however, size does tend to increase with increasing amounts of clay lost from a zone irrespective of the amount of clay formed *in situ.*

The Tama variant is the only profile with a definite zone of higher clay content. In the horizon from 29 to 43 inches the approximate percent of present clay that is illuvial is 0, 20, 30, 30, 30, 15, 6 and 0 respectively for each 2-inch increment starting at the 29-inch depth (Arnold, 1963). A curvilinear relationship exists between crystallite size and % of present clay accumulated by translocation if the average size for the three samples with 30% illuvial clay is used. The average size of matrix crystallites would decrease as small size particles are moved into the zone. In this soil the minimum size (ca 180 A°) corresponds to an accumulation of 5 to 10% of the clay. With increasing amounts of clay moving into the horizon, the average size tends to increase (to ca 350 A°) rather than to decrease further. The small crystallites may overlap and build up packets which are 50% or more wider than the initial average size (240-250 A°). Pawluk (1963) has reported that fine clay-size montmorillinite may be bound into larger particles by hydrated alumina hydroxy ions released from feldspars.

Soil clay minerals have received and are receiving much attention in research efforts yet little is known about actual sizes and distributions of naturally occurring crystallites. I feel that knowledge of the size of these particles would greatly supplement interpretations of the weathering, formation, and movement of clay minerals in the genesis of soils.

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