SPECIFICITY OF THE HALLOYSITE-RICH TROPICAL OR SUBTROPICAL SOILS

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INTRODUCTION

Some clayey soils of tropical or subtropical countries, deriving from volcanic pyroclastic rocks, are mostly made of halloysite and iron oxy-hydroxides. The specific properties of the halloysite-rich soils are not taken enough into account in soil taxonomy.

In the French Soil Classification (CPCS 1967), the halloysite-rich soils are related to either Brown Tropical or Ferralitic or even Fersialitic Soils. These Soils, which are still rich in calco-alkaline elements, are transitional between Brown and Ferralitic Soils. Segalen & al. (1979) proposed to distinguish a halloysitic Group amongst the Fermonosialsols.

The Soil Taxonomy (1975) takes into account the specific propertyg of halloysite clays only at the Family level. Due to their high C.E.C. (> 16 me/100g), the halloysite-rich soils cannot be Oxisols. Often estimated as Inceptisols, they are related to Tropepts or Ochrepts; but if they have an argilic horizon they are related to Alfisols or to Ultisols.

Herbillon & al. (1989) emphasized the intermediate property of halloysite, between the high and the low activity clays. The aim of this paper is : first, to show the distribution of

The aim of this paper is : first, to show the distribution of halloysite-rich soils and to present a scheme of their genesis; secondly, to recall the specific properties of these soils as well as the mineralogical features of halloysite clays which explain their specifity and which justify a peculiar place in soil taxonomy.

OCCURENCE AND DISTRIBUTION

The halloysite-rich soils are frequent on Pleistocene volcanoes, in wet tropical or subtropical countries, except on Holocene volcanic ashes where the soils are mostly rich in allophane. Almost all these soils derive from pyroclasts, rich in glass, without quartz or just a little.

In addition to halloysite (10-7Å), goethite and haematite, these soils eventually contain little allophane, kaolinite, 2:1 clays, or gibbsite. The youngest of them keep some weatherable minerals.

These soils have been mostly discovered for the last 30 years in most of volcanic tropical and subtropical countries. They are also found on the border of temperate lands, until 42°S (Chile, New Zealand) or 42 to 45°N (Japan, Italy, France). They are known in the following countries. In Africa : Cameroons, Ethiopia, Tanzania, Rwanda, Kiwu, Kenya, Spanish Guinea, Canary Islands. In Latino-America : Argentina, Chile, Brazil, Ecuador, Galapagos I., Colombia, Costa Rica, Nicaragua, Guatemala, Mexico, Antilles. In the Indian Ocean : Madagascar, Mauritius, La Reunion, Comores. In the Pacific Ocean : Hawaii, Tahiti, Fidji, New Hebrides, New Zealand, Salomon, New Guinea, Indonesia, Philippines, Japan. In Europe : Italy, France, Spain, Portugal, and Georgia (SSSR).

The halloysite-rich soils are located in a peculiar part of climotoposequences or of chronosequences, on the volcanic land forms.

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N° : 3/ 994 event Cpte : B 22 AVR. 1991 PNF Thus their formation is climate and time dependent. 1. Climotoposequences : In the tropics, the well known sequences are on volcanic islands as the Antilles (Colmet-Daage & al. 1965), La Reunion (Zebrowski 1975), the New Hebrides (Quantin 1972-78), and the New Guinea (Chartres & Pain 1984).

These studies show that halloysite-rich soils have been mostly formed under humid tropical climate (T 25-23°C, P 2000 - 3500 mm), with a less rainy season of 4-6 months or a dry season of 1-2 months. However some of them are found under less humid tropical or subtropical climate (P 1500 - 1000 mm, dry season of 4-5 months), for example in Cameroons (Sieffermann 1973), on the Canary Islands (Fernandez Caldas & al. 1982), or on the slopes of Andes (Faivre 1989). In all these cases, there has been a recent climate change.

2. Chronosequences : In the wet tropics the halloysite-rich soils are found on volcanic ash, at least 5000 years old or even 10,000 20,000 years old as for the most clayey soils. But their life time is limited; the more advanced soils of early Pleistocene or of the Tertiary are mostly made of kaolinite (Sieffermann 1973).

GENESIS OF HALLOYSITE

All the authors agree to say halloysite genesis is favoured by silicarich solutions, as well as under a tropical climate (Sieffermann 1973, Chartres & Pain 1984, Quantin & Rambaud 1987), as under a temperate one (Aomine & Mizota 1973, Wada 1980, Wada & al. 1985, Parfitt & Wilson 1985). The soils where this mineral grows are slightly acid, rich in calco-alkaline cations and also often in iron and manganesis (Wada 1987, Quantin & Rambaud 1987).

The richness in silica and cations of the solutions is due to two causes. First, the superimposition of pyroclastic materials : the youngest one on top, owing to the strong weathering of glass, grows allophane and at the same time it transfers to the subsoil solutions which are rich in silica, cations, as well as probably some alumina (Wada 1980). Secondly, a lowering of drainage at the bottom which favours the growth of clay, especially halloysite (Sieffermann 1973, Parfitt & Wilson 1985).

The halloysite-rich soils generally appear after allophane-rich soils. These latter often cover halloysitic soils, according to a chronostratigraphic sequence, as well as under tropical as under temperate climate. Thus we can infer the formation of halloysite by silicification of allophane. But in some cases halloysite grows directly from glass (Quantin & al. 1988), or in some older soils it appears from the surface. In fact halloysite can happen at the same time as allophane or later by dissolution of allophane. Then halloysite progressively becomes predominant. For instance we observed (Quantin & al. 1987) the progressive growth from allophane to spherical halloysite through a sequence of basaltic ash deposits from 500 to 1500 years.

Halloysite appears rapidly under a wet tropical climate : 1000-1500 years in the New Hebrides (Quantin & al. 1987); 300-2000 BP in New Guinea (Bleeker & Parfitt 1974); 4000 BP in Saint Vincent (Hay 1960). Its formation seems slower in temperate countries : 8000-10,000 BP in Japan (Aomine & Miyauchi 1963, Saigusa & al. 1978); 14;000 - 20, 000 BP in New Zealand (Kirkman 1975); although MacIntosh (1980) observed there its formation as soon as early stages of weathering.

About 10,000 years is the rate of full weathering of 1m deep basaltic ash into halloysite, have we estimated in the New Hebrides; meanwhile it can be 3 to 5 times more for a lava flow. Thus the halloysite-rich tropical soils are more than 10,000 years old if they derive from ashes, or at least 50,000 years if from lavas. But the halloysite is instable; it is slowly weathered to form kaolinite and some gibbsite, the rate of which increased to the top of soils which date from early Pleistocene or Pliocene (Sieffermann 1973, Churchman & Gilkes 1989). Thus the halloysite-rich soils within top soil are always younger than 1 MY.

SPECIFIC PROPERTIES

The halloysite-rich tropical soils like the kaolinitic ferralitic soils are made of 1:1 clay mineral and of iron oxyhydroxides, and eventually of gibbsite. The molar silica/alumina and silica/sesquioxides ratios range respectively from 2.2 to 1.3 and from 1.5 to 0.9. However the properties of the former are peculiar.

Firstly the halloysite-rich soils derive from pyroclastic rocks. Then in comparison with kaolinitic soils from the same material, they are younger and of lesser depth; they keep a large reserve in weatherable minerals or at least in calco-alkaline cations.

Then these soils have some peculiar physical and chemical properties, due to the surface properties of halloysite, which are more developed than those of kaolinite. Their original assemblage of clay particles allows a large microporosity. In addition, these properties change irreversibly after a strong dehydration. In the wet tropics they have the following specific properties.

1. Physical properties : A polyhedral nutty macrostructure, broader and less friable than for kaolinitic soils; shiny ped surfaces due to stress-cutans; a light bulk density $\sim 0.7 - 1.1 \text{g/cm}^3$ due to a large microporosity; an external specific area $\sim 100-200 \text{m}^2/\text{g}$ (while for kaolinite $\sim 15-40 \text{m}^2/\text{g}$); a large field water capacity (1 bar) $\sim 50-80\%$ (of dry soil at 105°C); a rate of irreversible dehydration $\sim 30-40\%$ (of initial field water capacity); an irreversible retraction due to a loss of interlaminar water.

2. Chemical properties: A C.E.C. (pH7) v 15-40 meq/100g of dry soil, equivalent to 30-60 meq/100g of clay (while for kaolinite v 5-15 meq/100g); a rate of pH dependent (9-4) C.E.C. 40-60% of maximum value; a permanent C.E.C. v 16-36 meq/100g; a specific absorption of K⁺ and NH4⁺ (Delvaux 1988); a P- retention capacity (Blakemore method) v 70-80%.

MINERALOGICAL INTERPRETATION

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The theoretical formula 2SiO₂, Al₂O₃, 4H₂O and X Ray Diffraction at 1 nm distinguish halloysite from kaolinite. But this state is metastable. By dehydration there is a transition from hydrated halloysite at 1 nm to dehydrated halloysite at 0,72 nm. That is almost irreversible. However, even dehydrated, halloysite can alsorb an interlaminar layer of bipolar organic liquid. For instance the intercalation of formamide (Churchman & al. 1984) re-establishes the 1 nm reticular space and allows to distinguish 0.7 nm halloysite from kaolinite.

The second criteria of halloysite is the curvature of the clay sheets, mostly in tubes or in spheres.

Numerous studies on the so-called halloysite clays show a wide variation of chemical composition, shape and properties (Kunze & Bradley 1964, Sieffermann 1973, Weaver & Pollard 1973, Tazaki 1982, Wada & Kakuto 1985, etc). There are various shapes, such as sphere, tube or crumpled lamella (Tazaki 1982). Tabular forms are rare. There is also a microspherical one, near allophane, which Wada and Kakuto (1985) named "Embryonic". In the tropical soils we observe the following evolution of clay forms: allophane \rightarrow micro-spherical or crumpled lamellar halloysite \rightarrow spherical H. \rightarrow Tubular H. \rightarrow kaolinite. The tubular form predominates in the more advanced soils. Tazaki (1982) showed a correlation between morphology and chemical composition. Along a chronosequence the first forms, in crumpled lamella or in sphere, are richer in iron and magnesium than the last one, in tube (Wada & Mizota 1982, Tazaki 1982, Wada & Kakuto 1985, Quantin & al. 1984, 1987, 1988, Delvaux 1988). Thus there is an evolution of form and composition to a mineral closer to kaolinite.

The third important criterion of halloysite is the cation exchange capacity (C.E.C.). Almost all halloysite clays of tropical soils are high charge clays. The C.E.C. of 1 nm halloysite ranges from 30-60meq/ 100g; that of halloysite 0.7 nm \sim 15-30meq/100g. A great part of this C.E.C. is due to permanent charges (Quantin 1984, Delvaux 1988). These high charges clay are always rich in iron and sometimes in magnesium. They differ from low charge halloysites (Churchman & Theng 1984), the properties of which are close to kaolinite. A good correlation has been established between the C.E.C. values and the internal area, the Fe and Mg contents, the effect of dehydration, as well as the selectivity of K⁺ adsorption before and after dehydration (Delvaux 1988, Delvaux & al. 1990).

These properties, in connection with permanent charges, cannot be explained by a structure of 1:1 clay with a substitution of Al by Fe and some Mg elements in the octahedral layer. It is necessary to do appear tetrahedral charges by substitution of Si by Al. In fact there are series of coherent properties which show that the high charge halloysite clays are probably an interstratification of 1:1 and 2:1 clay minerals, with smectite or vermiculite component in the ratio of 30 to 15% of whole clay (Quantin & al. 1984, 1988; Delvaux 1988, Delvaux & al. 1990).

This interpretation allowed some authors to propose some structural formulas of the 2:1 clays accompanying the halloysite (Quantin & al. 1984, 1988; Delvaux 1988). These formulas show high values of tetrahedral charges and they fit in well with almost all the observed properties. In addition Delvaux & al. (1990) showed a genetic sequence of these halloysite-smectite mixed clays, from those with high charge smectite to those with lower charge smectite, which makes a transition to low charge halloysite or to kaolinite.

CONCLUSIONS

Most of halloysite-rich soils, which derive from pyroclasts, could contain high charge halloysitic clays. These clays are probably made of an interstratification of halloysite-smectite or of halloysite-vermiculite minerals, of which the 2:1 clay component has high tetrahedral charges and the 1:1 clay is predominating.

This mixed clay mineral makes a transition between allophane and kaolinite. Thus it has a genetic meaning.

In addition, the abundance of this kind of clay has an important impact on the morphology, the structure, the chemical and physical properties of soils. Therefore the land use potentialities of these soils are much higher than those of kaolinitic soils.

Indeed in soil classification, the soils which are rich in high charge halloysite and iron oxy-hydroxide should never be related to Ferralitic soils, but rather to Fersialitic ones, or at least to be considered as intergrade. In the "Soil Taxonomy", their position should be recognised at a higher level than the Family, as it has be done for the Andisols.

REFERENCES

Aomine, S.; Miyauchy, N. Age of the youngest hydrated halloysite in Kyushu. Nature 199 : 1311-1312; 1963. Aomine, S.; Mizota, C. Distribution and genesis of imogolite in volcanic ash soils of Northern Kanto, Japan. Proc. Int. Clay Conf. Madrid 1972: 207-213; 1973. Bleeker, P.; Parfitt, R.L. Volcanic ash and its clay mineralogy at Cape Haskins, New Britain, Papua-New Guinea. Geoderma 11 : 123-135; 1974. Chartres, C.J.; Pain, C.F. A climosequence of soils on late Quaternary volcanic ash in highland Papua-New Guinea. Geoderma 32 : 131-156; 1984. Churchman, G.J.; Gilkes, R.J. Recognition of intermediates in the possible transformation of halloysite to kaolinite in weathering profiles. Clay Miner. 24 : 579-590; 1989. Churchman, G.J.; Whitton, J.S. & al. Intercalation method using formamide for differenciating halloysite from kaolinite. Clays and Clay Miner. 32 : 241-248; 1984. Colmet-Daage, F.; Lagache, P. Caractéristiques de quelques groupes de sols dérivés de roches volcaniques aux Antilles Françaises. Cah. ORSTOM Pedol. 3 : 91-121; 1965. C.P.C.S. Classification des sols. Note ENSA Grignon, France; 1967. Delvaux, B. Constituants et propriétés de surface des sols dérivés de pyroclastes basaltiques du Cameroun Occidental. Thesis; Catholic University of Louvain, Belgium; 1988. Delvaux, B.; Herbillon, A.J.; Vielvoye, L. & al. Surface properties and clay mineralogy of hydrated halloysitic clays. II Evidence for the presence of halloysite-smectite mixed-layer clays. Clay Miner.; (in press, 1990). Faivre, P. Lessivage et planosolisation dans les séquences de sols caractéristiques des milieux intrandins de Colombie. Thesis; University of Nancy I, France; 1988. Fernandez Caldas, E.; Tejedor, M.L.; Quantin, P. Suelos de regiones volcanicas, Tenerife, I. Canarias. Col. Viera y clavijo, 4; Universidad de la Laguna, Spain; 1982. Hay, R.L. Rate of clay formation and mineral alteration in a 4000 years old volcanic ash soil on St. Vincent, B.W.I. Am. J. Sci. 258 : 354-368; 1960. Herbillon, A.J.; Delvaux, B. & al. Halloysites from tropical ash derived soils at the border between high activity and low activity clays. Proc. Int. Soil Clas. Conf. Alma Ata; 1988; (in press, 1989). Kirkman, J.H. Clay mineralogy of some tephra beds of Rotorua area, North Island, New Zealand. Clay Miner. 10 : 437-449; 1975. Kunze, G.W.; Bradley, W.E. Occurence of a tabular halloysite in a Texas soil. Clays Clay Miner. 12 : 523-527; 1964. MacIntosh, P.D. Weathering products in Vitrandept profiles under pine and Manuka, New Zealand. Geoderma 24 : 225-239; 1980. Parfitt, R.L.; Wilson, A.D. Estimation of allophane and halloysite in three sequences of volcanic ash soils, New Zealand. Catena Supp. 7 : 1-8; 1985. Quantin, P. Atlas des Nouvelles Hébrides. ORSTOM, Paris; 1972-78. Quantin, P.; Gautheyrou, J.; Lorenzoni, P. Halloysite formation through in situ weathering of volcanic glass from trachytic pumices, Vico's Volcano, Italy. Clay Miner. 23 : 423-437; 1988. Quantin, P.; Herbillon, A.J. & al. L'"halloysite" blanche riche en ___ fer de Vaté (Vanuatu). Hypothèse d'un édifice interstratifié halloysitehisingérite. Clay Miner. 19 : 629-643; 1984. Quantin, P.; Rambaud, P. Genesis of spherical halloysite from basaltic ash, at Ambrym (Vanuatu). Proc. Int. Meet. Geochem. Earth Surface, Granada 1986 : 505-522; 1987. Saigusa, M.; Shoji, S.; Kato, T. Origin and nature of halloysite in Andosoils from Towada tephra, Japan. Geoderma 20 : 115-129; 1978. Segalen, P. & al. Projet de classification des sols. ORSTOM Paris; 1979. Sieffermann, G. Les sols de quelques régions volcaniques du Cameroun. Mémoire ORSTOM N°66, Paris; 1973. Soil Conserv. Serv. Soil Taxonomy, Agric. Handbook N°436. US. Dep. Agric. Washington; 1975. K. Analytical electron microscopic studies of halloysite Tazaki, formation processes. Morphology and composition of halloysite. Proc. Int. Clay Conf. Bologna 1981 : 573-584; 1982. Wada, K. Mineralogical characteristics of Andisols. in Theng, B.K., ed. New Zeal. Soc. Soil. Sci. : 87-107; 1980. Wada, K. Minerals formed and mineral formation from volcanic ash by weathering. Chemical Geology 60 : 17-28; 1987. Wada, K.; Kakuto, Y. Embryonic halloysites in Ecuadorian soils derived from volcanic ash. Soil Sci. Soc. Am. J. 49 : 1309-1318; 1985. Wada, K.; Mizota, C. Iron rich halloysite with crumpled lamellar morphology from Hokkaido, Japan. Clays Clay Miner. 30 : 315-317; 1982. Weaver, C.E.; Pollard, L.D. The Chemistry of Clay Minerals. Develop. Sedimentology 15, Elsevier; 1973. Zebrowski, C. Etude d'une climato-séquence dans l'Ile de la Réunion. Cah. ORSTOM Pedo1. 13 : 255-278.

SUMMARY

Halloysite-rich soils mostly appeared on pyroclastic rocks in tropical or subtropical countries. They form under a fairly wet climate, in a particular stage of climotoposequences and chronosequences. Their formation from volcanic ash needs at least 5000 years. The halloysite genesis is due to silica and cations rich solutions or a slow drainage. Halloysite-rich soils differ from kaolinitic ones by specific properties such as a high water retention and a high C.E.C. These properties are due to high charge halloysite clays. These ones are probably mixed layer clays, made of predominant halloysite and of high charge smectite or vermiculite. The peculiar properties of which are important for soil classification.