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Geochemistry and mineralogy of saprolite in Finnish Lapland

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Abstract

An ancient saprolite has developed on the Palaeoproterozoic granulite, granite gneiss and amphibolite bedrock of the Vuotso–Tankavaara area of central Finnish Lapland. The present day climatic regime in Finnish Lapland lies within the northern boreal zone and so the saprolite there can be regarded as fossil. Cores of saprolite were collected from 4 sections (42 samples) and analyzed chemically and mineralogically. In the study area, progressive weathering of the rocks has been marked by gradual enrichment in Al, Fe and Ti; and depletion of Na, K and Ca. The higher concentration of Fe(III) and water and reduced Na and Ca in weathered bedrock in the 4 sections are indicative of oxidation, hydration and leaching processes involved during weathering. The primary minerals in the saprolite are plagioclase feldspar, K-feldspar, quartz, garnet (almandine) and hornblende; the common secondary minerals are kaolinite, halloysite, and vermiculite in addition to minor amounts of sericite. Intense weathering is indicated by: (1) the presence of kaolinite and halloysite in 4 sections of different bedrock types, and (2) the comparatively lower SiO₂/Al₂O₃ (wt.%) ratio (2.30) of weathered granulites (3 sections) as compared to fresh granulite (4.33) and that of weathered amphibolite (2.68) as compared to fresh amphibolite (3.56). In general, kaolinite and halloysite have formed through the weathering of feldspars, garnet, and biotite. Vermiculite is the most probable alteration product of biotite. The formation of kaolinite and halloysite in Finnish Lapland indicates wetter and warmer climatic conditions during the time of their formation than at present. The possible time for formation of the saprolite is early Cretaceous–early Tertiary into Middle Miocene. © 2002 Elsevier Science Ltd. All rights reserved.

1. Introduction

Chemical weathering of rocks is one of the major processes that modify the earth's surface and is one of the vital processes in the geochemical cycling of elements (Berg, 1932). The rate and nature of chemical weathering vary widely and are controlled by many variables such as parent rock type, topography, climate and biological activity. The mobilization and redistribution of trace elements during weathering is particularly complicated because these elements are affected

by various processes such as dissolution of primary minerals, formation of secondary phases, redox processes, transport of materials, coprecipitation and ion exchange of various minerals (Harris and Adams, 1966; Nesbitt, 1979; Chesworth et al., 1981; Fritz and Ragland, 1980; Nesbitt et al., 1980; Cramer and Nesbitt, 1983; Fritz and Mohr, 1984). Nevertheless some general statements can be made (Middelburg et al., 1988).

Palaeoweathering remnants have been recorded in many regions that were severely glaciated during the Pleistocene epoch. In most cases these regions belong to shields or Paleozoic massifs where ice sheets and icecaps have covered low or moderate pre-glacial relief (Battiau-Queney, 1981, 1984; Bouchard and Pavich, 1989; Fogelberg, 1985; Godard, 1989; Hall, 1986; Lidmar-Bergström, 1988; Lundqvist, 1985; Peulvast, 1985; Roaldset et al., 1982). Research on Scandinavian saprolites (e.g. Whalley et al., 1997; Lidmar-Bergström,

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1988; Fogelberg 1985; Godard 1989) has highlighted the general issue of pre-Quaternary weathering in the region.

Alteration of feldspars to halloysite, kaolinite, and gibbsite in a deep weathering profile in Penninsular Malaysia has been reported by Eswaran and Bin (1978). Nesbitt et al. (1980) studied the continental weathering of Toorongu Granodiorite in Victoria, Australia and have reported the degradation of primary minerals and release of alkalis and alkaline earths from the weathering profiles. Allen et al. (2001) have carried out a weathering study in Arctic–Alpine Sweden (close to Finnish Lapland) and have found higher abundance of clay minerals which indicated either a long period of in situ chemical weathering or a greater period of development. Hill et al. (2000) have studied the geochemical evolution of paleolaterite in Northern Ireland where they have reported neo formed mineral assemblages consisting of hematite, gibbsite, goethite, anatase, meta-halloysite and kaolinite.

Systematic Quaternary geological and geochemical surveys (Kujansuu, 1967; Hirvas, 1992) have shown that almost the whole area of the Pleistocene ice divide of central Lapland is underlain by ancient saprolite. In Finnish Lapland, saprolite was recognized in 1836 (Fircks, 1907). Though the existence of ancient saprolite has been long known, no systematic weathering study has been done in this region. The present day climatic regime in Finnish Lapland lies within the northern boreal zone where saprolite formation does not occur. Therefore the saprolite can be regarded as fossil. The aim of this study is to decipher the elemental dispersion, weathering processes, mineralogical changes, and paleoenvironment of saprolite in Finnish Lapland.

2. Geology of the study area

The study took place in the Vuotso–Tankavaara area, which is located between 68°5′N and 68°12′N latitude, 27°05′E and 27°10′E longitude and is situated in central Finnish Lapland about 230 km north of the town of Rovaniemi and the Arctic Circle (Fig. 1). The present climatic regime in Finnish Lapland is Northern Boreal Zone (Tuhkanen, 1984) with annual mean temperature $-0.66\text{ }^{\circ}\text{C}$, relative humidity 78% and annual precipitation 485 mm (Atlas of Finland, 1987).

The Early Proterozoic Lapland Granulite Belt is a major geological feature of the northern Fennoscandian Shield forming a 50–90 km wide arc-shaped belt extending from the Scandinavian Caledonides in the NW to the Kola Peninsula in the SE (Marker, 1991) (Fig. 2). The Lapland Granulite Belt is chiefly composed of metaturbidites (khondalites) with intercalations of intermediate to ultramafic meta igneous rocks metamorphosed under medium to high pressure granulite facies conditions (Barbey et al., 1984). Structurally the Lapland Granulite within Finland can be divided into a northeastern part of unshaped anatectic granulites and a southwestern part of highly sheared granulites (Fig. 2). The bedrock of northern Finnish Lapland is dominated by granulite and granite-gneiss.

The landforms of the area are gently undulating, which indicates a long duration of glacial erosion and subsequent deposition of basal till. Basal till is the predominant type of Quaternary deposit (Kujansuu and Niemelä, 1984). The mean thickness of the Quaternary deposits in Finnish Lapland is 5.9 m (Mäkinen, 1975). The Quaternary history of Lapland differs a good deal from the rest of Finland due to the glacial ice divide.

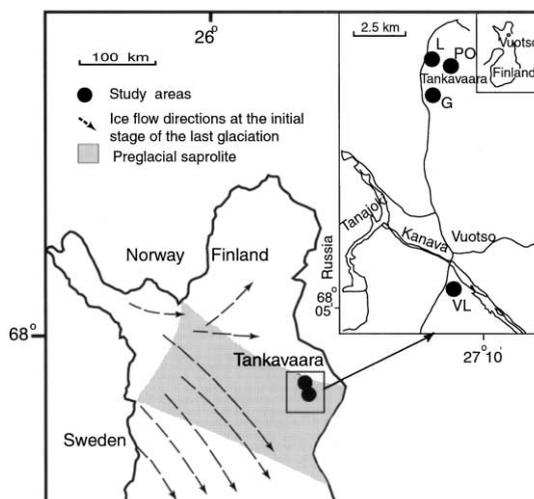


Fig. 1. Location map of the study area, Vuotso–Tankavaara, Finnish Lapland showing ice flow directions and preglacial saprolite (Modified from Koljonen, 1992). L, Lauttaojä; PO, Ponukuru; G, Tankavaara Gold museum; VL, Vuotso Air field section.

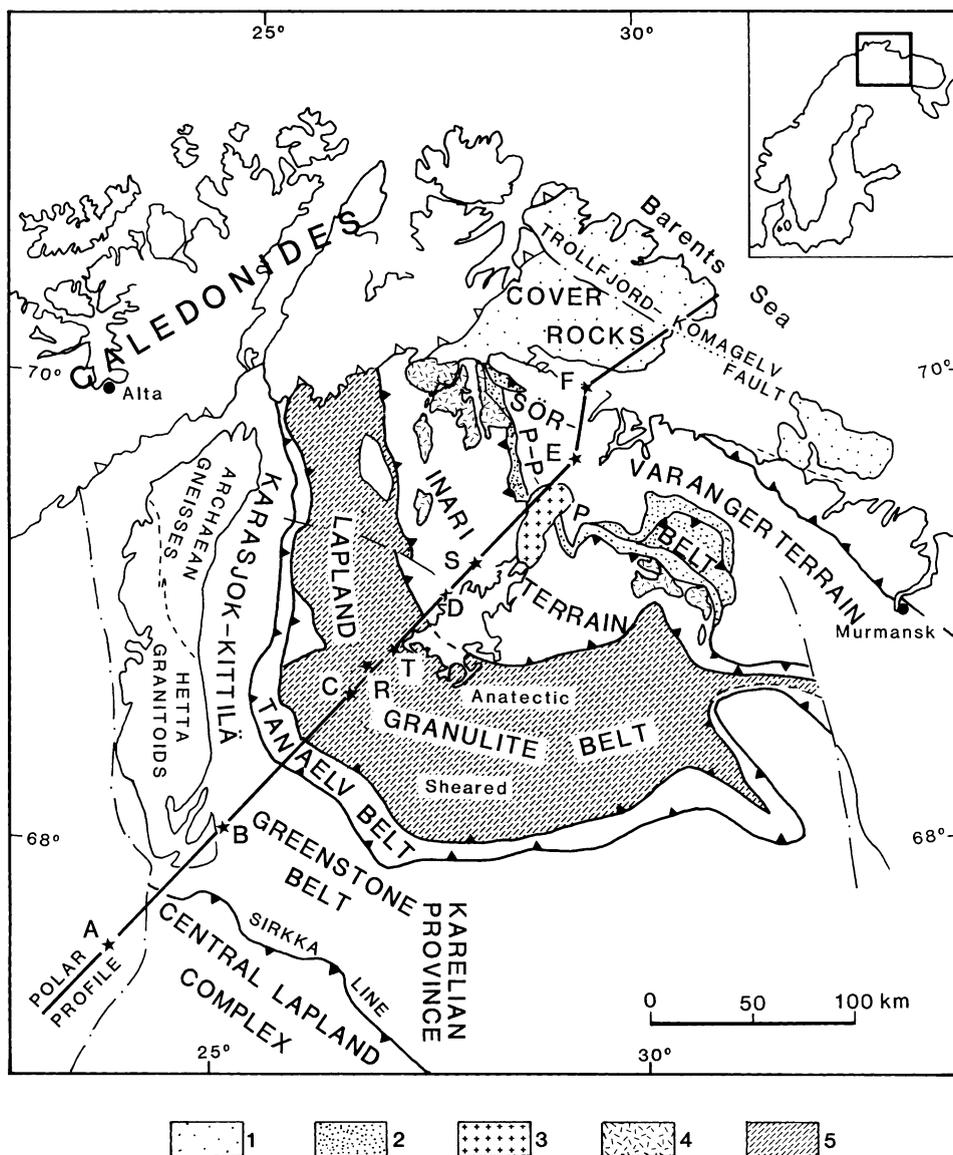


Fig. 2. Main tectonic units in the northern Baltic Shield. The EGT Polar Profile seismic line is shown with shot locations (A–F) marked by stars. 1, Late Proterozoic to Cambrian cover rocks; 2, Polmak–Pasvik–Pechenga Belt; 3, 1.8 Ga old granite bodies; 4, Ca. 1.95 Ga old dioritic to granodioritic intrusive rocks in the Inari Terrain; 5, Lapland Granulite Belt (after Marker, 1991).

The erosive action and transport capacity of the continental ice sheet was weaker in Lapland than in south Finland. In Lapland glacial erosion removed the upper portions of the saprolite. Ancient saprolite persists as isolated fragments elsewhere in Finland (Söderman, 1985).

In Finnish Lapland saprolite has various appearances due to parent material, grain size and degree of weathering. In western Lapland the upper portion of the saprolite is mixed with till to a varying degree, thus

demonstrating that the weathering took place before glaciation (Peuraniemi, 1989; Hirvas, 1992). In the Vuotso area, saprolite is common (Hyyppä, 1983). In some places saprolite has undergone intense chemical weathering and contains kaolinite and other clay minerals (Tanner, 1938). The thickness of saprolite varies substantially, being only a few meters in most places but tens of meters in others. At its thickest it may measure over 50 m (Sederholm, 1913) or even 100 m (Virkkala, 1955).

3. Field and laboratory methods

3.1. Field methods

During the summer of 1990, samples were collected from 4 weathering profiles developed on granulite and granite gneiss bedrock in the Vuotso-Tankavaara area of Finnish Lapland. The cores of saprolite were collected in steel boxes vertically from the bottom upward to till having a sample weight average of 6 kg. Sub-samples were selected on the basis of mineralogical, textural and field aspects and the subsampling interval was not less than 10 cm.

Two sampling sites are situated 2 km north of the Tankavaara Gold Museum, locally known as Lauttaoja and Ponukuru (Lat. 68°11'30"N: Long. 27°6'15"E). The elevation of the Lauttaoja and Ponukuru sites are 300 m and 340 m respectively. The distance between Lauttaoja and Ponukuru is about 400 m. Another sampling location is at the Tankavaara Gold Museum (Lat. 68°10'30" N: Long. 27°6'E) with an elevation of 310 m. All of these 3 locations lie on granulite. The fourth sampling site is Vuotso Airfield (Lat. 68°6'N: Long. 27°6'E) about 4 km south of Vuotso Village, situated on granite gneiss at an elevation of 260 m.

3.2. Laboratory methods

About 250 g of each weathered sample was oven dried at 100 °C for at least 72 h and then was crushed by a vibrating disk mill (Herzog, type: Hsm 100A). The crushed samples were used to determine the major element composition by X-ray fluorescence (XRF). Analysis was carried out using a Siemens SRS-303 AS XRF spectrometer with standard curves based on international rock standards [e.g. NIM-G and Ac-E granite, Sy-2 and Sy-3 Syenite, BIR-1 Basalt (Iceland), BHVO-1 Basalt (Hawaii), MRG-1 Gabro etc.] at the Institute of Electron Optics, University of Oulu, Finland.

For the determination of loss on ignition (LOI), the oven dried (110–120 °C) samples were ignited for 2 h at 950 °C and the LOI was determined.

For atomic absorption spectrometric (AAS) analyses 200 mg samples were dissolved in a Teflon crucible. One millilitre of HClO₄ and 2 ml of HF were added and the crucible was placed in a sand bath for heating. Afterwards, 2 ml of HCl was added to dissolve the samples and allowed to evaporate to dryness. Finally, the diluted fraction was analyzed for Cr, Cu, Ni, Pb and Zn using a Spectr AA300 atomic absorption spectrometer with an acetylene air flame.

Thin sections were prepared both from original core and mixed powdered saprolites for mineralogical analyses by petrographic microscope and electron microscope.

Differential thermal analysis (DTA) and thermogravimetric analysis (TGA) were carried out with the crushed and clay fraction (<0.002 mm) of the saprolite. Precalcined alumina was used as the inert material. Analyses were carried out by NETZSCH-STA 409 EP with a heating rate of 13 °C/min from 20 to 1100 °C in air.

For clay mineralogical studies, the clay fractions (<0.002 μm) were separated by centrifuging, then placed on a glass slide as a thin layer and allowed to dry at room temperature for analysis. X-ray diffraction (XRD) analysis was performed on the clay fraction using a SIEMENS D 5000 X-ray diffractometer with Ni filtered CuK_α radiation using 40 kV–40 mA. The step size was 0.02° and step time was 1 s. The scan range was 2–40°. Clay fractions were analysed 5 times after heating to temperatures of 20, 230, 450, 550 and 620 °C in order to identify the various clay mineral phases.

For electron microprobe (EPMA) analyses, minerals in a C coated polished thin section were analysed using a Jeol JXA-733 superprobe instrument by the wavelength dispersive technique (WDS). During the analysis, 15 keV accelerating voltage, 15 mA probe current and 10 μm beam diameter were employed. Natural and synthetic minerals were used as standards. Thin sections were also studied using a polarizing microscope.

Clay-size fractions were prepared for transmission electron microscopic (TEM) study by dispersing material in alcohol. The samples were placed on a formvar coated TEM grid (150 mesh) and examined with a Jeol, JEM-100 CX II electron microscope. A few thin sections of clay-size fractions coated with an Au–Pd conductor (Polaron SEM coating unit E 510) were examined morphologically under the Jeol JSM 6400 (with Link EXL, EDS) scanning electron microscope (SEM).

3.3. Method of calculation of geochemical data

The percentage changes to ratios, relative to the fresh parent rock, are calculated according to the following equation

$$\% \text{ change} = \left[\frac{(R_s - R_p)}{R_p} \right] \times 100 \quad (1)$$

where R_s is the ratio in a sample and R_p is the ratio of the same elements in the parent material.

4. Results

4.1. Field observations

The ancient saprolite found in the Lauttaoja section is soft and loose and can be dug easily by spade. The

colour of the crust is white to light grey interbedded with dark brown layers of material that contain mostly dark minerals (Fig. 3A). Some layers are totally white. The saprolite contains the primary minerals quartz and feldspar. The weathered bedrock still shows its original structures, although the upper part of the crust is mixed with overlying till material.

In the Ponukuru and Tankavaara Gold Museum sections, the lithological character is similar but the saprolite is darker and harder than that of the Lauttaoja section due to the presence of more unaltered garnet at the base of the saprolite profiles. Since these two sections are situated on the hilltops and the upper intensely weathered saprolites were washed away by glacial erosion, only the lower part with less intensely weathered saprolite are present.

The saprolite of the Vuotso Airfield section is reddish brown to grey in colour, indicating (Fig. 3B) a high degree of oxidation. The saprolite contains coarse to medium grained sandy material. In some places the saprolites contains very dark material that is amphibolitic and contains a high abundance of ferromagnesian minerals. The original structure of the bedrock can be seen in the saprolite.

4.2. Geochemical analyses

Geochemical analyses of saprolites on granulite, granite gneiss and amphibolite have been compared to

published analyses of unweathered rock samples (granulite, granite gneiss and amphibolite) from Finnish Lapland and other areas of Finland and from outside Finland (Tables 1 and 2).

According to these data, granulitic saprolite from Lauttaoja, Ponukuru and Tankavaara Gold Museum has been depleted significantly in Si, Ca, Na and K and markedly enriched in Al, Fe, Mg, Ti, Mn and water as compared to fresh bedrock (Fig. 4). Among trace elements, Cr and Ni have been enriched in the saprolite.

On the other hand, in Vuotso Airfield section, saprolite on the granite gneiss bedrock is enriched markedly in Fe and slightly in Ti, Mn, H₂O and Ni; it is depleted in Si, Al, Mg, Ca, Na, K, P and Cr. The saprolite derived from amphibolite (found in this section) has been enriched in Al, K, Fe, Ti, P and H₂O, and depleted in Si, Mg, Ca and Na (Fig. 4).

Furthermore, the relative changes in alkali metals and alkaline earths in the saprolite developed on the different bedrock types also can be seen in Fig. 4. Potassium has been depleted much more than Mg, Ca and Na ($K > Na > Ca > Mg$) in granulitic saprolite. On the other hand in granite-gneissic and amphibolitic saprolite the depletion trend is $Ca > Mg > Na > K$.

While there are no systemic increases or decreases in elemental concentrations with depth (vertical profile, Fig. 5), there are observable trends. SiO₂ and Al₂O₃ are significantly correlated ($r^2=0.46$) and are particularly high in the depth interval 1.05–1.35 m. MgO, Fe₂O₃ and

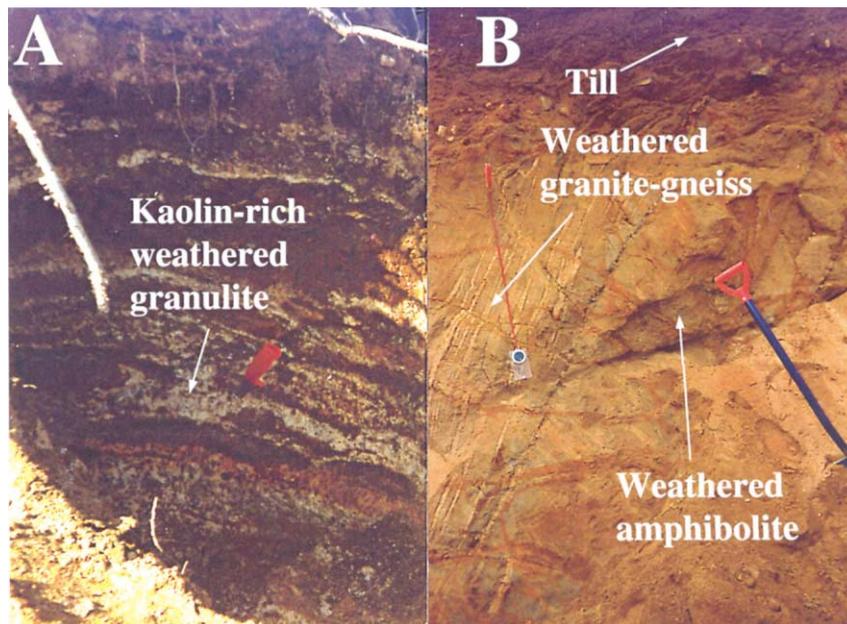


Fig. 3. (A) A typical weathering profile over granulite bedrock at Lauttaoja section. The alternating light (kaolin rich) and black-brown layers still show the original bedrock structure. (B) A weathering profile developed on the granite gneiss bedrock near the Vuotso Air field. Saprolite below the massive till can be seen. The black material along the cross-bedding (middle) is weathered amphibolite.

Table 1

Major and trace element composition of fresh rock types of granulite complex, granite gneiss and amphibolite from Vuotso and Inari areas, Finnish Lapland

Wt. %	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
SiO ₂	48.45	52.19	59.23	61.43	66.37	69.98	75.20	74.17	62.25	67.59	82.09	68.64	73.67	76.50	70.80	72.46	51.23
Al ₂ O ₃	15.61	20.00	19.29	19.86	16.71	16.49	16.02	13.62	16.62	19.74	9.09	15.42	13.92	12.52	15.25	11.35	14.38
MgO	7.84	5.23	3.92	2.93	1.76	2.19	0.08	0.33	3.38	4.05	0.97	2.15	0.26	0.20	0.92	0.21	6.99
CaO	11.28	7.49	5.95	0.37	3.90	1.77	1.74	0.93	1.65	0.45	1.25	1.56	1.19	0.56	3.15	1.85	9.97
Na ₂ O	1.85	3.26	3.36	0.99	3.23	2.35	3.16	2.19	1.53	0.85	2.10	3.34	3.44	4.00	4.68	2.95	2.69
K ₂ O	0.43	0.51	0.94	4.68	1.01	0.78	3.00	6.63	3.58	2.75	0.73	2.43	5.01	3.89	1.37	3.96	0.60
Fe ₂ O ₃	12.19	9.73	5.54	7.03	4.35	5.40	0.25	4.35	9.66	2.94	2.92	4.03	1.51	1.78	2.64	4.56	11.53
TiO ₂	1.19	0.46	0.39	0.78	0.57	0.53	0.03	0.06	0.56	0.07	0.31	0.58	0.16	0.12	0.37	0.32	1.31
MnO	0.23	0.33	0.08	0.04	0.06	0.12	0.005	0.01	0.14	0.01	0.04	0.04	0.04	0.05	0.04	0.09	0.20
P ₂ O ₅	0.10	0.05	0.13	0.03	0.23	0.06	0.08	0.08	0.03	0.03	0.05	0.08	0.04	0.05	0.10	0.04	0.13
LOI	0.53	0.04	0.62	1.28	1.60	0.67	0.56	0.80	0.70	1.68	0.57	1.54	0.51	0.35	0.50	2.19	–
Total	99.70	99.29	99.45	99.42	99.79	100.16	100.125	101.73	100.00	100.16	100.12	99.81	99.75	100.02	99.83	99.98	99.03
Cr	203	118	109	127	35	86	1	7	121	10	58	–	–	–	–	–	–
Ni	76	55	42	71	14	25	1	2	54	21	14	–	–	–	–	–	–

Note: 1. Hypersthene–clinopyroxene–plagioclase rock, Kuttura; 2. hypersthene–plagioclase rock, Kultala area; 3. garnet–hypersthene–plagioclase rock, Kuttura; 4. sillimanite–garnet–granulite, Kuttura; 5. sillimanite–garnet–granulite, NE Kuttura; 6. garnet granulite, W Kultala; 7. sillimanite–garnet–granulite, Sotajoki; 8. sillimanite–garnet–granulite, NE Kuttura; 9. garnet–cordierite metatexite, W Kultala; 10. garnet–cordierite diatexite, Ivalo; 11. garnet–feldspar quartzite, Mukkapalo; 12. biotite gneiss, man area, Finnish Lapland; 13. granite, man, Kyynejärvi; 14 and 16. arkose gneiss, man, Vaskojoki; 15. archaean grey gneiss, Suomussalmi–Kuhmo area; 17. amphibolite, Finnish Lapland SL. No. 1–11 (Hörmann et al., 1980); 12–14, 16 and 17 (Meriläinen, 1976) and 15 (Martin et al., 1983)

MnO are highly correlated and all are negatively correlated with SiO₂ and Al₂O₃, although overall patterns change more abruptly. CaO and Na₂O are also positively correlated ($r^2=0.56$) and show abrupt changes that are negatively correlated with MgO, Fe₂O₃ and MnO. K₂O tends to be relatively lowly concentrated at shallow depths and have relatively high concentration deeper in the profile.

4.3. Mineralogical analyses:

XRD: the X-ray data show (Fig. 6A) the primary minerals present in the samples are mainly quartz, feldspars (plagioclase and microcline) and almandine garnet. Sodium and Ca feldspars (plagioclase) can be identified by their 3.21–3.15 Å peak and microcline can be identified by its 3.24 Å peak (Brown and Brindley, 1980). Almandine is identified by its typical 2.57 and 2.87 Å peaks.

In addition to the presence of the primary minerals, the main clay minerals are kaolinite and halloysite. Vermiculite and illite were observed also in some samples. Kaolinite and halloysite can be identified through their common 3.56–3.62 Å (JCPDS, 1980), 4.43–4.44 and 7.2–7.5 Å (Brown and Brindley, 1980) peaks. Halloysite was identified by a broad (001) peak at 7 Å, by a strong asymmetric peak at 4.44 Å (Calvert et al., 1980; Fig. 7A; L & VL) and by plateau peaks between 2.50 and 2.31 Å.

Differentiation of kaolinite and halloysite, if they occur in the same samples, is, of course, very difficult or impossible on the basis of XRD alone. The most reliable means to recognize the difference between these two phases is electron microscopy (cf. Beutelspacher and Van der Marel, 1968). Vermiculite can be identified by its typical 14–14.6 Å peak range (Brown and Brindley, 1980) along with one at 3.36 Å. Illite (trioctahedral) can be identified by its 10 Å asymmetrical peak at low angles. Heating of a sample to 550 °C serves two important functions. It effects collapse of vermiculite which contains non-exchangeable interlayer Al complexes, and it destroys the kaolin minerals. Vermiculite is confirmed by heat treatment through the disappearance of the 14 Å peak followed by an increase in the 10 Å peak (Fig. 6B).

DTA and TGA: DTA–TGA graphs (Fig. 7) for the crushed samples and clay fractions (<0.002 mm) show that quartz and muscovite are the primary minerals; and kaolinite and halloysite are the secondary clay minerals in the saprolite.

The typical endothermic peak of quartz at 573 °C is found on most of the DTA curves. A very small trend of an endothermic peak in the range of 800–900 °C (Fig. 7B) and a downward trend of the curve at 1100 °C (Fig. 7C) is indicative of muscovite (Mackenzie, 1970). DTA–TGA graphs show the presence of kaolinite, but the first endothermic reaction in the DTA and the high initial weight loss in TGA imply also the presence of a more water bearing phase than normal kaolinite (Velde,

Table 2
Chemical composition (wt.%) of saprolite from Vuotso–Tankavaara area, Finnish Lapland

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21
SiO ₂	40.71	46.22	45.40	42.79	43.32	40.90	53.00	39.40	52.89	52.70	52.02	40.52	37.90	41.61	42.84	55.85	58.85	74.46	59.57	55.47	52.68
Al ₂ O ₃	23.65	21.60	22.50	17.73	23.24	21.83	27.57	23.96	25.06	26.74	27.53	25.20	21.87	23.03	24.84	16.03	15.97	11.52	16.54	18.82	22.30
MgO	3.53	4.72	3.43	5.19	5.25	6.48	1.30	5.76	1.70	1.84	1.74	3.27	6.28	5.51	5.26	3.47	3.63	1.91	1.66	1.17	0.74
CaO	3.53	3.52	3.78	4.29	3.58	2.58	5.10	2.50	5.56	4.41	3.87	3.02	1.85	2.86	3.99	1.58	2.00	1.43	0.92	1.90	0.25
Na ₂ O	2.46	2.24	2.20	0.25	1.91	1.10	4.17	1.10	3.08	3.30	3.00	1.87	0.56	1.59	2.24	1.72	0.94	1.39	0.53	0.28	0.25
K ₂ O	0.64	0.80	0.75	0.15	0.50	0.54	1.00	0.69	0.91	0.98	1.00	0.93	0.61	0.71	0.68	1.38	0.40	0.74	2.10	1.57	2.56
Fe ₂ O ₃	11.58	13.49	13.57	19.98	15.55	18.35	4.31	18.16	6.44	6.00	5.84	11.57	21.61	17.13	14.21	14.53	11.35	4.95	11.06	12.13	12.30
TiO ₂	0.28	0.43	1.32	2.55	0.20	0.26	0.05	0.61	0.03	0.04	0.06	0.23	0.60	0.28	0.59	0.88	1.37	0.45	1.11	1.50	1.02
MnO	0.13	0.16	0.15	0.25	0.25	0.29	0.07	0.27	0.06	0.10	0.09	0.14	0.28	0.26	0.20	0.11	0.14	0.07	0.05	0.05	0.22
P ₂ O ₅	0.05	0.04	0.04	0.03	0.04	0.04	0.03	0.04	0.04	0.03	0.04	0.04	0.06	0.05	0.04	0.11	0.12	0.04	0.12	0.13	0.13
LOI	5.89	3.79	4.56	3.78	2.51	3.35	3.10	3.88	3.23	3.47	4.76	5.27	3.85	2.97	1.67	5.12	4.39	1.98	6.17	6.96	7.15
																100.78	99.16	98.94	99.83	99.98	99.60
Total	98.45	97.01	97.7	96.99	96.35	95.72	99.70	96.37	99.60	99.61	99.95	98.06	95.47	96.00	96.52	–	164	96	110	173	111
Cr ppm	–	334	–	157	–	242	71	–	105	–	52	–	312	291	–	–	88	36	87	110	116
Ni ⁱⁱ	–	70	–	112	–	61	34	–	45	–	39	–	129	102	–	–	–	–	–	–	–
	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36	37	38	39	40	41	42
SiO ₂	55.29	52.37	45.49	37.80	46.00	56.83	71.60	69.49	70.91	71.71	71.67	73.77	72.27	74.18	73.93	67.80	74.76	71.84	72.63	39.72	47.44
Al ₂ O ₃	21.74	22.90	20.10	18.34	18.36	20.19	11.52	12.20	12.23	11.58	11.90	11.98	12.80	12.08	12.02	11.84	11.91	11.70	11.44	16.18	16.3
MgO	1.32	1.25	3.09	3.25	2.95	1.66	2.17	1.97	0.17	0.15	0.14	0.10	0.15	0.13	0.20	0.22	0.24	0.21	0.10	3.75	2.66
CaO	0.29	0.28	1.12	1.53	1.17	0.79	2.11	1.82	1.04	1.28	1.20	0.20	0.40	0.47	0.22	0.25	0.27	1.21	1.40	5.08	3.87
Na ₂ O	0.47	0.51	0.17	0.08	0.30	0.80	2.01	2.24	2.75	2.62	2.98	4.15	4.38	5.48	3.70	2.81	3.82	3.10	3.19	1.68	1.97
K ₂ O	3.04	3.77	0.61	0.84	0.84	1.31	1.96	1.97	3.04	3.00	3.46	2.98	2.33	0.74	3.17	2.87	3.22	3.38	3.45	1.5	1.3
Fe ₂ O ₃	9.52	11.09	18.03	25.28	18.69	10.56	5.47	6.46	6.23	6.44	5.53	5.10	4.74	4.52	3.70	9.50	2.86	40	5.32	25.31	20.25
TiO ₂	0.94	1.03	2.15	3.19	2.53	1.00	0.80	0.79	0.37	0.41	0.34	0.32	0.37	0.32	0.33	0.36	0.33	0.33	0.32	2.46	2.46
MnO	0.18	0.13	0.21	0.34	0.26	0.11	0.06	0.07	0.12	0.10	0.11	0.03	0.08	0.05	0.05	0.08	0.11	0.11	0.11	0.4	0.35
P ₂ O ₅	0.13	0.14	0.14	0.18	0.14	0.10	0.05	0.06	0.07	0.06	0.05	0.05	0.06	0.04	0.04	0.11	0.05	0.05	0.03	0.31	0.2
LOI	7.28	7.33	7.24	7.61	0.57	6.39	1.86	2.28	1.90	1.62	1.23	0.89	1.54	1.07	1.44	3.02	1.17	1.10	0.67	4.99	5.04
Total	100.20	100.8	98.35	98.44	97.81	99.74	99.61	99.35	98.83	98.97	98.61	99.57	99.12	99.08	98.80	99.11	98.47	98.72	98.66	101.38	101.84
Cr ppm	103	–	162	–	135	99	–	–	14	13	–	–	28	32	8	–	–	–	9	–	–
Ni ⁱⁱ	112	–	207	–	143	109	–	–	15	7	–	–	16	7	8	–	–	–	5	–	–

Sample Nos. 1–15, weathering crust from Lauttaoja section (granulite); sample Nos. 16–18, weathering crust from Ponukuru section (granulite); sample Nos. 19–27, weathering crust from Tankavaara Gold Museum section (granulite); sample Nos. 28–31, weathering crust from Vuotso Air field section (granite–gneiss); sample Nos. 32–40, weathering crust from Vuotso Air field section (granite–gneiss) and 41–42 (amphibolite).

1992). According to Velde (1992) the initial weight loss for halloysite is 7% and the weight loss (loss of crystalline water) between 500–600 °C is 13%. These are comparable with the TGA curves from Vuotso. According to Mackenzie (1970), halloysite gives a curve (DTA)

with a large dehydroxylation peak at 560 °C. According to Barshad (1965), the peak temperature is about 20–30 °C higher for kaolinite than halloysite; and the break ends at 625 °C for kaolinite and 595 °C for halloysite. The curves show similar phenomena as those of halloysite.

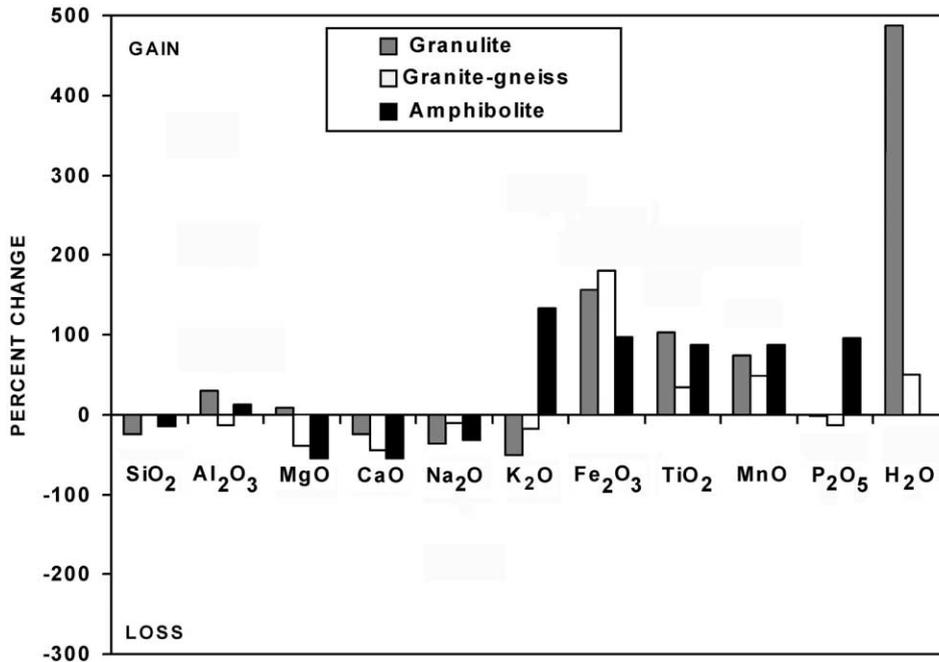


Fig. 4. Elemental percentage changes of the ratios (with respect to bedrock) for the weathered samples (major elements) from granulite, granite gneiss and amphibolite. Percentage changes use the average composition of the parent rocks as a reference.

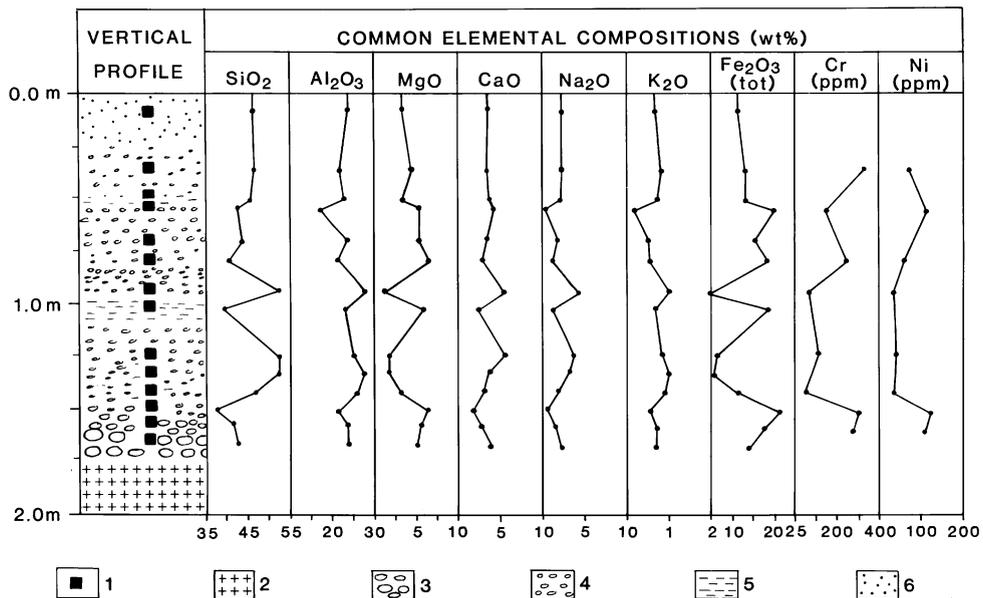


Fig. 5. Vertical profile of elemental composition within a typical section. In saprolite at Lauttaoja (granulite). 1, sampling spot; 2, bedrock; 3, youngest saprolite; 4, deep brown and harder saprolite; 5, light brown and softer saprolite; 6, till (massive).

TEM and SEM: TEM and SEM are commonly used as the main criteria distinguishing kaolinite from halloysite (Robertson and Eggleton, 1991; Singh and Gilkes, 1992). A well developed platy form of kaolinite

(Fig. 8C) and tubular halloysite (Fig. 8A–C) have been identified in the Tankavaara Gold Museum, Ponukuru, Lauttaoja and Vuotso Airfield sections (Figs. 8 and 9) which resemble those published by

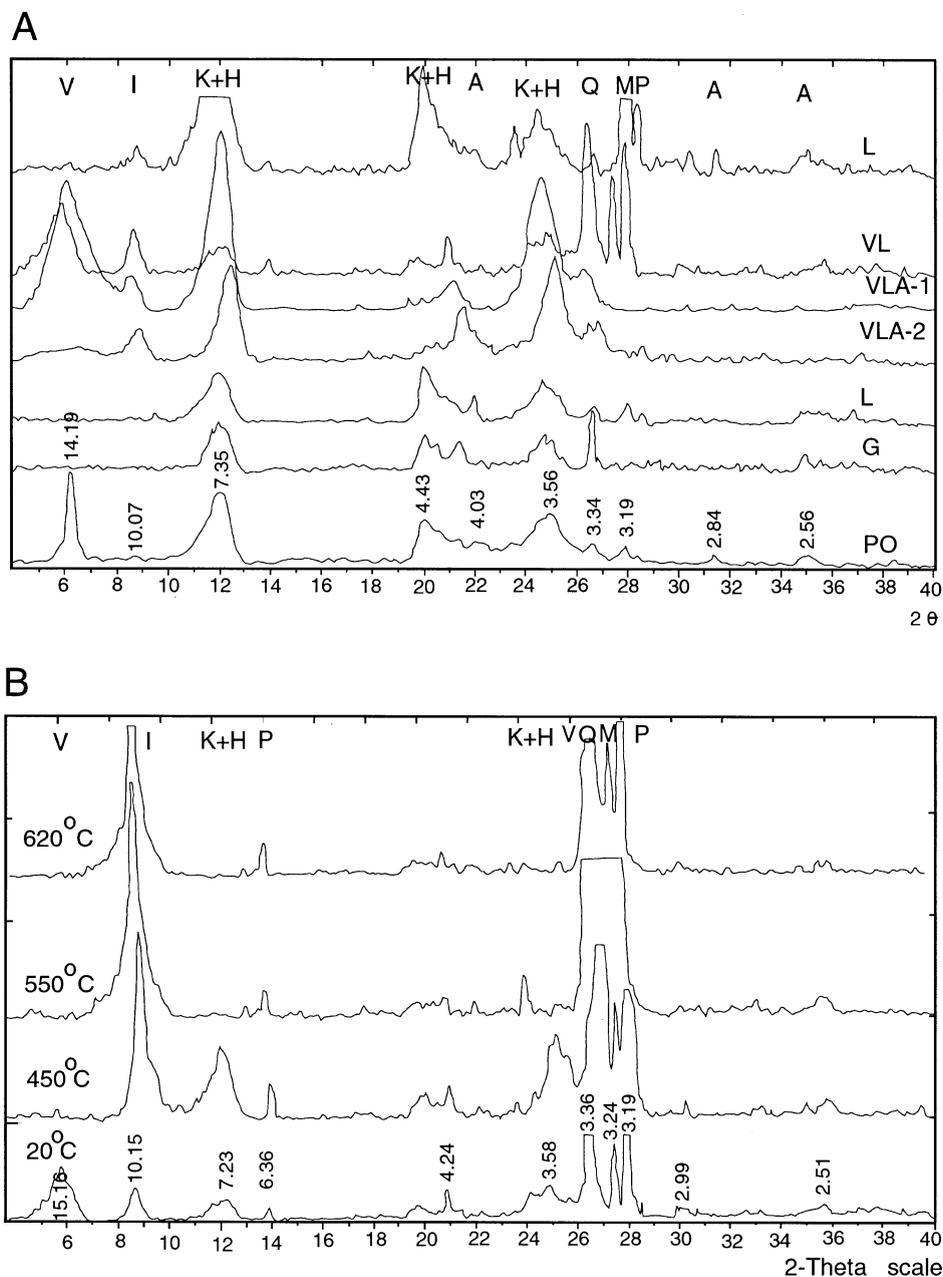


Fig. 6. (A). X-ray graphs for representative samples (clay fraction, 2 μm) with oriented glass slide: V, vermiculite; I, illite, K, kaolinite, H, halloysite; Q, quartz; M, microcline; P, plagioclase; A, Almandine. The mineral composition varies from place to place. The site locations are: L, Lauttaoja (0.90 m); VL, Vuotso Airfield (1.45 m; granite-gneiss); VLA1, Vuotso Airfield-amphibolite (2.00 m); VLA2, Vuotso Airfield-amphibolite (3.00 m); G, Gold Museum (1.00 m) and PO, Ponukuru (0.75 m). (B) Effect of heating, shown in X-ray diffraction diagrams from the Vuotso Airfield section (clay fraction) (1.45 m). Heating causes vermiculite to disappear, while there is an increase in the 10 Å peak. Kaolinite and halloysite have both disappeared. V, vermiculite; I, illite; K, kaolinite; H, halloysite; Q, quartz; M, microcline; P, plagioclase.

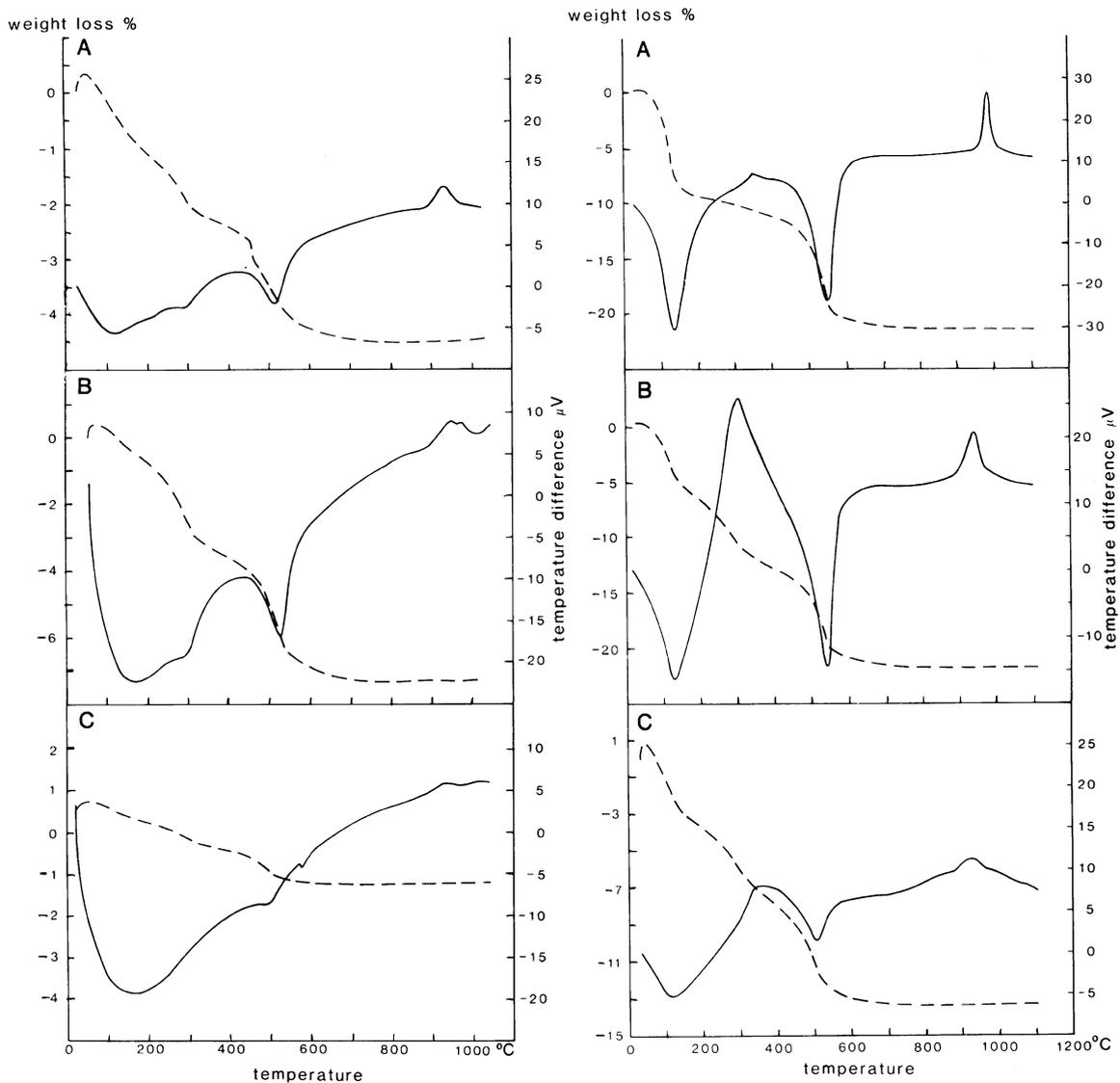


Fig. 7. DTA–TGA graphs for the crushed samples (left) and clay fraction (right). Solid line, DTA; dashed line, TGA; A, Lauttaoja (1.5 m); B, Gold Museum (1.2 m); C, Vuotso airfield (0.75 m).

Beutelspacher and Van der Marel (1968) and by Bates (1971).

Petrographic microscopy: the primary minerals found in the thin sections of weathered granulite under the petrographic microscope were plagioclase feldspar, quartz, K-feldspar, garnet, sericite, biotite and accessory minerals (Fig. 10A and B). In the granite gneiss from the Vuotso air field section quartz, plagioclase feldspar, biotite and some hornblende were found (Fig. 10C). The thin section of fine-grained black-green amphibolite contains mainly hornblende, quartz, plagioclase feldspar, K-feldspar, biotite and accessory minerals (Fig. 10C). The secondary minerals present in all of

these weathering crusts were extremely difficult to identify through the petrographic microscope.

EPMA: representative microprobe analyses of almandine garnet and its weathering products formed in the fracture plane of garnet from Lauttaoja section are given in Table 3. With respect to fresh almandine, its weathering product is enriched in Si and Al and depleted in Mg, Ca and Fe. The chemistry suggests that the weathering product formed in the fracture plane of almandine garnet is Fe-rich kaolinite (Deer et al., 1966). BEI has also documented initial etching of plagioclase (e.g. Fig. 11A) and severely weathered orthoclase grains (e.g. Fig. 11B).

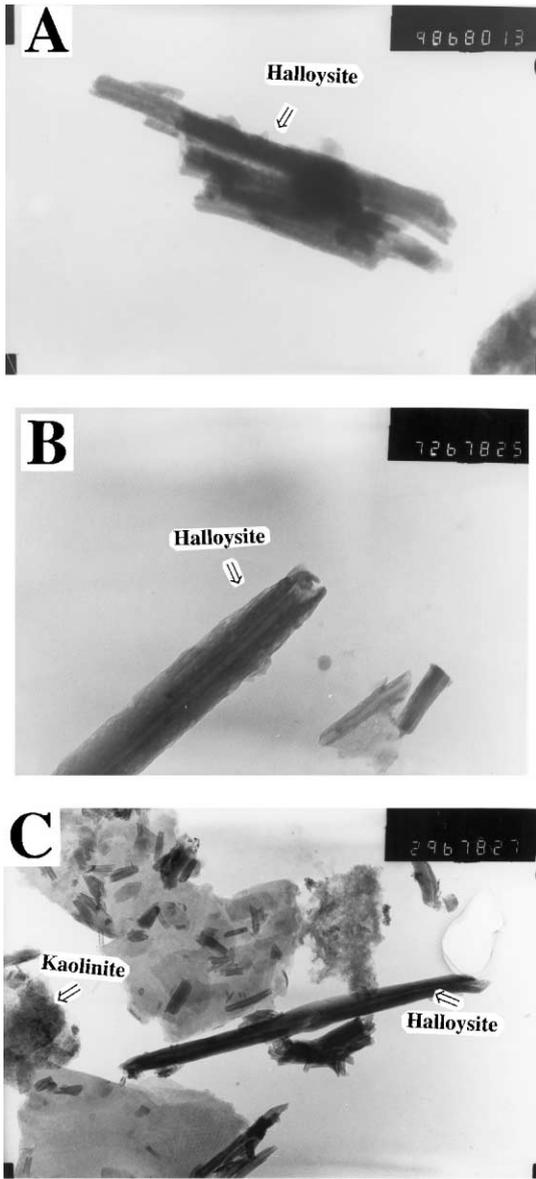


Fig. 8. Halloysite tubes (TEM) from A, Ponukuru ($\times 48,000$); B, Tankavaara Gold Museum ($\times 29,000$) and C, kaolinite and tubular halloysite in Ponukuru section ($\times 48,000$).

5. Discussion

5.1. Geochemistry

In the study area (4 sections) progressive weathering of the rocks and formation of saprolite has resulted in gradual enrichment in Al, Fe, Ti, Mn and H₂O; and in depletion of Si, Na, K and Ca (Fig. 4). The exception is amphibolitic saprolite, where K is enriched. The most direct indicator of weathering intensity of granulite and



Fig. 9. SEM photograph of Au–Pd coated oriented clay fraction (2 μ m) from Lauttaoja section showing kaolinite flakes and halloysite tubes ($\times 15,000$).

Table 3

Chemical composition of almandine garnet and its weathering product

Wt. %	Almandine	Almandine's weathering product
SiO ₂	38.75	44.33
Al ₂ O ₃	21.92	33.93
MgO	9.38	1.26
CaO	2.49	0.11
Na ₂ O	0.01	0.06
K ₂ O	0.01	0.22
FeO	26.65	8.09
TiO ₂	0.02	0
MnO	0.48	0
Cr ₂ O ₃	0.08	0.01
NiO	0	0.07
ZnO	0	0.06
V ₂ O ₃	0	0.03
Total	99.79	88.17
	On the basis of 24(O, OH ⁻)	On the basis of 18(O, OH ⁻)
Si	6.12	5.1
Al	4.08	4.57
Mg	1.57	0.13
Ca	0.41	–
Na	–	–
K	–	–
Fe	3.51	0.76
Ti	–	–
Mn	0.12	–
Cr	–	–
Ni	–	–
Zn	–	–
V	–	–

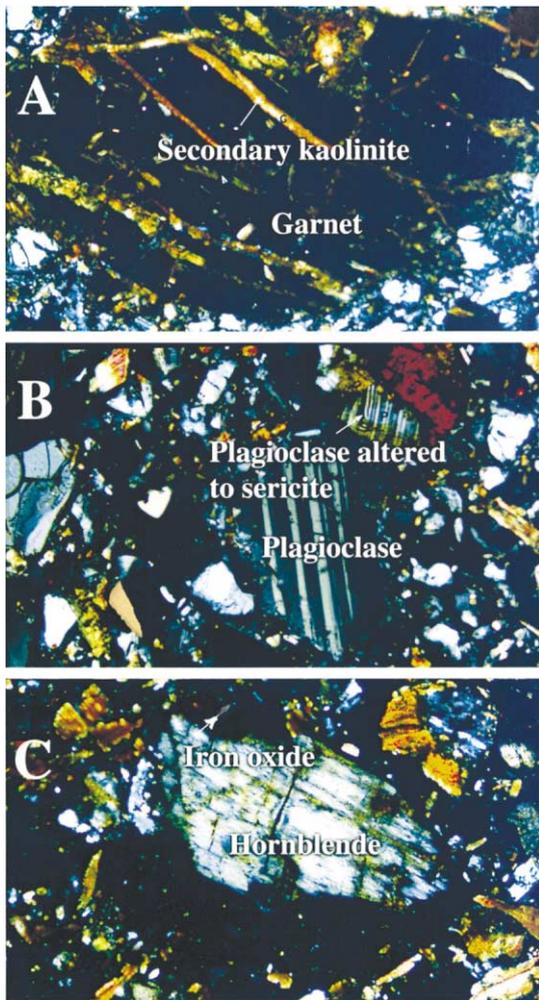


Fig. 10. (A) Thin section showing the effect of weathering on granulite from Lauttaoja section with etching along fracture planes of (almandine) garnet and secondary minerals formed in the fracture planes at a moderate stage of alteration; (B) granulite from Lauttaoja section showing etching at the early stage of alteration along the grain boundaries of plagioclase and plagioclase being altered to sericite (top), and (C) amphibolite from Vuotso airfield section showing the effect of weathering along the boundaries, surface, and cleavage planes of hornblende.

granite gneiss is the content of H_2O . With increased alteration, H_2O increases due to formation of clay minerals (kaolinization reaction). With increased alteration, saprolite shows relatively marked enrichment of H_2O and Al_2O_3 and relatively major depletion of silica indicating formation of kaolinite (Fritz, 1988). Buxton (1968) proposed a weathering index based on the SiO_2/Al_2O_3 ratio of the bulk soil. He found that fresh Si–Al rocks commonly have values ~ 4.0 – 4.5 , the ratio systematically decreased to 2.0 in bulk soil. In granulitic saprolite, the average SiO_2/Al_2O_3 ratio is 2.30

whereas the value of fresh granulitic bedrock is 4.33 which is indicative of intense weathering. In amphibolitic saprolite the SiO_2/Al_2O_3 ratio is 2.68 as compared to fresh amphibolite (3.56).

The marked increased concentration of Fe in saprolite from all the 4 sections is due to: (1) a higher population of biotite and (2) formation of Fe-hydroxide (amorphous) and to some extent higher concentrations of opaque minerals relative to that in the fresh rock. The higher concentration of Fe(III) and H_2O and reduced Na in the weathered bedrock of more or less all 4 sections in the study area is indicative of oxidation, hydration and leaching processes involved during weathering (e.g. Mikkel and Henderson, 1983). The enrichment of Ti is due to the higher abundance of sphene in the intensely weathered rocks, due to its stability during weathering.

The decreasing trends for Na, Ca and K in all 4 sections in Vuotso reflect the alteration of feldspars. In the granite gneiss saprolite, the Na and Ca are more depleted than K (Fig. 4) resulting from the rapid weathering of plagioclase and Na-feldspar in comparison to K-feldspar (Nesbitt et al., 1980). Furthermore, greater depletion of Ca than Na indicates that plagioclase feldspar has weathered more than sodic feldspar. The granulitic saprolite shows the depletion trend $K > Na > Ca$ which indicates that K-feldspar has weathered more than plagioclase and sodic feldspar. The occurrence of halloysite in the weathering crust is the result of weathering of K-feldspar.

The enrichment of K in the amphibolitic saprolite is due to the formation of illite. In this saprolite, leaching is strong. Mg, Ca and Na are being depleted extensively during weathering. The increase of Mg in granulitic saprolite compared to fresh rock is due to a higher proportion of biotite (including hydromica). In the Vuotso Air field section (granite gneiss and amphibolite), Mg content registers a depletion due to alteration of biotite.

The concentration of Ni in weathered granulite could be attributed to precipitation from percolating solutions, and the enrichment of Cr in the weathered profile is due to the formation of kaolinite (McLaughlin, 1959). The host minerals for Ni and Cr in the study areas are hornblende, garnet and biotite (Nockolds and Mitchell, 1948; Devore, 1955; Carswell, 1968a,b; Haack, 1969; Reimann and Caritat, 1998). The Ni is mobilized during weathering; it is, however, co-precipitated with Fe and Mn oxides. In oxidizing conditions, Ni is relatively mobile (Reimann and Caritat, 1998). According to Matzat and Shiraki (1978), Cr^{+3} , which closely resembles both Al^{3+} and Fe^{3+} in both chemical properties and ionic size, will behave similarly to these ions during the weathering process and will be concentrated in clays. As kaolinite and halloysite are the main minerals, the enrichment of Cr and Ni likely is caused by adsorption on these clay minerals and Fe oxide.

In the vertical profile, SiO₂ and Al₂O₃ correlation is primarily the result of the influence of plagioclase feldspars, garnet and clay minerals. A semiquantitative determination of mineralogical composition in a representative sample from weathered granulite shows plagioclase 75.59%, K-feldspar 3.14%, garnet 3.14%, and kaolinite and halloysite 12.33% (Table 4). The higher concentration of SiO₂ and Al₂O₃ in the depth interval 1.05–1.35 m is likely due to the influence of a higher abundance of unweathered plagioclase feldspar where Fe₂O₃ concentration is significantly low. The significant correlation between CaO and Na₂O indicates weathering effects on plagioclase feldspar where both calcic and sodic-feldspars are equally affected.

The enrichment of MgO, Fe₂O₃ and MnO indicates the relative abundance of garnet, biotite and the formation of Fe hydroxide, which is consistent with the deep brown and harder lithological character in the samples between the depths of 1.0 and 1.5 m (vertical profile). These phenomena are negatively correlated with CaO and Na₂O, indicating that one layer of saprolite is dominated by plagioclase feldspar while the other is dominated by garnet, biotite, and Fe oxides. In the vertical profile, the increasing concentration of K₂O with depth is due to the effect of intense leaching, which decreases with depth.

The development of a thick weathering profile with associated segregation of elements and extensive leaching of silica indicates extreme weathering conditions, similar to that observed in the tropic and subtropics today (Hill et al., 2000).

5.2. Mineralogy

The suite of analyses performed here by DTA, TGA, XRD, TEM, SEM, petrographic microscope and electron microprobe reveal that plagioclase feldspar, K-feldspar, quartz and garnet are the main primary minerals, and kaolinite and halloysite are the main secondary minerals found in granulitic saprolite. The

primary minerals found in the granite gneissic saprolite are mainly plagioclase, quartz, K-feldspar, garnet and hornblende. Amphibolitic saprolite contains mainly hornblende, plagioclase, quartz and garnet. Kaolinite, halloysite and vermiculite are the main secondary clay minerals formed both in granite–gneissic and amphibolitic saprolite. A semiquantitative estimation was done by SEM with one representative sample from granulitic saprolite (Table 4).

During the early stages of alteration there appears to be general dissolution and etching along surfaces and grain boundaries as well as along the cleavage and fracture planes of the minerals. Quartz persists throughout the saprolite, although many grains are fractured and contain veins of secondary minerals. In thin section, plagioclase shows clear evidence of etching on the surfaces at the early stages of alteration (Fig. 10B) and along the boundaries (Fig. 11A). At the extreme stage of weathering, plagioclase is altered to sericite (Harris and Adams, 1966) and clay minerals. Examination of sericite in soils is very limited because of its highly localized occurrence as thin weathering coatings around feldspar grains (Arocena et al., 1993). Exactly the same kind of feature is seen in the Fig. 10B (p). Arocena et al. (1993) reported that a soil thin section from the foothills of the Rocky Mountains in Alberta showed that most of the feldspars exhibit alteration products with interference colors similar to finely-grained mica or so called “sericite.” Sericite usually refers to the microcrystalline variety of muscovite (Kerr, 1977) but may include hydromica (Twenhofel, 1950). The same phenomenon is noticed in the sample from the Louttaoja section (Fig. 10B, upper right) where plagioclase is being altered to sericite. Orthoclase is affected severely by weathering along the surface of the mineral as shown by BEI (Fig. 11B). The K-feldspar is more altered compared to plagioclase in the granulitic bedrock.

Almandine is the typical garnet of the granulite facies that has been found in granulitic bedrock in Finnish Lapland and is highly fractured and altered due to weathering. Alteration of garnet starts along the boundaries and fracture planes as well as cleavage planes during the early stage of weathering. In the advanced stages, fracture planes become wider through the formation of secondary minerals. The secondary minerals formed in the fracture planes have been analysed by electron microprobe and identified as Fe-rich kaolin mineral [Fig. 10A and Table 3].

The nature of alteration of primary minerals in the Vuotso Air field section is more or less the same as described above for the granulitic saprolites. Hornblende, the main ferromagnesium mineral in amphibolitic saprolite is the most intensely altered mineral in this section. Amorphous Fe hydroxide is formed through the weathering of hornblende in weathered amphibolite (Fig. 10C). Alteration of biotite produces either smectite

Table 4
Semiquantitative determination of minerals in granulitic saprolite from SEM

Minerals	%
Plagioclase	75.59
K-feldspar	3.14
Quartz	2.88
Garnet	3.14
Biotite	0.78
Sericite	0.26
Kaolinite and halloysite	12.33
Unclassified	1.83
Total	99.95

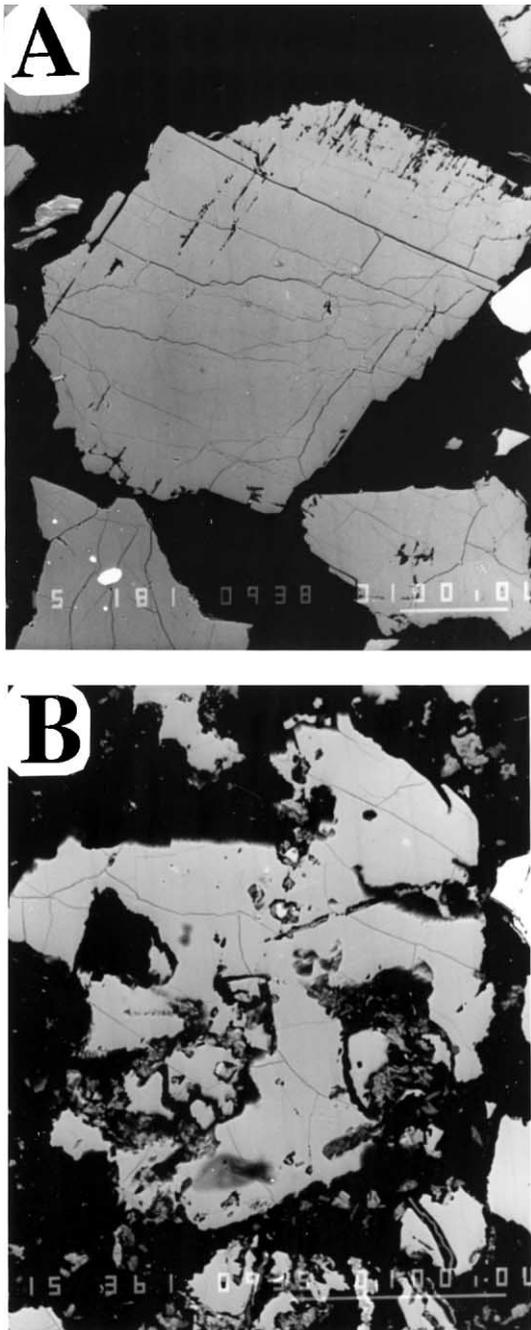


Fig. 11. Backscattered electron image (BEI) of a sample from Vuotso Air field section (granite gneiss) showing (A) the evidence of etching along the surface of plagioclase, and (B) intensely weathered orthoclase.

or vermiculite. In fact, all of the rock types of the 4 sections from the study area show common alteration of biotite to green biotite, muscovite and possibly also sericite. The biotite from amphibolite shows a lower oxide sum (about 88%) in microprobe analyses indicating the

presence of a more water-bearing phase, which is the form of hydromica.

The presence of kaolinite in amphibolitic saprolite indicates an intense weathering stage in which vermiculite has been decomposed to kaolinite. The review work carried out by Weaver (1989) pointed out the alteration of biotite to vermiculite (Torrent and Benyas, 1977) and/or smectite in temperate regions; conversely in the humid tropics it tends to weather directly to kaolinite or halloysite. The abundance of kaolinite and halloysite and the sparse occurrence of vermiculite in granite gneissic and amphibolitic saprolite indicates much warmer and more humid climate conditions than at present. Clearly temperature at the time of weathering was higher, and kaolinite and several forms of halloysite have formed through the weathering of K-feldspar as reported by Keller (1978) and Wilke et al. (1978).

Halloysite and kaolinite are important weathering products of rocks in temperate and tropical environments. Previous electron microscopic studies frequently reported the coexistence of both minerals in weathering profiles (Keller, 1977; Gilkes et al., 1980; Banfield and Eggleton, 1990). Their coexistence is of interest because the chemical composition of kaolinite is the same as that of halloysite except for weakly bound interlayer water (Weaver and Pollard, 1973; Newman and Brown, 1987), while the structure of halloysite differs from that of kaolinite in layer stacking sequence (Fig. 8A shows the stacking of halloysite in the study area). Thermodynamic studies predict that halloysite would convert into kaolinite with ageing (Huang, 1974; Tsuzuki and Kawabe, 1983). The overall observation from the SEM micrographs (Fig. 9) shows that the relative abundance of tubular halloysite is greater than platy kaolinite. As noted above, the saprolites of the study area represent almost the core of the weathering profile. The uppermost part of the saprolite profiles have been eroded due to glacial flow. The abundance of earlier formed halloysite at the weathering front should be followed by kaolinite, converted from halloysite at a later stage. More extensive microstructural study is required to elucidate the genetic relation between halloysite and kaolinite (Jeong, 1998).

5.3. Influence of glaciation

In spite of weak glacial erosion (Hirvas, 1992) in the ice divide zone in the study area, there have been some remarkable effects of glacial erosion. Saprolite is present in depressions and fracture zones, but absent or marginally present on higher ground. As the uppermost part of the saprolite has been eroded, only the bottom part of the saprolite now exists which has comparatively younger weathering products. For example, kaolinite and halloysite are the main clay minerals found in the

saprolite (e.g. in the core or root of the saprolite) of the study area. If the uppermost part of the mature saprolite would have remained uneroded, minerals indicative of more extreme weathering may have been present (i.e. gibbsite). This assessment is consistent with the identification of gibbsite in north Norway (Whalley et al., 1997), very close to Finnish Lapland.

5.4. *Paleoclimate and paleogeography*

The clay mineralogy provides a key to the weathering history of the region. Minerals in advanced stages of the weathering sequence, such as kaolinite, will persist unaltered throughout changes in climate and, therefore, can preserve the paleoclimatic record (Singer, 1980; Birkeland, 1984).

The dominant quartz and kaolin minerals in the saprolite of the Vuotso–Tankavaara areas yield information on the paleoclimatology and the paleogeography of the study area. The occurrence of kaolinite and halloysite indicates that a humid-warm climatic phase existed during the time of their formation.

Meyer (1976), Störr and Bellman (1975) and Pulvertaft (1979) reported the occurrence of a humid-warm climate in western Europe, East Germany and in Greenland during the early Cretaceous. The climatic conditions of the early Cretaceous could possibly explain the time of the formation of kaolinite and halloysite as well as the age of saprolite in the Vuotso–Tankavaara area. It should be noted that saprolites in southern Sweden have been interpreted as being of Jurassic–late Cretaceous age (Lidmar Bergström et al., 1997).

A kaolinitic weathering crust on the Iceland–Färö Ridge is overlain by Middle Eocene sediments (Nilsen and Kerr, 1978), and the same relationships are known from East Germany (Störr, 1975). Whalley et al. (1997) concluded that the age of the weathering products on the Blockfield, north Norway were probably Tertiary and could not have formed substantially at any stage during the Pleistocene which agrees with the findings of Basham (1974) and Hall and Mellor (1988).

Lidmar-Bergström (1982) concluded that European saprolite might be classified into two groups, namely an older one consisting of clays and a younger one comprising of sandy saprolites in accordance with conditions in the former Soviet Union (Kiselev, 1979). The older one formed during the Mesozoic and early Tertiary into the Middle Miocene. The younger one formed from the Late Miocene onwards.

From the above, it is clear that the clay minerals kaolinite and halloysite found in the saprolite of the Vuotso area indicate a climate that has been reported to exist during early Cretaceous, early Tertiary and Middle Miocene. To resolve this issue, O isotope data of the kaolinites could provide more precise climate data (Srodon, 1999).

6. Applied aspects

If we are going to understand anthropogenic climate change, we need to understand past natural variability in climate. This study provides some valuable information on natural variability in climate. Weathering systems are an essential clue to the understanding of earth surface processes. The geochemical analysis of the fossil saprolites discussed here indicates that Finland has undergone great climatic extremes over geologic time through a combination of the influence of plate tectonics and likely variations in the composition of gases like CO₂ in the atmosphere. It reinforces the fact that climate, whether anthropogenically influenced or not, is not immutable and can vary greatly over time.

The occurrence of considerable amounts of halloysite and kaolinite in Finnish lapland suggests the possibility of finding exploitable kaolin deposits. Finland is a big paper producer and its paper industry today must import kaolin from abroad. As in Korea, the kaolin deposits have been formed by residual weathering of anorthosite under the humid temperate climate and consist mostly of halloysite and kaolinite in various proportions (Jeong and Kim, 1992, 1993, 1994). Another economically interesting item is the possible lateritic Ni deposits, which may exist in the saprolite. Till geochemistry is a very important prospecting method in Finland. Till includes remnants from the ancient weathering crust and therefore knowledge of the geochemical and mineralogical development of the weathering crust is crucial in the interpretation of till geochemical results.

7. Conclusions

The present study shows that central Finnish Lapland underwent intense chemical weathering in the geologic past. The weathering is totally different to the present day weathering climate in the northern boreal zone. The pre-Quaternary saprolite is found only in central Lapland as it is situated on an ice divide zone. Elsewhere in Finland it is destroyed by strong glacial erosion. Global palaeoclimatic records show that tropical to subtropical climatic conditions never extended up to the present high latitude of Finnish Lapland (68°12N). This indicates that during the weathering, the palaeolatitudinal position of Finnish Lapland was at a much lower latitude than at present.

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