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Evidence for submarine weathering from metamorphosed weathering profiles on basaltic rocks, Tananao Metamorphic Complex, Taiwan¹

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Abstract

The metamorphosed paleo-weathering horizon(s) developed on basaltic rocks in the Tananao Metamorphic Complex forms distinct Al-Fe-rich rocks and occurs as two parallel belts, demonstrating its potential as a stratigraphic correlation unit in this structurally complicated metamorphic terrain.

These Al-Fe-rich rocks have been metamorphosed under amphibolite-/greenschist-facies conditions, leading to the formation of garnet-chloritoid rocks/chloritoid-rich rocks. The chemical changes along three profiles, from metabasite to garnet-chloritoid rock, show prominent K, Rb, Cs and Ba enrichment, yet, also, a large depletion of other elements if it is assumed that Ti, Zr, Nb, Ta and Hf remained relatively immobile. The REE's, however, behaved non-coherently. These chemical characteristics are attributed to pre-metamorphic processes and are shown to be similar to those observed with submarine weathering of basaltic rocks. In contrast, the chloritoid-rich rocks reveal distinct K, Rb, Cs and Ba depletion. It is suggested that this resulted from subaerial weathering processes.

K enrichment is not an uncommon feature in Paleozoic/Precambrian paleosols. The present case study demonstrates that submarine weathering should be considered as a probable cause.

1. Introduction

The study of paleosols of Quaternary age has made a significant contribution to stratigraphic and paleoenvironmental research (e.g., Birkeland et al., 1971; Campbell, 1986). Similar applications when extended to pre-Quaternary paleosols, however, are restricted due to the possibility of complicated geohistory (see

[SB] ¹ Institute of Earth Sciences Contribution No. IESEP94-014. Wright, 1986). For example, since soils often change their characteristicss along strike and more than one soil may be present in a stratigraphic sequence, their use as correlation units in a metamorphic terrain may be somewhat ambiguous (Barrientos and Selverstone, 1987). On the other hand, using Paleozoic and Precambrian paleosols as indicators of ancient atmospheric conditions (Gay and Grandstaff, 1980; Schau and Henderson, 1983; Holland, 1984; Retallack, 1986) is meaningful only when the alteration processes can be correctly identified (Hol-

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land and Feakes, 1989; Palmer et al., 1989a, b; Retallack, 1989; Rainbird et al., 1990). This paper presents chemical changes of the metamorphosed paleo-weathering profiles developed on basaltic rocks, probably of late Permian age in the Tananao Metamorphic Complex of Taiwan. The anomalous K enrichment along the profiles is as most likely a result of submarine weathering. It is postulated that such a process should be considered a probable cause for those Paleozoic and Precambrian paleosols with similar chemical variation trends. In addition, the spatial distribution of such metamorphosed weathering horizon(s) in the Tananao Metamorphic Complex suggests its potential as a stratigraphic indicator.

2. Geological settings and occurrences

The Tananao Metamorphic Complex is the oldest geologic-tectonic element outcropping in Taiwan. Schists, metabasite, marble and granitic gneiss are the major rock types, with the presence of subordinate amounts of metasandstone, metaconglomerate, metachert and ophiolitic rocks. Four mappable lithologic units were proposed (Wang Lee, 1982; Hsu, 1988). Unit I consists of a metabasite-metachert-marble association. A few deformed fossils of late Permian age were recovered from the marbles in this unit (Yen, 1953). It was shown that most metabasites in this unit exhibit chemical characteristics similar to those of within-plate basalt, and it was postulated that this unit may represent ancient ocean (marginal/back arc basin) floors/islands associated with chert and calcareous sediments (Yui et al., 1990a). Unit II is composed of pure and massive marble and was interpreted as reefy limestones (Yui et al., 1988, 1990b). Unit III contains phyllite, graphite schist, metasandstone and metaconglomerate, associated with minor blocks of metasandstone, metaconglomerate, metachert, marble and metabasite. Its mélange nature (Lu, 1986) suggests that it could represent a trench or near-trench deposit. Dinoflagellate fossils were reported recently (C.H. Chen, 1989), indicating that part of this unit may have ranged in age from the middle Jurassic to the early Cretaceous. The last unit, Unit IV, consists of granitic gneisses intruding into the above lithologic units at $\sim 97-85$ Ma (Jahn et al., 1986; Yui et al., 1993). The contact between rock types is generally concordant. The foliation of all these rocks generally strikes in the NE-SW direction and dips predominantly to the NW. However, due to the intensive deformation and polymetamorphism as well as the lack of diagnostic fossils, stratigraphic relationships among these rocks are still unclear. Tentative tectonic models for this metamorphic complex were discussed by Liou and Ernst (1984) and Yui et al. (1988, 1990b).

The chloritoid-rich rocks, occurring as a series of lenticular pods and lenses in the chloritoidbearing greenschist intercalated with the marble in Unit I, were first reported by Yen (1959). P.Y. Chen (1963) reported that the size of these lenticular bodies may range from <1 up to ~8 m in thickness and a few tens of meters in length. These lenticular bodies can be traced for $\sim 1 \text{ km}$ along the rock foliation. Based on drilling data, at least two successive layers of such rocks were noted in some places. Later, Liou and Chen (1978) gave petro-/mineral-chemical data and concluded that these chloritoid-rich rocks are Al-Fe-rich and were most probably derived from paleo-lateritic soils formed from a basaltic layer in a limestone terrain. The mineral assemblage, chloritoid + chlorite + calcite + opaques (mainly magnetite and ilmenite) \pm quartz \pm corundum \pm white mica \pm epidote \pm sphene \pm rutile, demonstrates that these paleosols were later metamorphosed under greenschist-facies metamorphic conditions (i.e. $350-450^{\circ}$ C, ~ 2 kbar) (Liou and Chen, 1978).

In their paper, Liou and Chen (1978) showed four sample localities for the chloritoid-rich rocks (Fig. 1, sample localities 1-4). In recent years with further field work, the authors of this study have found the chloritoid-rich rocks in another four localities (Fig. 1, sample localities 5-8). These newly-found occurrences are all intercalated with marble, similar to those reported by P.Y. Chen (1963) and Liou and Chen (1978). It is most surprising that all these chloritoid-rich



Fig. 1. Simplified geologic map of the study area (modified after Lan, 1989), showing the distribution of the lithologic units mentioned in the text and the sample localities. Sequential samples from the LI, P and LII profiles are from locality 8; sample 7999, from locality 7; sample 7925, from locality 6; sample 7966, from locality 5; sample 0070, from locality 4; sample 8430, from locality 3; sample 7523, from locality 2; and samples 1120 and 1121, from locality 1.

rock occurrences spatially form two belts, roughly parallel to the main trend of the rock foliation (Fig. 1). Although, due to the complicated structures of the area, the present study does not speculate on how these two belts are related to each other; such a spatial distribution, as well as the similar rock association, at least give a reasonable argument for trying to use these rocks as a stratigraphic indicator in the Tananao Metamorphic Complex. Similar recommendations concerning the paleosols in the Eastern Alps of Central Europe were also proposed by Barrientos and Selverstone (1987) and Krois et al. (1990).

Among the newly-found occurrences, the chloritoid-rich rocks at locality δ show two distinct aspects. First, they reveal a different mineral assemblage from others. At this locality, the mineral composition includes garnet, chloritoid, white mica, chlorite, plagioclase, quartz, epidote and ilmenite (hereafter referred to as the garnet-chloritoid rock), indicating amphibolite-facies metamorphic conditions. Secondly, this garnet-

chloritoid rock is associated with metabasite and the gradational changes from one to the other can be observed. The metabasite at this locality is ~ 8 m thick and can be divided into three units: the lower lava flow (LI) (~ 3.5 m), the middle pillow (P) (~ 1 m) and the upper lava flow (LII) $(\sim 3.5 \text{ m})$. The lower 3 m of each lava flow and the pillow core are typical metamorphosed amphibolite-facies basaltic rocks, containing hornblende, plagioclase, biotite, white mica and opaques. The garnet-chloritoid rocks developed on the upper 30-50 cm/outer 10-20 cm of the lava flows and the pillows, respectively, with the amount of garnet, chloritoid and white mica increasing upward/outward progressively. The grain size of the garnet reaches 3-5 mm and is easily recognized in the field. Sequential samples, from metabasite (sample numbers with suffix A) to garnet-chloritoid rock (sample numbers with suffix B, C, or D), were collected from three profiles of LI, P and LII to see their chemical changes. Sampling was focused on the garnet-chloritoid rocks, while sampling distances were arbitrary. For comparison, eight samples of chloritoid-rich rocks from other localities were also studied.

3. Analytical methods and results

All chemical analyses were performed at the

| Table | 1 |
|-------|---|
|-------|---|

| Chemical com | positions of t | hree profiles from | metabasite to garnet- | -chloritoid rock at | locality 8 |
|--------------|----------------|--------------------|-----------------------|---------------------|-------------|
| Chonnour com | | mee promes nom | monuousno to guinet | vinor i out u | , iocuity o |

| | LI-A | LI-B | LI-C | LI-D | PI-A | PI-B | PI-C | LII-A | LII-B | LII-C | LII-D |
|--|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|------------|------------|------------|
| (%): | | | | | | | | | | | |
| SiO ₂ | 45.48 | 39.16 | 35.40 | 28.01 | 47.54 | 39.17 | 31.99 | 45.83 | 34.82 | 33.29 | 23.83 |
| TiO ₂ | 1.66 | 2.86 | 4.41 | 6.76 | 1.66 | 2.05 | 3.76 | 1.65 | 3.76 | 4.60 | 5.42 |
| Al_2O_3 | 15.59 | 19.96 | 23.47 | 26.01 | 14.13 | 17.19 | 25.68 | 13.81 | 24.05 | 25.67 | 29.95 |
| FeO ^a | 11.88 | 16.15 | 20.18 | 23.13 | 11.06 | 17.97 | 18.85 | 12.22 | 20.13 | 19.23 | 19.91 |
| MnO | 0.18 | 0.21 | 0.28 | 0.09 | 0.18 | 0.22 | 0.15 | 0.22 | 0.25 | 0.21 | 0.15 |
| MgO | 9.79 | 9.61 | 4.61 | 2.65 | 10.49 | 8.06 | 4.85 | 12.73 | 6.18 | 3.99 | 4.94 |
| CaO | 11.26 | 5.39 | 1.68 | 1.32 | 11.01 | 8.78 | 2.58 | 8.16 | 1.12 | 2.95 | 3.40 |
| K ₂ O | 0.12 | 0.89 | 2.73 | 2.17 | 0.03 | 0.15 | 1.97 | 0.01 | 2.29 | 2.12 | 1.03 |
| Na ₂ O | 1.58 | 0.92 | 0.63 | 0.37 | 1.44 | 0.60 | 1.10 | 0.16 | 0.46 | 0.98 | 0.35 |
| P_2O_5 | 0.26 | 0.38 | 0.71 | 0.88 | 0.25 | 0.35 | 0.16 | 0.27 | 0.09 | 0.35 | 0.61 |
| LOI | 2.20 | 4.16 | 5.40 | 7.53 | 2.23 | 4.57 | 8.08 | 4.45 | 6.10 | 5.92 | 9.68 |
| Total | 100.00 | 99.69 | 99.50 | 98.92 | 100.02 | 99.11 | 99.17 | 99.51 | 99.27 | 99.31 | 99.27 |
| (ppm): | | | | | | | | | | | |
| Nb | 25 | 49 | 54 | 97 | 23 | 27 | 59 | 23 | 54 | 72 | 89 |
| Zr | 117 | 190 | 322 | 434 | 118 | 141 | 320 | 119 | 278 | 409 | 471 |
| Ŷ | 20 | 34 | 57 | 127 | 22 | 25 | 27 | 22 | 30 | 54 | 56 |
| Sr | 600 | 232 | 243 | 251 | 233 | 170 | 382 | 194 | 166 | 252 | 285 |
| Rb | 4 | 34 | 87 | 65 | 2 | 5 | 47 | 3 | 54 | 65 | 34 |
| Ва | 54 | 383 | 580 | 681 | 37 | 52 | 1,560 | 28 | 1,560 | 808 | 279 |
| Zn | 77 | 98 | 144 | 248 | 88 | 135 | 187 | 103 | 158 | 169 | 264 |
| Ni | 284 | 318 | 459 | 1,005 | 279 | 355 | 315 | 270 | 382 | 371 | 212 |
| Со | 61 | 65 | 96 | 149 | 59 | 72 | 86 | 63 | 87 | 89 | 75 |
| Cr | 529 | 976 | 1,081 | 1,206 | 522 | 735 | 750 | 496 | 1,340 | 948 | 1,001 |
| V | 277 | 331 | 629 | 889 | 253 | 343 | 453 | 265 | 427 | 590 | 545 |
| Та | 1.23 | 2.84 | 3.28 | 5.91 | 1.14 | 1.46 | 3.46 | 1.34 | 3.04 | 4.19 | 3.04 |
| Hf | 2.68 | 4.31 | 7.84 | 9.36 | 2.5 | 3.27 | 7.34 | 2.75 | 6.32 | 8.17 | 5.75 |
| Sc | 32.8 | 41.8 | 54.2 | 77.5 | 32.6 | 39.3 | 46.7 | 32.1 | 51.8 | 52.9 | 53.4 |
| Cs | 0.21 | 0.98 | 2.35 | 1.15 | n.d. | n.d. | 0.89 | n.d. | n.d. | 2.52 | n.d. |
| Th | 1.9 | 4.4 | 4.2 | 6.8 | 1.6 | 2.2 | 4.6 | 1.6 | 3.6 | 5.6 | 3.2 |
| La | 15.8 | 37.2 | 43.6 | 109.0 | 16.5 | 19.1 | 23.1 | 16.6 | 39.8 | 131.0 | 74.9 |
| Ce | 33.4 | 78.5 | 97.6 | 244.0 | 36.4 | 44.0 | 39.9 | 37.3 | 66.7 | 210.0 | 110.0 |
| Nd | 16.7 | 36.4 | 54.3 | 131.0 | 19.6 | 22.7 | 25.7 | 20.3 | 37.7 | 57.5 | 65.4 |
| Sm | 3.88 | 7.04 | 12.20 | 32.80 | 4.50 | 5.27 | 6.52 | 4.67 | 7.49 | 18.10 | 12.90 |
| Eu | 1.48 | 2.52 | 4.20 | 6.92 | 1.39 | 1.85 | 1.09 | 1.34 | 2.05 | 4.35 | 2.15 |
| Gd | 4.22 | 6.88 | 12.50 | 20.80 | 4.29 | 5.05 | 5.87 | 4.13 | 7.19 | 14.20 | 8.19 |
| Ib | 0.72 | 1.09 | 2.03 | 2.82 | 0.66 | 0.79 | 0.83 | 0.62 | 1.04 | 1.87 | 1.10 |
| Yb | 1.96 | 2.96 | 4.75 | 7.96 | 1.73 | 2.18 | 2.05 | 1.67 | 2.25 | 3.51 | 2.54 |
| Lu | 0.30 | 0.47 | 0.68 | 0.96 | 0.23 | 0.28 | 0.27 | 0.23 | 0.37 | 0.49 | 0.42 |
| (La/Yb) _N CIW ^b | 5.3 40 | 8.3 64 | 6.0 85 | 9.0 90 | 6.3 39 | 5.8 50 | 7.4 80 | 6.6 48 | 11.7 90 | 24.6 79 | 19.5 82 |

n.d. = not determined.

^a Total Fe as FeO.

^b CIW (weathering index) = $[Al_2O_3/(Al_2O_3 + CaO + Na_2O)] \times 100$ (molecular proportions) (Harnois, 1988).

 Table 2

 Chemical compositions of chloritoid-rich rocks from localities 1–7

| | 1120 | 7925 | 0070 | 8430 | 7523 | 7966 | 7999 | 1121 |
|--------------------------------|-------|-------|-------|-------|--------|--------|-------|-------|
| (%) | | | | | | | | |
| SiO ₂ | 31.92 | 29.01 | 27.11 | 26.59 | 22.49 | 17.66 | 16.15 | 12.57 |
| TiO ₂ | 5.05 | 4.73 | 4.31 | 5.43 | 5.63 | 4.08 | 5.03 | 6.57 |
| Al ₂ O ₃ | 28.10 | 31.14 | 32,91 | 30.98 | 35.16 | 45.49 | 43.30 | 42.33 |
| FeO ^a | 25.15 | 25.40 | 22,90 | 26.50 | 24.91 | 21.72 | 24,97 | 26.61 |
| MnO | 0.37 | 0.19 | 0.20 | 0.19 | 0.21 | 0.19 | 0.10 | 0.16 |
| MgO | 1.26 | 0.88 | 1.61 | 1.00 | 2.19 | 1.05 | 1.87 | 1.84 |
| CaO | 0.71 | 0.90 | 1.13 | 0.89 | 0.36 | 3.19 | 1,13 | 0.20 |
| K ₂ O | 0.15 | 0.01 | 0.32 | 0.00 | 0.14 | 0.00 | 0.00 | 0.00 |
| Na ₂ O | 0.00 | 0.00 | 1.45 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 |
| P_2O_5 | 0.36 | 0.67 | 0.32 | 0.69 | 0.19 | 0.79 | 0.56 | 0.19 |
| LOI | 6.18 | 6.62 | 6.83 | 7.34 | 7.73 | 6.01 | 6.57 | 8.74 |
| Total | 99.24 | 99.57 | 99.08 | 99.60 | 99.00 | 100.18 | 99.69 | 99.20 |
| (ppm) | | | | | | | | |
| Nb | 62 | 49 | 60 | 59 | 85 | 65 | 55 | 79 |
| Zr | 341 | 295 | 281 | 319 | 440 | 441 | 361 | 383 |
| Y | 29 | 18 | 42 | 21 | 21 | 18 | 10 | 10 |
| Sr | 90 | 32 | 499 | 31 | 39 | 236 | 36 | 2 |
| Rb | 6 | 3 | 7 | 3 | 4 | < 2 | 2 | < 2 |
| Ba | 59 | 27 | 190 | 6 | 81 | 45 | 20 | 6 |
| Zn | 238 | 160 | 174 | 170 | 216 | 130 | 166 | 175 |
| Ni | 356 | 298 | 428 | 352 | 267 | 78 | 158 | 41 |
| Co | 94 | 49 | 105 | 55 | 87 | 57 | 61 | 27 |
| Cr | 298 | 788 | 1,017 | 872 | 654 | 1,103 | 1,622 | 1,324 |
| V | 475 | 418 | 611 | 482 | 564 | 437 | 335 | 688 |
| Ta | 2.88 | 2.82 | 3.32 | 3.15 | 6.90 | 3.34 | 3.14 | 4.36 |
| Hf | 6.19 | 6.80 | 6.28 | 7.34 | 16.90 | 7.32 | 7.77 | 8.25 |
| Sc | 36.8 | 39.1 | 58.3 | 45.2 | 53.4 | 42.6 | 67.9 | 65.3 |
| Cs | n.d. | n.d. | n.d. | n.d. | < 0.20 | n.d. | n.d. | n.d. |
| Th | 3.0 | 2.9 | 2.7 | 3.1 | 5.6 | 3.5 | 3.8 | 3.7 |
| La | 44.9 | 14.9 | 36.0 | 39.1 | 40.5 | 62.0 | 47.7 | 60.7 |
| Ce | 103.0 | 30.4 | 78.7 | 80.7 | 85.7 | 128.0 | 80.9 | 124.0 |
| Nd | 58.6 | 18.4 | 44.7 | 63.1 | 47.1 | 64.0 | 44.2 | 56.9 |
| Sm | 14.80 | 6.84 | 9.54 | 16.80 | 10.70 | 13.70 | 6.21 | 11.10 |
| Eu | 1.63 | 2.53 | 2.35 | 3.45 | 4.61 | 1.86 | 0.77 | 1.95 |
| Gd | 6.73 | 8.72 | 8.95 | 7.45 | n.d. | 7.65 | 4.86 | 4.90 |
| Тb | 0.80 | 0.95 | 1.21 | 1.18 | 1.32 | 0.92 | 0.45 | 0.50 |
| Yb | 2.55 | 2.07 | 2.91 | 2.21 | 1.80 | 1.53 | 1.49 | 1.04 |
| Lu | 0.33 | 0.18 | 0.48 | 0.28 | 0.22 | 0.19 | 0.16 | 0.13 |
| (La/Yb) _N | 11.6 | 4.8 | 8.2 | 11.7 | 14.9 | 26.8 | 21.1 | 38.5 |
| CI₩° | 96 | 95 | 88 | 95 | 98 | 89 | 95 | 99 |

n.d. = not determined.

^a Total Fe as FeO.

^b CIW (weathering index) = $[Al_2O_3/(Al_2O_3 + CaO + Na_2O)] \times 100$ (molecular proportions) Harnois, 1988).

University of Western Ontario. Ten major and eleven trace elements were analyzed by X-ray fluorescence spectroscopy (XRF) following the methods suggested by Norrish and Hutton (1969) and Feather and Willis (1976). Spectral interferences were corrected from pre-calculated



Fig. 2. Al_2O_3 -FeO-SiO₂ diagram showing the compositions of the rocks in the present study. *Gar.*=garnet; *Ctd.*=chloritoid; *Cd.*=cordierite. Also shown for comparison is the compositional field of the chloritoid-rich rocks reported by Liou and Chen (1978). *Closed circles*=sequential samples from the LI profile; *open circles*=sequential samples from the P profile; *crosses*=sequential samples from the LII profile; *stars*=chloritoid-rich rocks. *Large arrow* shows the chemical variation trend from metabasite to garnet-chloritoid rock. See text for details.

interfering factors. In general, the precision was better than \pm 5% for all major-element determinations, and \pm 10% for most trace-element analyses. The REE's (rare-earth elements), Ta, Hf, Sc, Cs and Th, were analyzed by the instrumental neutron activation analysis (INAA) method as described by Gibson and Jagam (1980). Analysis of gamma-ray spectra was done by SAMPO[©] computer program (Routti, 1969). The reproducibility was better than \pm 5% for Sm, Eu, Ta, Sc and Cs, but \pm 10% for Ce, Gd, Tb, Yb, Lu, Hf and Th, and \pm 15% for La and Nd. The results for the sequential samples from the three profiles at locality 8 are shown in Table 1, while those for the chloritoid-rich rocks from other localities are presented in Table 2. In both tables, the total iron is expressed as FeO. Owing to the reducing environment of metamorphism, the iron is mainly present as ferrous ion allocated in chloritoid, almandine garnet and ilmenite in the garnet-chloritoid rocks, although magnetite was also reported in the chloritoid-rich rocks (Liou and Chen, 1978).

As shown in Table 1, the compositions of the sequential samples show gradational systematic changes, i.e. from metabasite to garnet-chloritoid rock, Ti, Al, Fe, K, and most of the trace elements increase progressively, whereas Si, Mg and Ca decrease significantly. In the SiO_2 -Al₂O₃-FeO diagram (Fig. 2), the Al₂O₃-FeO enrich-





ment trend is evident. In comparison, the chemical compositions of the chloritoid-rich rocks in the present study (Table 2) are similar to those reported by Liou and Chen (1978) (Fig. 2). In the SiO₂-Al₂O₃-FeO diagram, both the chloritoid-rich rock and the garnet-chloritoid rock are enriched in Al and Fe relative to the metabasite, but the former shows more prominent SiO₂ depletion (Fig. 2).

The chondrite-normalized REE patterns for these samples are presented in Fig. 3. The metabasites (samples LI-A, P-A and LII-A) are enriched in the light REE's (LREE's), similar to other metabasites in the Tananao Metamorphic Complex reported by Yui et al. (1990a). The garnet-chloritoid rocks are higher in the REE concentrations, more enriched in the LREE's and sometimes exhibit negative Eu anomaly. The chloritoid-rich rocks display similar REE characteristics, except that they do also show positive Eu anomaly occasionally (Fig. 3d-f).

Even though the rocks in the present study were metamorphosed under greenschist-/amphibolite-facies conditions, the chemical compositions, except the LOI content, of the metabasites are still similar to those of the present-day unaltered basalts (BVSP, 1981). The compositions of the metabasites are also rather homogeneous (Table 1). It is, therefore, assumed that the metamorphism was mainly an isochemical process with respect to the non-volatile components.

4. Discussion

Chloritoid is relatively uncommon in low- to medium-grade metamorphic rocks and is restricted to special bulk-rock compositions (Halferdahl, 1961; Hoschek, 1967). Halferdahl (1961) gave a very detailed compilation about the occurrence of the chloritoid-bearing rocks. Based on his work, chloritoid may occur in hornfelsed pelitic schists, in emery deposits, in hydrothermally altered rocks/hydrothermal veins and in regionally metamorphosed pelitic sediments, particularly those rich in Al and Fe, but poor in Ca, Mg, K and Na. In low-grade metamorphic rocks, chloritoid is usually accompanied by quartz, chlorite, muscovite, corundum, diaspore, calcite, rutile and/or iron oxide. In higher-grade rocks, almandine, staurolite and kyanite are common associates. The protoliths of these rocks could be (altered) basalt, (altered) shale and residual deposits (or their sedimentary equivalents) (e.g., Halferdahl, 1961; Hoschek, 1967; Iwao, 1978; La Tour et al., 1980; Phillips, 1988; Palmer et al., 1989a). Since the protoliths cover a wide variety of rocks, the chemical compositions of the chloritoid-bearing rocks vary considerably. For example, their silica content may vary from nearly zero to ~ 90 wt% (Halferdahl, 1961). Despite such component variations, some characteristic chemical features do exist for the chloritoid-bearing rocks. e.g., more alumina than the total mafic oxides; an excess of alumina after calculating K₂O, Na₂O and CaO as micas, plagioclase or epidote; more FeO+MnO than MgO; and more FeO+MnO than Fe₂O₃ (Halferdahl, 1961). Hoschek (1967) also concluded that to contain chloritoid, rocks should simultaneously meet the following chemical criteria: low abundances of alkalis and CaO: $(FeO^* + MgO) / (FeO^* + MgO + Al_2O_3) < 0.63$ and $FeO^*/(FeO^*+Al_2O_3) < 0.58$. The garnetchloritoid and chloritoid-rich rocks in the present study are relatively low in silica content (Tables 1 and 2), compared to the normal shales and basalts. Most samples do have low alkali and CaO contents and show chemical characteristics compatible with those documented by Halferdahl (1961) and within the chemical limitations postulated by Hoschek (1967). Some samples (e.g., samples LI-B and PI-B in Table 1), however, exhibit relatively high Fe-Mg contents that do not concur with Hoschek's chemical limitations. In studying the chloritoid-bearing metabasite, La Tour et al. (1980) reported bulk-rock chemical compositions with very high Fe content and suggested that the chemical limitations proposed by Hoschek (1967) may not be valid since the presence of accessory minerals, such as magnetite and ilmenite, could accommodate excess Fe.

The similar high Al and Fe contents in the garnet-chloritoid and chloritoid-rich rocks in the present study are significant. These rocks with similar chemical characteristics but with different metamorphic grades suggest that their distinct bulk compositions were largely developed either prior to metamorphism or in the early stages of their metamorphic history. This is consistent with the suggestion made by Liou and Chen (1978) that these Al-Fe-rich rocks could be of lateritic origin derived from a basaltic layer. Therefore, the observed gradational chemical changes along the three profiles at locality 8 may have most likely resulted from weathering. However, Schreyer et al. (1981), Phillips (1988) and Palmer et al. (1989a) argued that hydrothermal alteration, either caused by pre-metamorphic postvolcanic exhalative processes or by synmetamorphic hydrothermal processes, could also produce rocks with high Al content in metamorphic terrains. Although it is difficult to distinguish the pedogenic vs. hydrothermal origins for the Al-Fe-rich rocks based solely on the chemical data (e.g., Retallack, 1989), the latter proposition is discounted in the present study since any hydrothermal alteration should not restrict its effects only to the garnet-chloritoid/chloritoid-rich rocks or to the top of the lava flows. The regional distribution of the Al-Fe-rich rocks in the present study as two parallel belts mentioned above does not favor a hydrothermal origin either. Although the granitic gneiss intruded the marble-metabasite rock association in the study area, the present contact is largely tectonic. No contact metamorphism or hydrothermal alteration is observed around the granitic gneiss.

The composition-volume changes of weathering profiles could be quantitatively analyzed by a series of equations as proposed by Gresens (1967). However, a simple "isocon diagram" as a solution to Gresens' equations was provided by Grant (1986). Fig. 4, then, employs Grant's method for sequential samples that were collected from the lower lava flow (LI) unit. The metabasite sample, LI-A, in this profile is assumed to be the least altered and its composition may closely represent that of the igneous precur-

Fig. 4. Isocon diagram for samples from the LI profile. *Solid line*=constant Ti and Zr; *long-dashed line*=constant mass; *short-dashed line*=constant volume. See text for more details.



sor. The composition of each element, multiplied by a proper scaling factor, was plotted in the figure for successive samples. As pointed out by Grant (1986), elements that remain immobile should fall on a single straight line passing through the origin. Such a line is called an "isocon". Elements that were truly immobile throughout the profile should define isocons in each diagram in Fig. 4, although the slope of the isocon may change from sample to sample. Careful inspection of Fig. 4 shows that only Ti and Zr can be closely fitted to isocons throughout the profile. This result is consistent with the conventional knowledge that these two elements are generally considered to be immobile during weathering/metamorphism. Elements which fall to the right/left of the isocon represent losses/ gains, respectively, relative to Ti and Zr. Note that although the chemical compositions of the sequential samples show apparent progressive Al and Fe enrichment in Table 1, these two elements are actually leached (Fig. 4). Only K, Rb, Cs and Ba were consistently enriched throughout. The densities of samples LI-A, -B, -C and -D are 3.132, 3.168, 3.172 and 3.397 g cm⁻³, respectively. Calculations yield integrated (including weathering and metamorphic effects) mass losses of 38%, 64% and 73%, and volume decreases of 39%, 65% and 75% for samples LI-B, -C and -D, respectively.

Similar features also hold for the pillow lava profile and the upper lava flow profile at locality δ , although the relevant isocon diagrams are not shown. To give a clearer and more quantitative idea, the percent change of each element (X) relative to the least altered rock vs. the degree of weathering along the profiles is shown in Figs. 5 and 6 for major and trace elements, respectively. The percent change is calculated assuming Ti remains constant:

$$(\text{percent change}) = (1)$$

$$\left(\frac{(X/Ti)_{\text{sample}}}{(X/Ti)_{\text{least altered rock}}} - 1\right) \times 100$$

The degree of weathering is measured by the weathering index (CIW) as defined by Harnois (1988):

$$CIW = [Al_2O_3/(Al_2O_3 + CaO + Na_2O)]$$

 $\times 100$ (molecular proportions) (2)

It is noted that the upper lava flow is overlain by marble. Field observations show that the garnet-chloritoid rocks developed on this unit might have been contaminated by some carbonate clasts. This is manifested by the progressive increase in CaO content (or epidote content in mineralogy) for the garnet-chloritoid rocks (LII-B, -C and -D in Table 1). The calculated CIW for this profile, therefore, may be a little misleading.

As shown in Fig. 5, Si, Mn, Mg, Ca, P, and to a lesser extent, Al and Fe, are progressively depleted as CIW increases in all three profiles. Na behaves a little differently in the LII profile, probably an apparent feature due to the very low Na content in sample LII-A (Table 1). K, however, is significantly enriched. Trace elements Nb, Ta and Hf, in addition to Zr, also remain relatively constant, but (V,Cr) < Zn < (Sc,Co) < Ni < Sr (in this order of increasing mobility) decrease successively. Y and Th either increase or decrease, while Rb, Cs and Ba show prominent increases (Fig. 6).

The chloritoid-rich rocks from localities 1-7occur as thin pods or lenses in the well-foliated chloritoid-bearing greenschist. Compared with the metabasites, the chemical compositions of the chloritoid-bearing greenschist show different, albeit small degrees of Al-Fe-Ti enrichment (Liou and Chen, 1978). Gradational changes from parental rocks to chloritoid-rich rocks, like those in locality 8, cannot be observed. Similar data comparisons to those discussed for the three profiles from locality 8 are not possible. However, if the parent rocks (i.e. the metabasites) in the study area all exhibit similar chemical characteristics, as documented by Yui et al. (1990a), the largest contrasting feature between the chloritoid-rich rocks and the garnet-chloritoid rocks would be that the former show significantly lower abundances of K, Rb, Cs and Ba for rocks with a comparable weathering index (Tables 1 and 2).

The element mobility during weathering is a function of many variables, such as the stability of the primary minerals, the chemical composi-



Fig. 5. Percent change, normalized with respect to Ti, of major elements vs. the weathering index (CIW). Note the vertical scales in (g) and (h). *Closed circles*=sequential samples from the LI profile; *open circles*=sequential samples from the P profile; *crosses*=sequential samples from the LII profile.

tion and the surface properties of the secondary minerals, the compositions of the fluid phase, the topography as well as the local Eh-pH environment (e.g., Fritz and Ragland, 1980; Nesbitt et al., 1980; Cramer and Nesbitt, 1983; Fritz and Mohr, 1984; Middelburg et al., 1988). Despite these complexities, studies on the weathering of basalts under subaerial conditions generally show distinct K, Rb, Cs and Ba depletion (e.g., Furnes, 1978; Chesworth et al., 1981; Colman, 1982; Noack et al., 1990; Marsh, 1991), similar to the weathering results of other rock types (e.g., Goldlich, 1938; Nesbitt et al., 1980; Minarik et al., 1983; Kronberg et al., 1987; Middelburg et al., 1988; Nesbitt and Young, 1989). The chemical characteristics of the chloritoid-rich rocks are, thus, consistent with the postulate that they are of lateritic origin (Liou and Chen, 1978). The K, Rb, Cs and Ba enrichment for the garnetchloritoid rocks, on the other hand, requires an alternative explanation.

The anomalous K enrichment in weathering profiles is not uncommon. It was reported for the paleo-weathering profiles of Paleozoic age (Wahlstrom, 1948; Retallack, 1985; Krois et al., 1990) as well as those of Precambrian age (Wil-



Fig. 6. Percent change, normalized with respect to Ti, of trace elements vs. the weathering index (CIW). Note the vertical scales in (n)-(p). *Closed circles* = sequential samples from the LI profile; *open circles* = sequential samples from the P profile; *crosses* = sequential samples from the LII profile.

liams, 1968; Button, 1979; Gay and Grandstaff, 1980; Sokolov and Hieskanen, 1984; Palmer et al., 1989a; Holland et al., 1990; Rainbird et al., 1990; Sutton and Maynard, 1991). Retallack (1986) postulated that such an anomalous K enrichment most probably originated from burial diagenesis of the weathering horizon. Based on kinetic, thermodynamic and mass-balance considerations, Nesbitt and Young (1989) concluded that the chemical changes of the weathering profiles are predictable, and they also favored the suggestion that diagenesis in continental environments may account for such K enrichment in some paleo-weathering horizons, although external K sources are needed. Such external K was usually attributed to diagenetic fluid which had equilibrated with K-feldspar in the overlying clastic sediments and percolated downward (e.g., Zbinden et al., 1988; Retallack, 1989). However, this postulate may not be applicable in the present case. The rocks which overlie the present weathering profiles are marbles, rather than metamorphosed clastic rocks. In fact, the former do not contain any K minerals. From the mass-balance point of view, if the upper 30 cm of the LI profile in this study have an averaged K₂O content of 2.45% with a volume decrease of 70% and if the K content of the groundwater in the basaltic rock and limestone area is assumed to be $\sim 1-7$ ppm (White et al., 1963), then rough calculations would result in an unreasonable integrated water/rock mass ratio of $\sim 2,000-20,000$ during diagenesis to account for the observed K enrichment. This ratio could be reduced if the diagenetic fluid had an abnormal K content. However, since both the garnet-chloritoid rocks and the chloritoid-rich rocks in the study area might have experienced a similar post-weathering geohistory, it seems implausible to assume that only the garnet-chloritoid rocks suffered from such K enrichment during diagenetic reactions. Although the garnetchloritoid rocks were subjected to slightly higher metamorphic P-T conditions than the chloritoid-rich rocks (i.e. the amphibolite facies vs. the greenschist facies), this difference should not be responsible for such a chemical contrast either.

The association with marble as well as the pil-

low structure of the metabasite at locality 8 demonstrate that the basalt was probably emplaced in a shallow submarine environment. Low-temperature seawater-basalt interaction (i.e. submarine weathering) causes complicated chemichanges of basalts under variable cal environments (Honnorez, 1981 and references therein). In spite of the complexities, such reactions almost invariably cause significant K, Rb and Cs enrichment in weathered basalts. Ba enrichment is not uncommon either. Altered basalts from Hole 417A [International Phase of Ocean Drilling (IPOD) Legs 50, 51 and 52] at the southern end of the Bermuda Rise in the North Atlantic even exhibit an apparent K_2O content of up to 6% (Donnelly et al., 1979), which was suggested to be the result of 10-20-Ma seawater-basalt interaction. Bienvenu et al. (1990) likewise reported one altered basalt sample which, having been exposed on the seafloor for ~ 35 Ma, showed significant apparent depletion in Si, Mg and Ca, and enrichment in Fe, Ti, K, Rb and Cs. These chemical changes are, in fact, similar to the chemical characteristics of the profiles discussed previously in this paper. Though the tectonic setting may not be the same, the above examples and the field occurrence of the garnet-chloritoid rocks, i.e. on top of the lava flows and around the pillows, do indeed show that a long-term submarine weathering may have been the cause for most, if not all, of the chemical changes observed along the three profiles at locality 8 of this study.

The chloritoid-rich rocks at localities 1-7 might also have experienced similar submarine weathering processes. If so, the submarine-weathering chemical signatures must have largely been erased by later subaerial processes, which could also explain why the protoliths of the chloritoid-rich rocks are more mature (i.e. more Al-Fe-enriched and Si-depleted, see Fig. 2) than those of the garnet-chloritoid rocks.

Most of the paleosols of Paleozoic/Precambrian age with anomalous K enrichment were either developed on basaltic rocks/greenstones or covered by marine clastic sediments (e.g., Button, 1979; Holland et al., 1990; Krois et al., 1990; Sutton and Maynard, 1991). Like the present study, the submarine-weathering process should be considered/explored as one of the possible causes responsible, or partially responsible, for the K enrichment. This mechanism overcomes the cumbersome mass-balance problem which results when considering the external K sources required in the continental diagenetic model (Nesbitt and Young, 1989).

The percent change of the REE's vs. atomic number for the three profiles from locality δ are shown in Fig. 7a-c. Combined with the chondrite-normalized pattern (Fig. 3), the following features are most distinct: (1) the LREE's were enriched, whereas the heavy REE's (HREE's) were leached from the two lava-flow profiles; (2) the REE's were all leached from the pillow profile; (3) preferential leaching as well as accumulation of the Eu were observed; and (4) a marked negative Ce anomaly in the chondrite-normalized pattern, a common feature for weathered submarine basalts, was not observed in the present study.

Since the REE systematics are generally not thought to be affected by metamorphism (e.g., Bau, 1991), the above features are ascribed to low-temperature seawater-basalt interactions. The mobility of the REE's during submarine weathering of basalts depends on the environment, in particular the water/rock ratio and the duration of interaction time. Both the uptake of the LREE's and the leaching of all REE's were reported previously (Frey et al., 1974; Ludden and Thompson, 1978; Staudigel et al., 1979; Staudigel and Hart, 1983). The uptake of the LREE's was attributed to a high water/rock ratio, long exposure time, and, hence, equilibration with seawater. Under the opposite conditions, all REE's were leached. Equilibration with seawater was also invoked to explain the negative Ce anomaly observed in some weathered submarine basalts (Frey et al., 1974; Robertson and Fleet, 1976; Ludden and Thompson, 1979; Humphris, 1984). However, if seawater, e.g., shallow seawater, does not exhibit a negative Ce anomaly (Shimizu and Masuda, 1977), then neither would the weathered basalts. The preferential removal of HREE's from the basalts is not uncommon and might be due to the fact that



Fig. 7. Percent change, normalized with respect to Ti, of REE's vs. atomic number: (a) sequential samples from the LI profile; (b) sequential samples from the P profile; (c) sequential samples from the LII profile; and (d) representative samples for the chloritoid-rich rocks.

they may have formed more soluble ionic complexes than the LREE's in seawater. This relative mobility, nevertheless, is also a function of pH (Cantrell and Byrne, 1987; Brookins, 1989). The observed REE characteristics of the three profiles, thus, demonstrate heterogeneous weathering environments.

Both preferential leaching and accumulation of Eu were observed in the profiles. Since under weathering conditions, Eu should be dominantly in the trivalent state in aqueous fluids and not be fractionated from other REE's (Sverjensky, 1984; Brookins, 1989), the observed anomalous Eu behavior can only be accounted for by mineralogical controls. Significant alteration of plagioclase may be the reason for the preferential leaching of Eu in more severely weathered samples (Fig. 7a-c). In the pillow-lava profile, although the REE's were mainly leached, the two altered samples show almost mirror-imaged REE percent change patterns, especially Eu (Fig. 7b). Due to changes in pH as the weathering fluid percolated through the pillow lava, partial precipitation/adsorption of the REE's, leached from the more weathered pillow rind (sample PI-C) on the less weathered portion (sample PI-B), may explain this observation. A similar phenomenon was reported by Nesbitt (1979) from a weathering profile developed on Australian granodiorite.

Assuming the protoliths of the chloritoid-rich rocks have similar REE compositions to those of the metabasites from locality δ , representative percent change of REE plots are shown in Fig. 7d. Similar features to those of the garnet-chloritoid rocks are also observed, except that only one sample (sample 7966) shows LREE enrichment. This may be accounted for by the superimposed effect of the inferred subaerial processes.

5. Conclusions

The metamorphosed Al-Fe-rich rocks in the Tananao Metamorphic Complex of Taiwan may be indicative of paleo-weathering horizon(s) developed on basaltic rocks. These rocks form two belts parallel to the main foliation trend of the complex. Such spatial distribution demonstrates the possible potential of these rocks to serve as a stratigraphic indicator in this structurally complicated metamorphic terrain.

These Al-Fe-rich rocks have been metamorphosed under different conditions, leading to different mineral assemblages: the garnet-chloritoid rocks of the amphibolite facies (locality 8) and the chloritoid-rich rocks of the greenschist facies (localities 1-7). The chemical characteristics, as well as the field occurrences of the chloritoid-rich rocks, indicate a lateritic origin. In contrast, three profiles from metabasite to garnet-chloritoid rock, developed either on lava flows or around pillows, show distinct K, Rb, Cs and Ba enrichment. Ti, Zr, Nb, Ta and Hf remained relatively immobile, while Si, Mn, Mg, Ca, P, Al, Fe, Sr, Ni, Co, Sc, Zn, Cr and V were all depleted from the garnet-chloritoid rock. It is concluded that all these chemical changes might not have been caused by metamorphism/hydrothermal alteration/diagenesis, but may have largely resulted from submarine weathering. The non-coherent behavior of the REE's also demonstrates heterogeneous submarine weathering environments. The chloritoid-rich rocks might have experienced similar submarine weathering processes but must have later been superimposed by subaerial processes, leading to K, Rb, Cs and Ba depletion and more intensive Al-Fe enrichment relative to Si.

K enrichment is not an unusual feature, especially for those paleosols of Paleozoic/Precambrian age. It is recommended, in addition to the diagenesis in continental environments, that submarine weathering also is to be considered a probable cause.

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