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Autor: Verdoya, Massimo / Chiozzi, Paolo / Pasquale, Vincenzo

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Heat-producing radionuclides in metamorphic rocks of the Briançonnais-Piedmont Zone (Maritime Alps)

MASSIMO VERDOYA[1], PAOLO CHIOZZI[1] & VINCENZO PASQUALE[1]

Key words: Radioactive elements, field γ spectrometry, heat production rate, background radiation, NW Italy

ABSTRACT

A wide set of radioactive heat-production data of gneisses, migmatites, amphibolites, and micaschists is presented for the eastern part of the Briançonnais-Piedmont Zone (NW Italy). They were obtained from determination of concentrations of uranium, thorium and potassium by means of ground γ-ray spectrometry. We selected as a reference site for the local background evaluation an area with serpentinitic outcrops showing negligible radioactivity, and verified the possible variation of the background radiation due to changes in elevation. This allowed an assessment of more realistic radioelement concentrations. Migmatites show the largest concentrations of U (6.1 ppm), whereas Th is more abundant in orthogneisses (17.8 ppm) as well as K (4.62%). Although rocks were affected by metamorphic and alteration processes, the Th/U and K/U ratios showed differences in radioelement concentrations that mostly reflect the chemical composition of protolith. In particular, in gneisess the Th/U ratio decreases exponentially with the uranium content. Due to the low content in heat-producing radioelements, amphibolites show an average heat-production rate of 0.30 μW m^{-3}. The other lithotypes have comparable U, Th and K contents, and therefore the range of their heat production is rather narrow, varying from 2.29 (paragneiss) to 2.99 μW m^{-3} (migmatite).

RIASSUNTO

Vengono presentati dati di produzione di calore radiogenico di alcune rocce (gneiss, migmatiti, anfiboliti, micascisti) della parte orientale della zona Brianzonese-Piemontese delle Alpi Marittime. Essi sono stati ottenuti da determinazioni di concentrazione di uranio, torio e potassio *in situ* mediante spettrometria a raggi gamma. In un sito di riferimento, su affioramenti serpentinitici caratterizzati da radioattività naturale trascurabile, è stata valutata la radiazione di fondo e verificata la sua variazione con l'altitudine, per una precisa stima della concentrazione dei radioelementi. Le migmatiti mostrano la concentrazione più elevata di uranio, mentre il torio e il potassio sono più abbondanti negli ortogneiss. I rapporti Th/U e K/U presentano differenze in concentrazione di radioelementi, che riflettono soprattutto la composizione chimica dei protoliti e non i processi metamorfici e di alterazione cui sono stati soggetti. In particolare, gli gneiss hanno un rapporto Th/U che diminuisce esponenzialmente con il contenuto in uranio. Le anfiboliti mostrano il più basso contenuto di elementi produttori di calore. Tutti gli altri litotipi hanno contenuti di U, Th e K confrontabili e un intervallo ristretto di variazione di produzione di calore.

Introduction

The major naturally occurring radionuclides (^{40}K, ^{235}U, ^{238}U and ^{232}Th) and their decay products are of significant importance to various geological problems. Applications span from geochronology and geochemistry to studies of ore deposits, energy potential and environmental radiation monitoring (e.g. Lentz 1994; Bea 1996; Chiozzi et al. 1998a). They contribute to the earth's thermal budget and therefore are of special interest in understanding and interpreting the terrestrial heat-flux data.

Surface exposures of rocks forming the crust and mantle represent a possible source of information for estimating the distribution of the heat-producing elements (HPEs) and thus for the variation with depth of the radiogenic heat production (RHP) rate (e.g. Nicolaysen et al. 1981; Ashwall et al. 1987; Jaupart et al. 1998; Verdoya et al. 1998). The RHP rate is a fundamental thermophysical property for the estimation of temperatures and thermal regime of the lithosphere (e.g. Pasquale et al. 1990 and 2001; Okaya et al. 1996).

In this paper, we present results from an extensive research program on the thermal properties of rocks. We focus on metamorphic lithotypes of the eastern part of the Briançonnais-Piedmont Zone (Maritime Alps, NW Italy). A field γ-ray spectrometry equipment was utilised for quantitative assessment of K, U and Th. As the investigated area shows a rather rough topography, particular care is given to the evaluation of the background radiation that can cause uncertainty in the HPE determination. Results of HPE concentrations are discussed in relation to parent rock composition, metamorphic and alteration processes, and their relative contribution to the RHP rate is analysed.

[1] Dipartimento per lo Studio del Territorio e delle sue Risorse, Settore di Geofisica, Università di Genova, Viale Benedetto XV, 5, I-16132 Genova, Italy.
E-mail address: verdoya@dipteris.unige.it

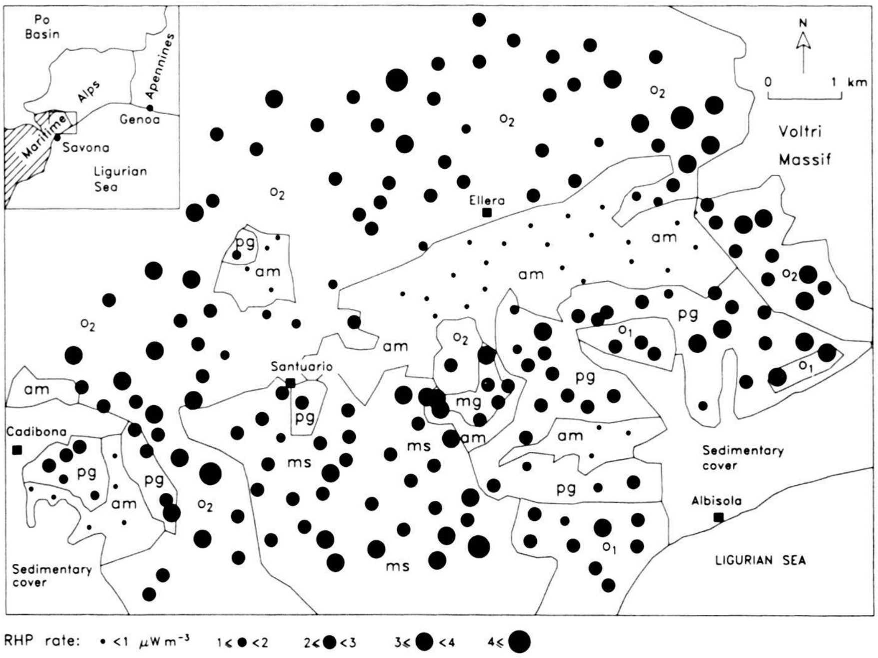

Fig. 1. Schematic lithological map of the eastern part of the Briançonnais-Piedmont Zone (Maritime Alps, NW Italy) and sites at which the radiogenic heat production (RHP) rate was determined. Pre-Carboniferous basement: o_1 – orthogneiss I, o_2 – orthogneiss II, pg - paragneiss, mg - migmatite, am - amphibolite. Permo-Carboniferous cover: ms - micaschist. Inset shows the main tectonic units of the Maritime Alps: Briançonnais-Piedmont Zone (hatched) and Voltri Massif (dotted).

Lithological setting

Orthogneisses, paragneisses, migmatites, and amphibolites are the main lithotypes constituting the Briançonnais-Piedmont Zone in the surveyed area (Fig. 1). These rocks, forming the pre-Carboniferous crystalline basement, are exposed as allochthonous masses. They overlie the Permo-Carboniferous metasedimentary cover, which consists of micaschists cropping out in a tectonic window (Vanossi et al. 1994).

Two types of orthogneiss (I and II) occur in the study area. Orthogneisses I derive from felsic rocks, essentially intrusive (granitoids) and subordinately effusive (rhyolites), and differ from orthogneisses II for compositional variety, prevalently monzogranitic and granodioritic. Paragneisses are related to pre-Alpine metamorphism of metapelites and metagreywackes, and are characterised by abundance of quartz, oligoclase and micas. Migmatites derive probably from anatexis of metapelites at the transition from eclogite to granulite facies.

Amphibolites, whose major constituents are hornblende and plagioclase, and are sometimes garnet or seldom quartz bearing, probably derive from basalts with tholeiitic affinity. These rocks form both massif and lenticular, stratified bodies, trending parallel to gneissic masses. Micaschists stem from continental fine-grained sediment that underwent Alpine metamorphism (Cortesogno et al. 1993, 1997).

Method and results

We have recently implemented a portable γ-ray device for in situ determinations of the main HPEs (Chiozzi et al. 2000). Our apparatus consists of a 76.2 mm x 76.2 mm thallium-activated sodium iodide [NaI(Tl)] scintillation detector and a 256-channel spectrometer unit. The detector is enclosed in a single integral unit with a photomultiplier tube, a high-voltage supply and a signal preamplifier. A reference isotopic source of ^{137}Cs, with an approximated activity of 15 kBq, is used to control automatically the system gain and to stabilise the spectrum by preventing shifts caused by temperature effect or component ageing.

For the quantitative analysis of HPEs, three energy windows were investigated, by recording γ-rays associated with characteristic peaks in the decay spectra of ^{40}K, ^{238}U and ^{232}Th (Chiozzi et al. 1998b). Most of natural uranium is formed by ^{238}U (99.28%) and ^{235}U (0.71%). Natural thorium is, instead, totally constituted by ^{232}Th. The isotopic abundance of ^{40}K, in weight, is 0.01193%. The determinations of uranium and thorium were based on measurements of γ radiation from the decay of ^{214}Bi (1.76 MeV) in the ^{238}U decay series and from ^{208}Tl (2.62 MeV) in the ^{232}Th series. The primary decay of potassium ^{40}K (1.46 MeV) was measured directly. The total count energy window was set from 120 to 3000 keV.

The spectrometer was calibrated by means of standard spectra acquired at three concrete pads enriched in K, U and Th (e.g. Killeen 1978). These sources were cylindrical in shape, 2 m in diameter and 0.5 m thick. A fourth calibration pad, made by lead, was used to measure the preliminary background count rate. Based on counting statistics, in common rocks the uncertainty on the measured concentration is minimum for potassium (< 3%), but is maximum for uranium (up to 8%). For the adopted measuring time of 500 s, the detection limit is estimated to be 0.2 ppm for uranium, 0.3 ppm for thorium and 0.03% for potassium (Chiozzi et al. 2000).

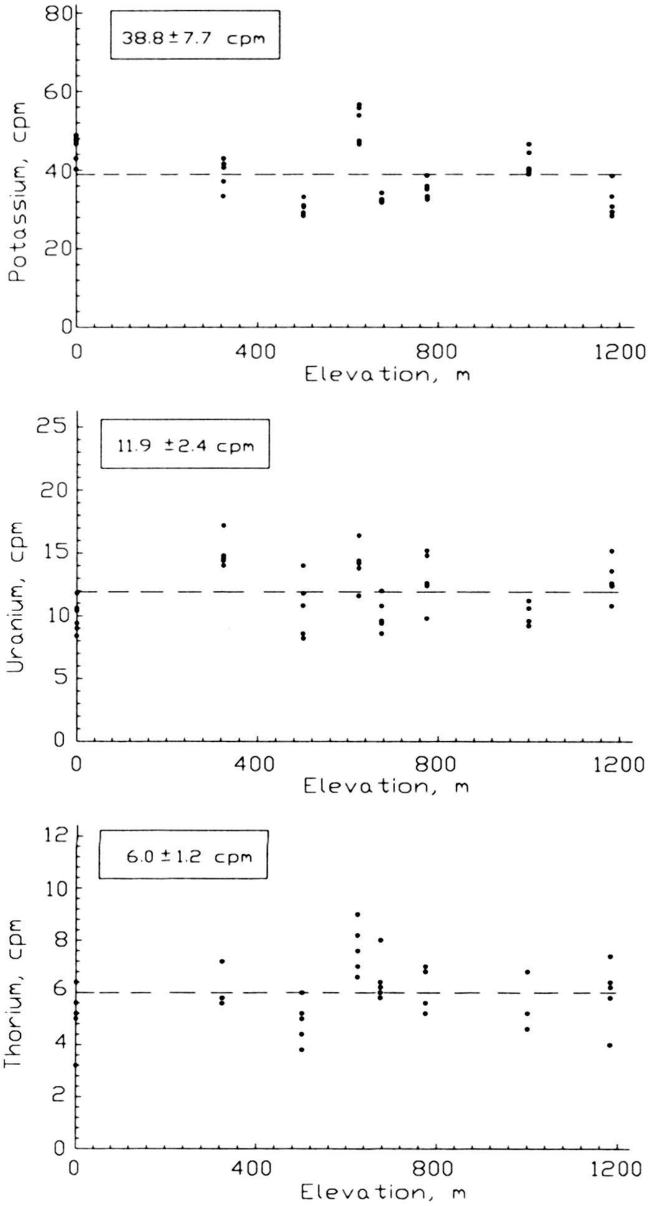

Fig. 2. Count rate in counts per minute (cpm) of the background γ radiation in the potassium, uranium and thorium regions of interest measured on sea and at different elevations over serpentinitic outcrops of the Voltri Massif. Average values are also shown together with standard deviations.

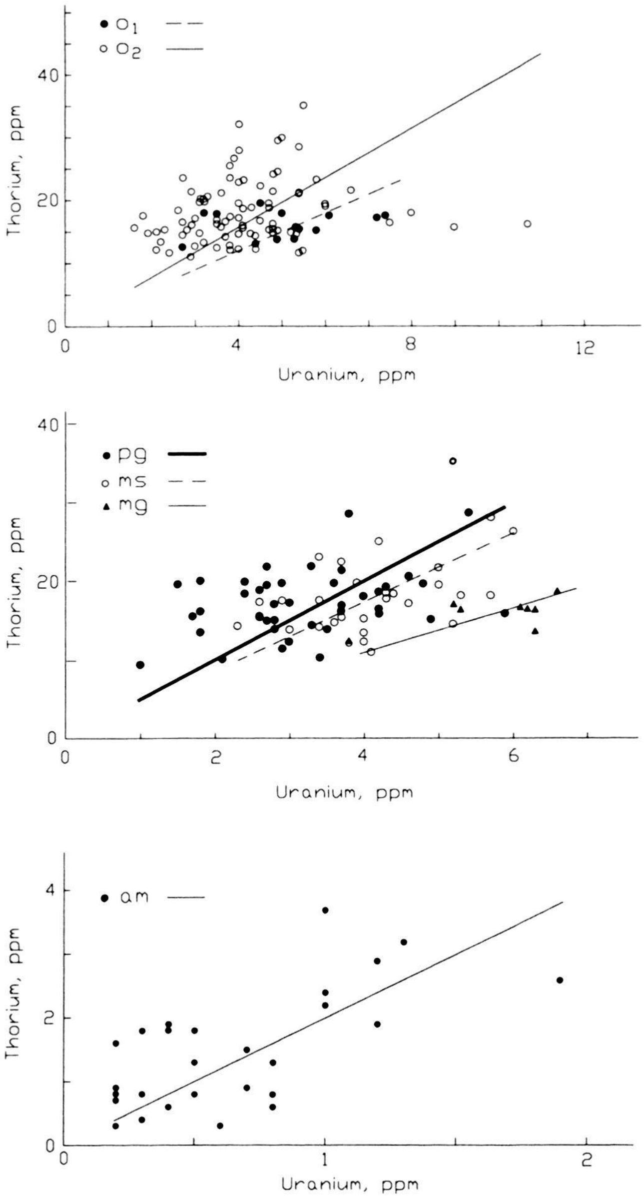

Fig. 3. Thorium versus uranium concentrations in the analysed rocks. The best-fitting linear relation is plotted. The rock code is as in Fig.1.

The concentrations c_K, c_U and c_{Th} at each site were deduced by solving the matrix

$$(1)\quad \begin{vmatrix} c_K \\ c_U \\ c_{Th} \end{vmatrix} = \begin{vmatrix} k_{11} & k_{12} & k_{13} \\ k_{21} & k_{22} & k_{23} \\ k_{31} & k_{32} & k_{33} \end{vmatrix} \begin{vmatrix} N_1 - \delta_1 \\ N_2 - \delta_2 \\ N_3 - \delta_3 \end{vmatrix}$$

where $k_{11},...,k_{33}$ are the spectrometer calibration constants, and N_1, N_2 and N_3 are the count rates for the regions of interest (ROIs) of the γ spectrum, centred on the ^{40}K, ^{214}Bi, and ^{208}Tl peaks, and δ_1, δ_2 and δ_3 are the background count rates in the same ROIs. Before starting the field radiometric survey, the local background was assessed in a nearby area (Voltri Massif, Fig. 1) over serpentinitic rocks that exhibit extremely low radioactivity, since their amount of K, U and Th is nearly negligible. A series of laboratory analyses on a few samples from this area indicates concentrations below the detection limit of the portable apparatus.

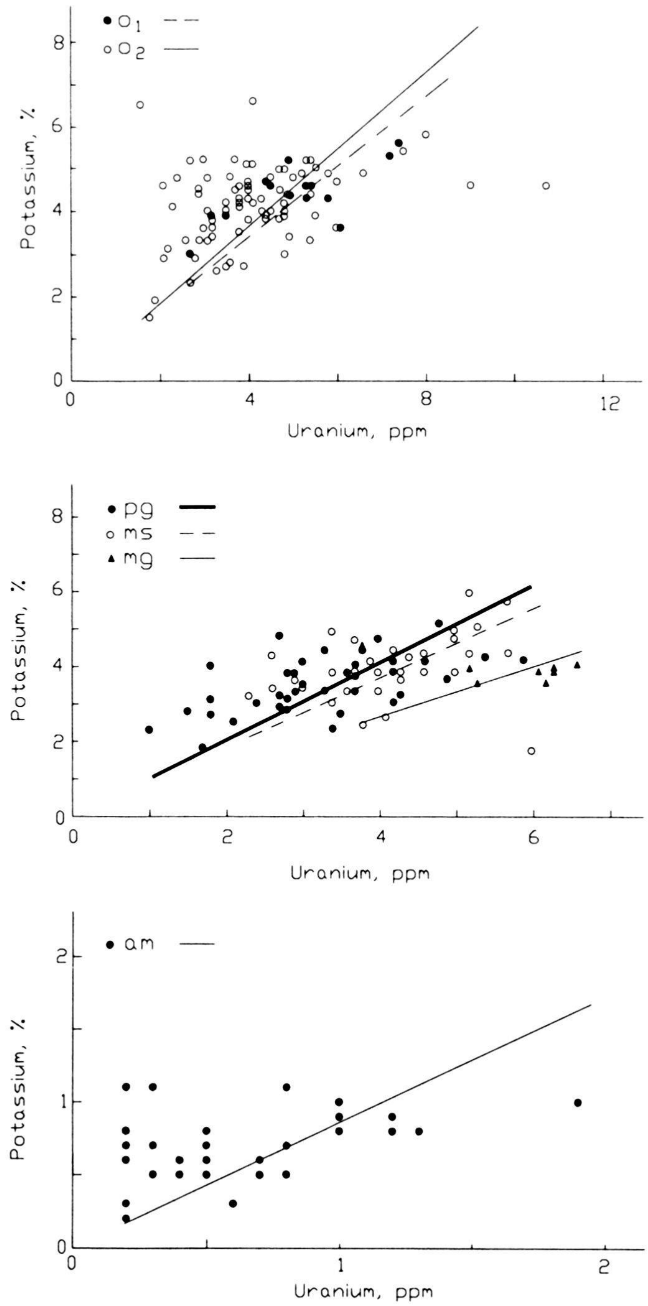

Fig. 4. Potassium versus uranium concentrations in the analysed rocks. Details as in Fig. 3.

As the investigated sites range in altitude from about 100 to 800 m above sea level, the possible variation of the background radiation was also verified by choosing outcrops at different elevations. The "zero background" count rate was assessed over the sea, the γ activity of seawater being negligible. The background count rate measured within each energy window vs. elevation does not show any particular trend (Fig. 2). The relatively small deviations in count rate (maximum for K

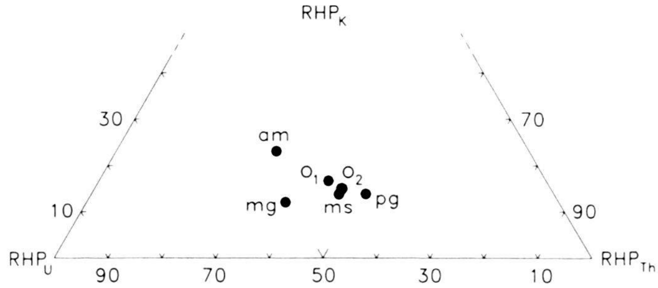

Fig. 5. Relative contribution of each radioelement to the total RHP rate. The code next to each point refers to rocks of Table 1.

and minimum for Th) fall within a reasonable range of "statistic" variability. Therefore, average count rates in each window were assumed as the reference background value δ_1, δ_2, δ_3.

Figure 1 shows the sites of HPE measurements and the corresponding RHP rates. A total of 211 outcrops was analysed. Such a number of data and the spatial distribution of measurements can be considered as representative of the possible variations in HPE concentrations for the different lithologies of the Briançonnais-Piedmont Zone. Figures 3 and 4 depict the correlation of U with Th and K, respectively, for all data.

Particular care was taken to avoid outcrops affected by weathering or not presenting a good source-detector geometry, which otherwise could have given biased results (cf. Chiozzi et al. 2000). Moreover, we applied a statistical procedure (Chauvenet's criterion) for individuating extreme values suspected of being of lower quality. This implied the rejection of two data for each lithotype collected in the field work.

Table 1 summarises the results in terms of mean concentration of radioelements, Th/U and K/U ratios and RHP rate. The heat production constants, that give the heat generated for unit volume and time of U, Th and K on the basis of well known isotopic decay schemes, half lives and mass differences, together with the average rock density, allow the calculation of RHP (Rybach 1988). The average contribution of each HPE to the total RHP rate, depending on the radioelement concentration and the value of its heat-production constant, is given in figure 5.

Discussion

The distribution of uranium, thorium and potassium of igneous rocks is controlled by processes involved in magma genesis. Such elements are defined "incompatible", as in the fractional crystallisation process they denote affinity to the liquid silicic phase wherein they increase in concentration with the increasing of the differentiation degree. Th and U have a different behaviour from K, as they do not form proper mineral phases, but they are present as trace elements in accessory minerals such as apatite, rutile, titanite and zircon. They enter the crystal lattice to replace other elements, as their ionic radius is

Tab. 1 HPE content after three stepwise eliminations of the extreme values and radiogenic heatproduction rate (RHP) of the analysed rocks. n is number of sites; in round brackets the standard deviation of the average values and in square brackets the sample number for density determinations

Rock type (code)	n	U ppm	Th ppm	K %	Th/U	K/U $\times 10^4$	Density 10^3 kg m^{-3}	RHP μW m^{-3}
	14	5.1 (1.4)	16.0 (2.1)	4.43 (0.69)				
Orthogneiss I	13	4.9 (1.2)	16.3 (1.9)	4.54 (0.58)				
(o1)	12	4.7 (1.1)	16.6 (1.8)	4.62 (0.53)	3.54 (0.89)	0.99 (0.25)	2.66 (0.05) [7]	2.74 (0.19)
	84	4.2 (1.5)	18.0 (5.0)	4.18 (0.93)				
Orthogneiss II	83	4.1 (1.3)	17.9 (4.7)	4.22 (0.89)				
(o2)	82	4.0 (1.2)	17.8 (4.4)	4.18 (0.85)	4.40 (1.74)	1.03 (0.38)	2.68 (0.06) [4]	2.65 (0.26)
	40	3.2 (1.1)	17.2 (4.1)	3.48 (0.73)				
Paragneiss	39	3.1 (1.0)	16.6 (3.3)	3.52 (0.69)				
(pg)	38	3.1 (1.0)	16.8 (3.1)	3.47 (0.65)	5.47 (1.96)	1.13 (0.41)	2.71 (0.06) [4]	2.29 (0.19)
	8	5.7 (0.9)	16.7 (2.9)	3.86 (0.32)				
Migmatite	7	6.0 (0.6)	16.3 (1.5)	3.77 (0.20)				
(mg)	6	6.1 (0.5)	16.8 (0.9)	3.87 (0.17)	2.84 (0.25)	0.61 (0.03)	2.73 (0.05) [3]	2.99 (0.15)
	33	0.6 (0.4)	1.3 (0.9)	0.69 (0.23)				
Amphibolite	32	0.6 (0.3)	1.2 (0.8)	0.70 (0.21)				
(am)	31	0.5 (0.3)	1.2 (0.7)	0.69 (0.15)	2.21 (1.94)	1.30 (0.84)	2.89 (0.07) [5]	0.30 (0.06)
	32	4.2 (1.0)	18.4 (5.1)	3.92 (0.89)				
Micaschist	31	4.1 (0.9)	17.9 (4.2)	3.99 (0.81)				
(ms)	30	4.1 (0.9)	17.5 (3.8)	3.92 (0.74)	4.22 (1.27)	0.95 (0.27)	2.72 (0.07) [7]	2.66 (0.21)

comparable to that of the replaced ions. They often lie as impurities in the lattice vacancy of minerals such as quartz, feldspars and micas (Van Schmus 1995).

The results of HPE concentration (Fig. 1 and Table 1) show that the distribution of radioelements in the metamorphic rocks of magmatic origin of the Briançonnais-Piedmont Zone is mainly related to composition and degree of evolution of the parent rock. In felsic rock types, the RHP rate is not very variable, being in the range 2.3 to 3.0 μW m^{-3}. The lowest value of RHP (0.3 μW m^{-3}) was found in amphibolites.

Amphibolites are clearly distinct compared to the other lithotypes for their low U, Th and K content. They show radioelement concentration values by about 8, 14 and 6 times lower than orthogneiss I and II and migmatites, which, on the average, vary within 4.0–6.1 ppm in U, 16.6–17.8 ppm in Th and 3.9–4.6 % in K. Orthogneisses I have radiogenic element concentrations slightly different from those of orthogneisses II. The larger SiO_2 content in orthogneisses I, corresponds to a larger U content. In orthogneisses II, HPEs increase from the granodioritic to the monzogranitic terms, the latter being more rich in quartz. The distribution of radiogenic elements in migmatites is also related to the anatectic process, during which, melting and crystallisation control their concentration (e.g. Kissling et al. 1978). Therefore it is not surprising that these rocks show the largest U concentration, and their Th and K content is comparable to that of orthogneisses and micaschists, another possible source of anatectic melts.

In metasedimentary rocks, HPE concentrations are not so different from migmatitic rocks and igneous protoliths. Paragneisses show Th content comparable to that of orthogneisses I and II, whereas U and K are slightly lower. This could be ascribed to the fact that all these rocks might derive from a common protolith (metapelites) of felsic composition.

In principle, it cannot be excluded that metamorphism might have played a role in the redistribution of HPEs. With the increase of the metamorphic grade, depletion is expected in Th and U. At high pressure and temperature, ions Th^{4+} and U^{4+} in octahedral coordination migrate towards zones unaffected by metamorphism. U^{4+} is easily soluble and tends to oxidise into ion U^{6+}, characterised by higher mobility. Ion mobilisation processes are controlled by the degree of solubility and the energy coming into play, and are favoured by the presence of circulating water. Therefore, after an alteration phase the more soluble ions are mobilised and transported in reducing environments, where they precipitate within sedimentary deposits.

The mineral assemblages of the investigated rocks suggest different phases of re-equilibrations, and possible mobilizations during the metamorphic episodes or hydrothermal late-Alpine tectono-metamorphic phases (Cortesogno et al. 1997). The metamorphic peak of amphibolites, orthogneisses and paragneisses should be at 0.6–0.8 GPa of pressure and 600–650 °C of temperature, corresponding to the amphibolitic facies. Migmatites originated at higher thermobaric conditions

(1.2–1.7 GPa, 650–750 °C) at the transition from eclogite to granulite facies. However, this does not seem to have affected the U, Th and K distribution, which seems more dependent on protolith composition and partial melting processes.

Quantitative determination of U and Th with γ spectrometry assumes secular radioactive equilibrium between parent isotopes ^{238}U and ^{232}Th with their decay products ^{214}Bi and ^{208}Tl, respectively (e.g. Faure 1986). For the ^{238}U series, which results from the relative mobilities of uranium itself and its decay products ^{234}U, ^{226}Ra and ^{222}Rn, this might not be valid. Instead, the ^{232}Th series may be considered in equilibrium in most geological environments. The Th/U and K/U ratios in the sampled localities range from 2.2 to 5.5 and from 6100 to 13000, respectively. The K/U ratio estimates are highly variable similarly to those reported in the literature (e.g. Roger & Adams 1969). If one excludes migmatites and amphibolites, the Th/U ratio ranges from 3.5 to 5.5, that is not very different from that generally observed in igneous rocks. This seems to indicate that the rock radioelements have not been significantly fractionated during weathering or involved in metasomatic activity. However, gneisses show a Th/U ratio which decreases exponentially with the uranium content, similarly to what observed by Kissling et al (1978) for Variscan granites of the Gotthard massif.

Amphibolites have relatively lower Th/U ratio and, consequently, show some uranium enrichment. This implies that the parent rock of basaltic composition could have been reworked in sedimentary, subaerial (oxidising) environment. However, it must be stressed that U and Th concentration in amphibolites at several sites is very close the instrumental detection limit (cf. Figs. 3 and 4). Therefore, the Th/U and K/U ratio could be affected by some uncertainty.

Figure 5 shows the relative contribution to the radiogenic heat production supplied by U, Th and K for the investigated lithotypes. The largest contribution is given by U (43–51%) in orthogneisses I, migmatites and amphibolites, and by Th (46–51%) in the other rocks. The heat produced by K is maximum (23%) in amphibolites.

Concluding remarks

Field γ-radiometric surveying proved to be a good tool for mapping lithological variations over a rough topography on the basis of detailed and rapid reconnaissance of the HPE concentration. Provided that the γ-spectrometry apparatus is appropriately calibrated with special reference to the background radiation and weathered outcrops are avoided, the distribution of U, Th and K can give information about the fine variation in chemical composition, and results are not dissimilar from laboratory measurements. The quantitative assessment of HPE and consequently of RHP is immediate and related to large rock volumes, being thus statistically more representative than laboratory samples.

The Briançonnais-Piedmont Zone rocks underwent a complex metamorphic history with superposition of different p-T conditions, corresponding to the main phases of Alpine tectonism. Their HPE distribution seems, however, more dependent on the parent-rock composition, as metamorphic reactions and/or superficial processes are difficult to ascertain. RHP is higher in orthogneisses (2.66 $\mu W\ m^{-3}$) and migmatites (2.99 $\mu W\ m^{-3}$). The lowest rate of heat production was found in amphibolites (0.30 $\mu W\ m^{-3}$). Para-derived rocks show as a whole RHP of 2.45 $\mu W\ m^{-3}$, i.e. comparable to that of ortho-derived rocks, thus indicating a probable common protolith. The substantial homogeneity in composition is also evident from the relative contribution of each HPE to the total RHP. In most of the analysed rocks, the heat production is almost equally partitioned between U (43–51%) and Th (46–51%).

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