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The Clermont radiometric reference rocks: a convenient tool for dosimetric purposes

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Abstract

Boreholes drilled in 8 different rocks situated in the vicinity of the Clermont TL laboratory

have been used for testing and calibrating methods devoted to measure the natural

radioactivity. The present paper gives updated data for the nuclide contents and internal dose-

rates for those reference media. Two examples of application are presented. The first example

shows dose-rate calibration of a scintillator gamma probe using a threshold method. The

second example shows a routine technique for TL dosimetry, based on the rocks for dose-rate

reference.

1 – Introduction

Various experiments aimed at testing or calibrating techniques devoted to the measurement of natural gamma and X-ray emissions necessitate reference homogeneous media of well known radioactivity.

For such purpose, the Clermont TL group (Clermont-Ferrand, France) has been using natural rocks for many years (Sanzelle et al., 1988, Miallier et al., 1988, Soumana et al., 1994). This approach has also been adopted by other luminescence dating groups (Prescott et al., 1988). The use of rocks is an alternative to built blocks such as those prepared at the Oxford University with doped concrete (Murray, 1982, Aitken, 1985) or those made of building bricks available in different laboratories (for instance, at Gif-sur-Yvette and Bordeaux). One advantage of natural rocks is that they can easily provide homogeneous media, large enough for representing an infinite volume from the point of view of natural gamma emissions. This is particularly the case for lava flows, which, moreover, are numerous in the vicinity of Clermont-Ferrand. In contrast, the laboratory blocks have generally dimensions lower than the maximal range of the most energetic natural gamma rays. For instance, the cubic Oxford blocks (side = 50 cm) have a thickness (50 g.cm^{-2}) which is only equal to around two mean free paths for the 2.61 MeV photons emitted by ^{208}Tl. Additionally, it is suspected that the Oxford blocks are affected by radon loss in proportions which might attain 10 % (Aitken, Pers. com., 2009 and Bowman, 1976). Such effects have to be accounted for in certain calculations, such as, for example, evaluating the factor between the nuclide content and the derived induced radiation dose rate (Soumana, 1993, Rhodes and Schwenninger, 2007). However, the inner radioactivity of naturally occurring rocks is not determined in the course of construction, as it can be theoretically with doped blocks; this can be a drawback for certain experiments using rocks.

So, since the beginning of the 80's, a dozen of boreholes were drilled in a variety of rocks whose bare surface was exposed either naturally or by human action. These different sites are situated within a distance shorter than 25 km from the laboratory. Recently, in order to reduce the distance to be travelled for experiments, two large blocks, weighting 10.5 t and 9.5 t were transported close to the laboratory (Fig.1). On the other hand, some sites are no longer accessible at present, so that 8 reference rocks are currently available (and possibly 2 more, slightly less convenient). The boreholes are horizontal, they have a diameter of 70 mm and a length between 0.39 m and 1.5 m. The cores extracted in the course of drilling were crushed and homogenized before being distributed among several laboratories for elemental analyses, including nuclide assessment. Some of the nuclides analyses gave rise to inter-laboratory comparisons (Faïn et al., 1997). Dose rates in the different rocks were derived from the nuclide contents. Dose rate measurements were also performed *in situ*, in the holes, by independently calibrated means including TL dosimeters and NaI gamma probes.

The present paper is aimed at giving updated data for those reference media and at presenting two examples of their use. The first one concerns the calibration of a field probe for gamma dose rate measurements. The second one is a simple and advantageous method for TL dosimetry, where the reference rocks are used as radiation-dose-sources, avoiding the use of a laboratory calibrated source.

2 – Data concerning the reference rocks

The reference rocks include two granites, one sandstone and six lavas (Table 1). Apart from the two blocks settled close to the laboratory (C341 and C347), the rocks do not appear as free blocks, but as outcrops in geologic massive formations. Homogeneity at the metre scale around the boreholes is assumed from the geological nature of the rocks, which are not

bedded, and from visual inspection. The water content is nil, or low but stable. Certain lavas are porous; however the pores are not interconnected, so that time-variation in water content is not possible. Although the holes are horizontal, they may sometimes be wet inside, due to rain, but this cannot affect the dose rate.

(Table 1 here)

The nuclide contents of the rocks were assessed by various laboratory methods, comprising: low background gamma spectrometry, ICP-MS, NAA and flame photometry (for K). ^{230}Th was measured by means of alpha spectrometry for recent lavas since these can show a slight disequilibrium in the U-series.

 The nuclide contents were also verified *in situ* for C341 and C347 by using a NaI gamma probe. The laboratories or persons who participated to the measurements are listed in Annex 1.

(Table 2 here).

The gamma dose rates in the different media were calculated on the basis of the conversion coefficients published by Adamiec and Aitken (1998), and the updated coefficients given by Guérin and Mercier (submitted) for ^{40}K and Th-series. Allowance was made for disequilibrium where necessary. The cosmic contribution was evaluated using the data of Prescott and Hutton (1994).

Radon loss has not been specifically measured. It has been considered as negligible for all the rocks, because of their compactness and it has been indirectly verified by the good agreement between the calculated dose-rates and the *in situ* measured dose-rates. Moreover, the activity ratios ^{210}Pb/^{226}Ra for the rocks are not significantly differing from 1.

(Table 3 here)

3 – Application to the calibration of a portable gamma spectrometer

The Clermont reference rocks were used for calibrating a probe aimed at being a convenient tool for gamma dose rate determinations in the field. The detecting cell of the probe is made of a 1.5" x 1.5" scintillation crystal (LaBr$_3$). According to the manufacturer (Canberra), this material exhibits a higher detection efficiency and a higher resolution than the more classical NaI(Tl) crystals. An example of spectrum recorded in a cubic block made of building bricks (side: 1.5 m) available at the CRP2A laboratory (Bordeaux) is given in Fig. 2. However, this probe has the disadvantage of a relatively high background (Fig. 2), due to the presence of the ^{138}La isotope in the crystal lattice which generates γ– rays at 1436 and 789 keV, this second emission following a beta decay (end-point energy: 255 keV) that induces counts in the 789-1044 keV range. Thus, in the background spectrum, one also notices the presence of a line at 1461 keV, probably due to the summation of gamma rays of 1436 keV and X-rays following electron capture, as well as peaks related to the Th-series.

Countings were performed with this probe in the following Clermont reference rocks: C347, C341, PEP, LMP, MAZ and GOU, as well as in the Bordeaux block of bricks in order to get a point at ~2000 µGy/a. As each spectrum is a mixture of γ-rays coming from the ^{40}K and U- and Th-series, we analyzed the spectra in applying two threshold techniques (Guérin and Mercier, submitted). With the first technique, the total number of pulses above a fixed threshold was cumulated and counted. This threshold value was determined with the Geant4 code simulating a LaBr$_3$ cell (encapsulated in a 1 mm thick duraluminium layer) placed in infinite media. These media had various compositions similar to sediments rich in carbonates,

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silicates, organic materials or lavas; the different radio-emitters - ^{40}K and those of the U- and Th-series- were distributed uniformly. According to these Monte-Carlo simulations, the number of counts detected per unit time by the LaBr$_3$ cell above an energy of 300 keV (Σ Ni , E > 300 keV), normalized to 1 µGy/a, is independent of whether the γ-rays originated from K, U or Th, and this value was then used as the low threshold. A high threshold of 2800 keV was set in order to eliminate the cosmic contribution to the counts (see Prescott and Clay, 2000). Fig. 3 shows the total count above this threshold between 300 keV and 2800 keV as a function of the gamma dose-rate, indicating that 0.0338 ± 0.008 counts per second for a 1 µGy/a dose-rate are detected with this probe.

The analysis of the same simulated data indicated that above 165 keV, the sum of the energy per unit time i.e. Σ Ni. Ei (which is, for each channel, the product of the number of counts in this channel with the corresponding energy) is also independent when normalized to 1 µGy/a, of the origin of the gamma-rays. In applying this second threshold technique, a larger part (about twice in counts number) of the recorded spectrum is used, which improves the counting statistics used for the dose rate determination. Moreover, Guérin and Mercier showed that this technique is less sensitive than the previous one to the repartition of the gamma dose-rate between ^{40}K and the U- and Th-series. The same Geant4 simulations indicated that this technique is also nearly insensitive to the medium composition.

With this second technique, a good linearity was found between the signals (Σ Ni . Ei) and the gamma dose rates (Fig.4).

4 – In situ TL dosimetry using the reference rocks

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4 – 1 Verification of the consistency of the dose-rate data.

A campaign of TL dosimetry was especially devoted to check the internal consistency of the dose-rate data (Table 3). For that purpose, powdered Al_2O_3:C (provided by Landauer Inc., Stillwater Crystal Growth Division) was used (grain size : 200-315 μm). The Clermont laboratory routine dosimeter consists of a stainless steel tube (1.5 mm thick, 0.25 cm^3 of capacity) completely filled with the zeroed powder. The cap is maintained in place by a heat shrinkable tube, slightly longer than the steel tube (+ 1 cm at both ends). When in place and very hot, the shrinkable tube is pinched at both ends so as to form a water proof bag around the steel tube, in the form of a sweet paper.

Such dosimeters were inserted in the holes for a few months and they were afterwards measured in the laboratory, all at the same time. The intensities of the TL peaks were plotted against the known dose rates (Fig. 5). No correction was applied to the results, because possible undesirable effects, like self dose and fading, would be exactly the same for all the dosimeters and are therefore not supposed to affect the linearity of the curve. However, this is not true for attenuation by the walls of the tube, which actually depends both on the gamma spectrum – that is the respective proportions of U, Th and K in the rocks – and on the composition of the rock (see e.g., Valladas, 1982, Faïn et al., 1985). The exact attenuation in the different media for the given configuration has not been computed yet. It can be expected from preliminary results that the difference of attenuation between the samples is in the range 0 – 3 %.

A good correlation between TL and estimated dose-rates was obtained (Fig.5), thus proving the validity of the data.

4 – 2 TL dosimetry

Routine methods for evaluating the dose acquired *in situ* by a TL dosimeter imply the use of a calibrated radiation-source (X, beta or gamma) for getting the correspondence: TL vs dose.

Conversion of the measured equivalent dose in terms of *in situ* annual dose may require

application of correction factors such as those evoked above for fading or self-dose; such

corrections are routinely made by certain laboratories. Moreover, it necessitates a precise

knowledge of the dose-rate delivered by the laboratory source. So, inevitably, errors and

uncertainties accumulate in the conversion. To overcome such problems, the Clermont TL

group proceeds as follows.

When a set of dosimeters is to be placed in sites to be dated, 3 more dosimeters are prepared

and placed in 3 of the 8 reference boreholes at the same time. After the dosimeters have been

collected months later, all the phosphors are measured at the same time. Then, the TL

intensities are plotted vs annual dose for the three known ones. The dose-rates in the different

measured sites are then obtained by a simple interpolation (Fig.6). The overall uncertainty

(systematic + statistic) on the result is estimated at 4.5 % of the annual radiation dose. The 3

reference rocks that have been chosen are C341, C347 and PEP, considering the advantages

of such choice: (i) the representative points on the initial experiment (see Fig.4) are on the

mean regression straight-line (least squares method), thus meaning that the dose-rates have

been correctly evaluated for those 3 holes; (ii) the dose-rates are significantly different from

each other, allowing exploration a large domain of dose-rates and, (iii) the sites are the most

convenient, being the closest to the laboratory.

4 – 3 Discussion of the method

The method offers advantages that have already been mentioned: several possible sources of

error with the standard techniques have no effect here because they are the same for all the

dosimeters and, accordingly, they are automatically corrected for by the interpolation. The

method avoids the use of a laboratory source, with the relevant difficulties (authorization and

calibration). Moreover, the number of TL (or OSL) measurements is limited: for N dosimeters, the total number of useful series of measurements is N+3 only (a "series "corresponds to the several measurements necessary for reducing statistical uncertainty for a given sample). Additionally, there is no need to measure the background (noise) or to worry about possible spurious signal.

However, the drawback of the method is that the sites of application must not be too far away from the laboratory. For remote sites, it is necessary to introduce complementary measurements for evaluating the travel-dose; in any case, the *in situ* duration must be long enough for the corresponding dose to be significantly higher than the travel-dose. Improvements of the method could consist: (i) in using a housing that would induce less dependence of the dose acquired by the TL phosphor on the composition of the measured medium and (ii), in evaluation of the attenuation for a given medium with, e.g., a Monte Carlo code. Note that this applies to the standard methods too.

5 – Conclusions

The reference rocks established around the Clermont laboratory can be considered as a convenient tool for various dosimetric experiments. The accuracy and precision of the data published in the present paper can still be improved by new independent measurements, either in the boreholes or in the assessment of the nuclide content of samples from the cores.

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Table captions

Table 1 : Composition and nature of the reference rocks.

Table 2: Nuclide contents of the reference rocks. The ratios ^{230}Th/^{238}U and ^{226}Ra/^{230}Th are given for activities. For the older rocks, PEP, LAS, LMP, MPX, and C347, these ratios were not measured; they were inferred from the ages of the rocks (3 Ma for the youngest one, LMP). Uncertainties are quoted at the 95% level of confidence.

Table 3: Gamma and cosmic dose- rates in the reference rocks. Gamma dose-rates were calculated using the data of Adamiec and Aitken (1998) revised for ^{40}K and Th-series by Guérin and Mercier (submitted). Cosmic dose-rate was derived from the data of Prescott and Hutton (1994).

Figure captions

Figure 1: Drilling the block of granite C347, weighting 9500 kg.

Figure 2: Time-normalized spectra (1000 s) recorded with a LaBr3 probe placed in the block of bricks available at CRP2A (BDX- gamma dose rate= 2010±35 µGy/a) and in a lead housing (Background). High resolution gamma spectrometry performed on bricks used for building the CRP2A block allowed determination of their average radioisotopic contents : U(^{238}U)=4.27±0.68 ppm, U(^{226}Ra)=4.10±0.14 ppm, Th= 13.7±0.3 ppm, K=3.50±0.05%. Note that the Y-scale is logarithmic.

Figure 3 : Net counts per second (ΣNi for E> 300 keV) deduced from measurements in the Clermont reference rocks and in the Bordeaux block of bricks as a function of the gamma dose-rate (see values in Table 3). The upper discrimination level was fixed at 2800 keV, in order to eliminate the counts due to cosmic rays ~~sum all the gamma counts~~. Notice the good linearity and that the intercept is compatible with 0.

Figure 4 : Time-normalized signal (ΣNi. Ei for 165 keV < E < 2800 keV) deduced from measurements in the Clermont reference rocks and in the Bordeaux block of bricks as a function of the gamma dose-rate. The background was subtracted after time normalization.

Figure 5: Plot of TL signal vs dose-rate for 8 reference rocks

Figure 6: Plot of TL signal vs dose-rate for 3 selected reference rocks (C341, C347 and PEP) based on a different campaign of measurements than the one for Fig.5. The dotted line shows

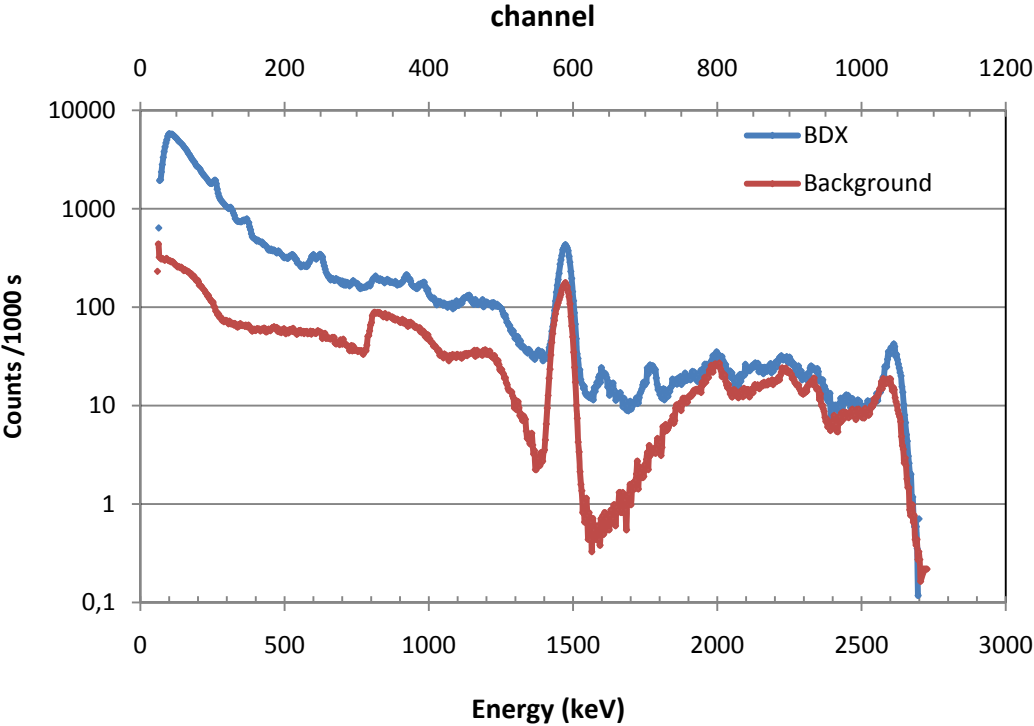the determination, by interpolation, of the annual dose for a dosimeter placed in a site to be dated.
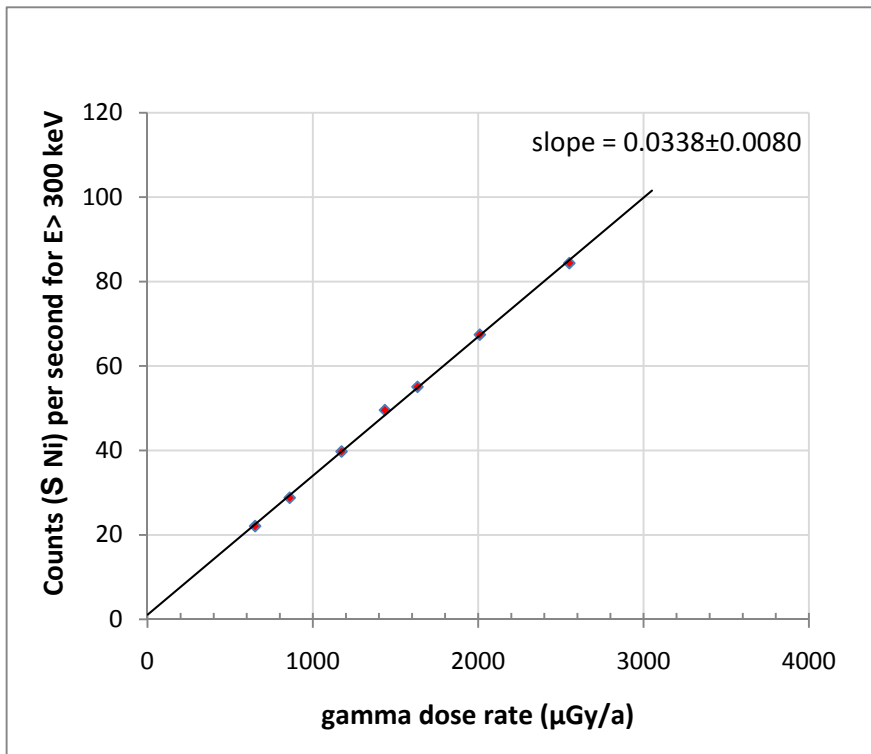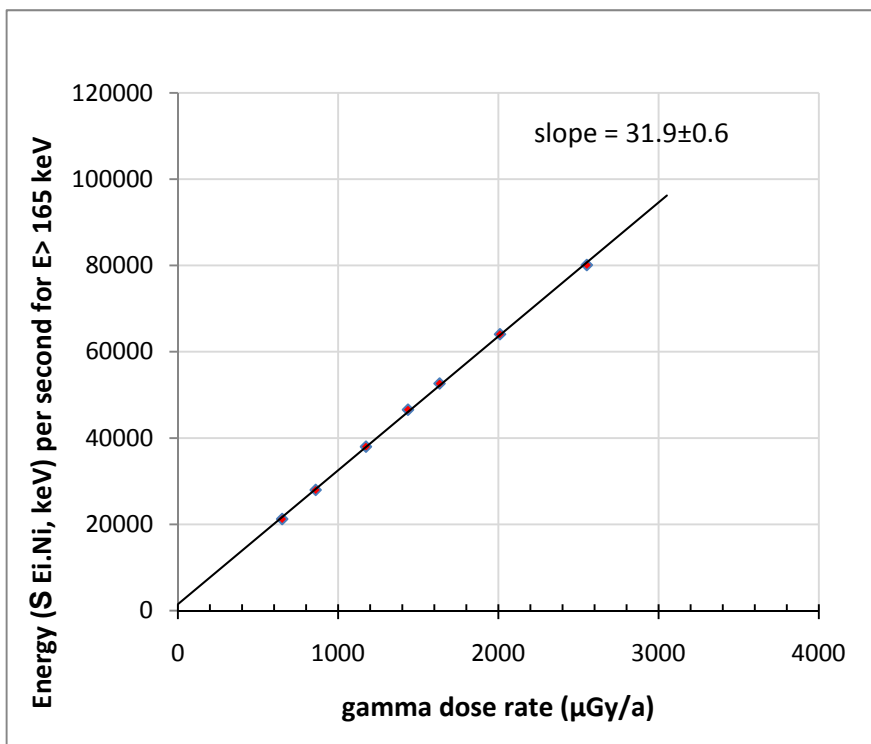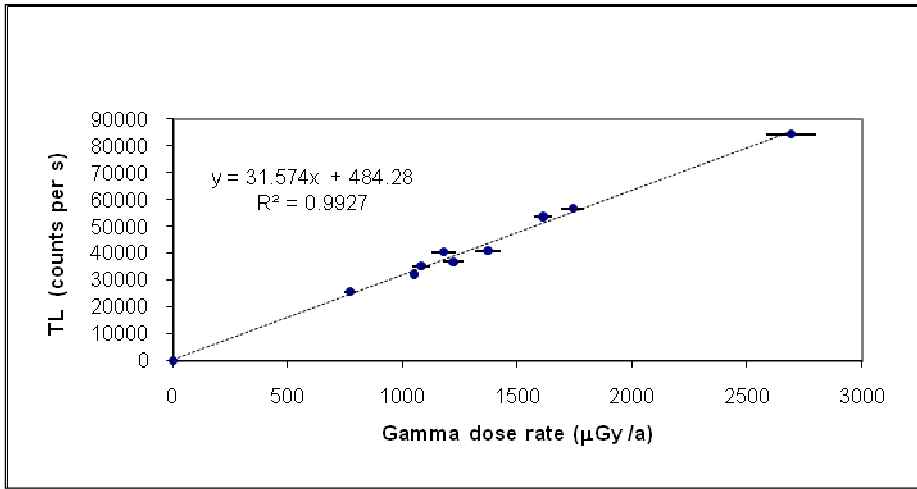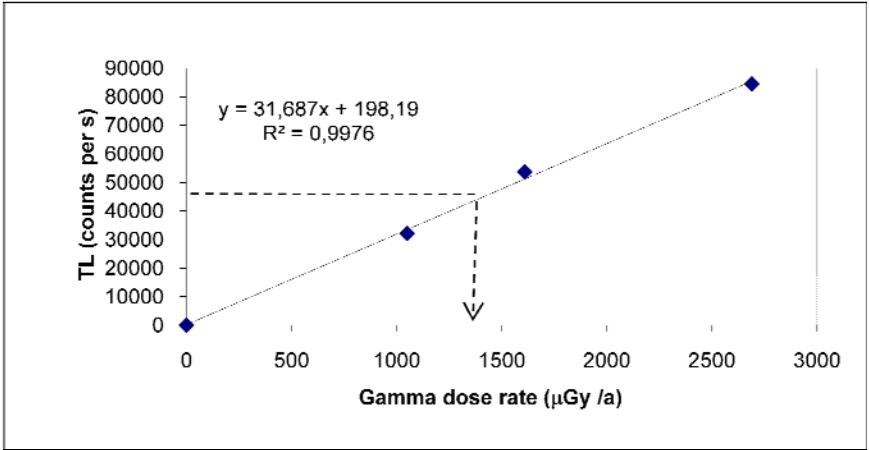
Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Reference	SiO$_2$	Al$_2$O$_3$	Fe$_2$O$_3$	MgO	CaO	Na$_2$O	K$_2$O	TiO$_2$	P$_2$O$_5$	MnO	H$_2$O (1000°C)	H$_2$O (110°C)	total	Nature
PEP	60.0	16.2	5.6	2.85	3.19	3.36	4.63	0.84		0.08	2.1	0.11	98.96	granite
LAS	49.5	16.0	10.6	5.67	8.68	4.21	2.33	2.25		0.18	0.21	0.26	99.89	basalt
LMP	42.9	12.6	12.8	10.6	11.1	2.64	0.87	2.84		0.17	2.1	1.06	99.75	basalt
MPX	86.0	6.89	0.83	0.11	0.07	0.19	3.08	0.05		0.01	1.35	0.1	98.59	sandstone
MAZ	49.19	17.13	12.12	4.69	8.47	4.36	2.24	2.39		0.2	0	0.33	100.07	trachyandesite
GOU	54.9	18.2	7.91	2.25	5.47	5.4	3.13	1.38		0.22	0	0.03	98.9	trachyandesite
C 347	73.09	15.05	1.45	0.38	0.89	3.62	4.27	0.13	0.19		0.82		99.79	granite
C 341	46.95	16.67	12.15	6.16	9.59	3.71	1.65	2.44	0.57	0.19	0.17		100.28	trachybasalt

Table 1

Reference	[U] ppm	±	[Th] ppm	±	$^{230}Th/^{238}U$	$^{226}Ra/^{230}Th$
PEP	6.00	0.20	19.0	2.0	1	1
LAS	2.14	0.04	7.57	0.15	1	1
LMP	1.60	0.14	5.91	0.09	1	1
MPX	1.38	0.03	3.61	0.20	1	1
MAZ	2.39	0.08	8.58	0.04	1.00 ± 0.01	1.07 ± 0.01
GOU	3.18	0.12	11.95	0.06	1.00 ± 0.01	1.11 ± 0.01
C 347	2.84	0.12	4.67	0.10	1	1
C 341	1.80	0.05	6.42	0.40	1.10 ± 0.01	1.0 ± 0.01

Table 2

Reference	Dγ $\mu Gy.a^{-1}$	Dcosmic $\mu Gy.a^{-1}$	Total
PEP	2552 ± 110	140 ± 14	2692 ± 110
LAS	1096 ± 10	120 ± 20	1216 ± 22
LMP	651 ± 18	120 ± 20	771 ± 27
MPX	971 ± 13	110 ± 15	1081 ± 20
MAZ	1174 ± 12	200 ± 20	1374 ± 23
GOU	1633 ± 17	110 ± 11	1743 ± 23
C347	1436 ± 25	170 ± 18	1606 ± 29
C341	860 ± 21	170 ± 18	1030 ± 26

Table 3

Annex 1

List of the persons who contributed to the assessment of the nuclide content and/or gamma dose-rate in the reference blocks.

Laboratory	Person	Method
Centre de Recherches en Physique Appliquée à l'Archéologie, CRP2A, Université de Bordeaux 3.	P. Guibert, N. Mercier	Gamma spectrometry (Hp- Ge detector), field gamma-probe.
Laboratoire des Sciences du Climat et de l'Environnement, LSCE, Gif-sur-Yvette.	N. Mercier, H. Valladas	TL, field gamma-probe.
Institut de Paléontologie Humaine, IPH Paris	J.J. Bahain, C. Falguères	Gamma spectrometry (Hp- Ge detector)
Laboratoire Pierre Sue, CEA, Saclay	J.L. Joron	NAA
Laboratoire Magmas et Volcans, Clermont-Ferrand.	M. Condomines	ICP-MS, alpha spectrometry
Laboratoire d'analyses et mesures, CRN, Strasbourg	J. Tripier	TL
Laboratoire de Physique Corpusculaire, LPC, Clermont-Ferrand	J. Faïn, H. Erramli, D. Miallier, T. Pilleyre, S. Soumana, S. Sanzelle	Gamma spectrometry (Hp- Ge detector), TL
Centre de Recherches Pétrographiques et Géochimiques, CRPG, Vandoeuvre-lès-Nancy		ICP-MS
Hokkaido University of Education, Japan.	Y. Ganzawa	NAA