A real-time compact monitor for environmental radiation: Cosmic rays and radioactivity (*)

S. CECCHINI (¹)(²), A. COTTA RAMUSINO (²), F. FRONTERA (³), M. GALLI (⁴)

I. LONGO (⁴), M. RICCIOTTI (⁵) and R. SACCHETTI (⁶)

(¹) Istituto TESRE-CNR - Bologna, Italy

(²) Istituto Nazionale di Fisica Nucleare, Sezione di Bologna - Bologna, Italy

(³) Dipartimento di Fisica, Università di Ferrara - Ferrara, Italy

(⁴) Dipartimento di Fisica, Università di Bologna - Bologna, Italy

(⁵) Agenzia Regionale Protezione Ambientale - Rimini, Italy

(⁶) Ministero Pubblica Istruzione - Bologna, Italy

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Summary. — We report here about the possibility of using a compact scintillation NaI(Tl) detector, long-term stable and reliable, to monitor separately the components of the environmental radiation, *i.e.* in the energy range 0.28–2.8 MeV, due to very low energy secondary (Ultrasoft) cosmic radiation and radioactivity, airborne and from environment matter. We suggest some procedures to accomplish time variation analysis, by using a sample of data collected in Bologna.

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1. – Introduction

As it is known, the pulses of an organic or inorganic scintillation counter, corresponding to more than 50 keV of energy lost in the scintillator, originate mainly from the most degraded secondary cosmic rays, from the airborne radioactivity, from the surrounding materials and from the detector itself. This radiation, that we will call *environmental radiation* (ER), at ground gives a counting rate that may exceed by more than two orders of magnitude the cosmic-ray (CR) total ionizing component with energies greater than 5–8 MeV (Bernardini and Ferretti, 1939; Rossi, 1948).

The real-time monitoring of the ER and the variation of its components, besides warning in real time about possible radiation anomalies of artificial origin (nuclear

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reactor accidents, nuclear bombs, pollutants, etc.) or astrophysical and geophysical origin (solar flares effects, solar activity), it is surely important for those activities for which it is necessary to know the ER *in itself* or as a background because it can seriously interfere, for instance, with the operation of a health diagnostic apparatus (Theodorsson, 1994).

The NaI(Tl) scintillation detector of ER we have developed has a free air statistics at sea level of ~ $0.5 \cdot 10^6$ counts/h in the band 0.28–2.8 MeV, and ~ 10^4 counts/h around the photoelectric peak of 40 K; it has a long-term stability absolutely reliable; the whole set has a small volume (~ $130 \times 70 \times 70$ cm³); it has a reasonable cost, and requires a fair operation charge; it is, therefore, suitable to be used as a monitor of ER.

In what follows we report some observations and procedures used also to obtain a separate monitoring of the various components of radioactivity, besides the monitoring of the very low-energy secondary CR component, that was observed long time ago by Bernardini and Ferretti (1939) and called by them *Ultrasoft component* or *Mollissima*.

We will also show a sample of the results that can be obtained.

2. - The detection of the environmental radiation

A sketch of the structure of our scintillation detector is shown in fig. 1. The monocrystal of NaI(Tl), which has a diameter of 8" and a height of 4", has been supplied by BICRON Corp., New Bury (OH, USA). The scintillations of the crystal are seen through a light guide by a photomultiplier tube (PMT) of 5" diameter also supplied by



Fig. 1. – Drawing of the detector unit (not in scale). Dimensions are shown.

TABLE I – The pulse height stability relative to the photopeak of 40 K and of 208 Tl for all the 191 three hourly spectra of the considered interval.

Nuclide γ-line energy	$^{40}{ m K}$ 1.461 MeV	²⁰⁸ Tl 2.615 MeV
Order number of channel (14 keV/channel)	103;104;105	181; 182; 183; 184;185
Frequency of the order number	0;576;0	0; 42; 477; 7; 0

BICRON Corp. The PMT is provided of a mu-metal shield in order to avoid gain variations due to varying magnetic fields. The PMT photocathode is electrically connected to its metalized glass and to the the mu-metal shield.

The output signals from the PMT are sent to a charge pre-amplifier and then amplified and energy analysed. The energy analysis is obtained through a multichannel analyzer of 1024 channels of variable width, 7.5–100 keV/channel. A complete description of the apparatus will be made elsewhere.



Fig. 2. – Three hourly pulse spectra for the channel interval 16 to 200: average (curve "a"), minimum (curve "m"), and maximum (curve "M") during the period May 17th 00:03–June 9th 24:00 LT. The minimum spectrum lies on a regular curve , $I_r(n)$, that we call "reference virtual spectrum". In the insert we can see the average cosmic-ray spectrum with energy > 2.8 MeV.



Fig. 3. – Examples of reduced spectra $(I(n, t) - I_r(n))$ corresponding to the minimum and the maximum spectra of the considered time interval. Notice that the ¹³⁷Cs peak at 661.6 keV is barely visible in the maximum spectrum as a deformation of the ²¹⁴Bi peak at 609.3 keV.

An important feature of our detection system is its time stability. It has been achieved by a feedback gain control based upon the electric pulse height, corresponding to a given photoelectric peak in the energy spectrum. After fixing the channel in which the peak is desired to be, the possible variation of the corresponding pulse heights are recognized and corrected by the electronics by means of a feedback action on the high-voltage supply of the PMT. In our case we have considered the peak corresponding to the 1.461 MeV gamma line of ⁴⁰ K, naturally present in the surrounding solid material and in the detector itself. By this system the overall pulse height gain can be stabilized within a few permill, as shown in table I. In that table the variation of the channel number of the ²⁰⁸Tl peak appears to be consistent with the effect of the counting Poissonian fluctuation of $\sim 0.5/183 = 2.7\%c$.

In fig. 2 we show, in decimal semi-logarithmic coordinates, the behaviour of the average spectrum and also of the minimum and the maximum spectra in the energy interval corresponding to channels n = 16 to n = 200 with 14 keV/channel, obtained during the time interval 00:00 h 17th May 1994–24:00 h 9th June 1994, altogether 191 three hourly intervals; the monitor was located on the roof of the building of the Physics Department of Bologna University under a layer of material not greater than 2 g/cm^{-2} .



Fig. 4. – An example of separation and determination of line intensity from a reduced three hourly spectrum. In a first approximation this is possible by knowing the energy resolution (see text) and the difference *peak minus adjacent valley to the right*. Within the maximum spectrum of the ²¹⁴Bi (609.3 keV) line, one can see the separation of the ¹³⁷Cs (661.6 keV) line and of the two bismuth lines around the ⁴⁰K line. The dotted continuous lines show the smaller peaks coming out after subtraction of the big peak of bismuth and the one of ⁴⁰K from the maximum spectrum. The FWHM of the subtracted and resolved peaks are indicated.

In this representation all the spectra I(n, t) show undulations whose amplitude decreases towards the lower energies where the general level increases rapidly, and seem to correspond to photoelectric peaks of about the same order of magnitude. The greatest of them correspond to lines of ²⁰⁸Tl, ⁴⁰ K, the ²²² Ra daughters (that we shall call radon) and of e[±] annihilation. The continuous behaviour on which these peaks lay is due to the continuous radioactive background (Compton continuum, backscattering, etc.) plus the ultrasoft cosmic-ray gamma radiation that can be considered as the end of electronic cascades of various origin (Rossi, 1948).

3. - Measuring the amplitudes of the photoelectric peaks in the pulse spectrum

From our 191 three hourly spectra we take the one corresponding to the minimum total number of counts and fit to it a *regular curve* that approximate as much as possible to its undulations from the lower side. We shall call it *virtual reference* spectrum $I_r(n)$.



Fig. 5. – Photoelectric peak and the continuous spectrum 40 K. It indicates the energy resolution of the present detector at 1.461 MeV. It has been obtained through the difference of one hour counting with and without a kg of potassium hydroxide on top of the detector.

When subtracting this curve from any three hourly spectrum, we will obtain a *residual spectrum* in which we can clearly see a number of peaks laying on a quasi-horizontal baseline. They appear to be bell-shaped, more or less overlapping and having a width increasing with energy, according to the resolving power of the detector (Knoll, 1979).

We find that $I_{\rm r}(n)$ according to the multichannel energy division of 14.0 keV//channel may be expressed by the formula

(1)
$$I_{r}(n) = \exp\left[2.3c(n)\right] + G(n; a, p, L),$$

with

$$c(n) = B - kn + A \exp\left[-n/d\right],$$

n being the order number of the channel, B the intercept on the ordinate axis of the straight line continuing the spectrum for E > 2.8 MeV (see fig. 2), k the angular coefficient of the mentioned line and

(2)
$$G(n; a, p, L) = a^{2(-(n-p)/L)^2}$$

is a Gauss bell-shaped curve with amplitude a, maximum at channel p, and FWHM of 2L channels.



Fig. 6. – Scatter diagram of the counts/3 h at channel 41 (containing the peak 609.3 keV of ²¹⁴Bi) minus the counts/3 h of channel 50, (P - V) vs. the counts/3 h at channel 41 (P) for all the 191 three hourly subintervals considered. As we suppose the difference (P - V) to vanish when radon disappears, the extrapolated value of P when P - V is zero, at constant atmospheric conditions, would give the mean level of ultrasoft cosmic rays plus non-radon radioactivity. The correlation coefficient is r = 0.9972. Data refer to reduced spectra.

With reference to the mentioned data interval, the following parameters appear to be adequate:

 $A = 75\,000$, p = -2, L = 10, B = 3.95, k = 0.0104, d = 40.

In fig. 3 we can see the residual three hourly spectra corresponding to the minimum and the maximum total counting rate between channels 16 and 200. Here we can see several peaks more or less overlapping according to their mutual distances and amplitudes. Assuming that each one is bell-shaped of type (2), when adding $\sum_i G_i(n; a_i, p_i, L_i)$ to the reference spectrum $I_r(n)$ of eq. (1), where a_i, p_i, L_i are suitably chosen for the peak expected at the n_i -th channel, we can obtain the amplitudes of a number of lines that otherwise would have been of difficult observation.

The half-width at half-maximum L_i can be determined as a function of the energy E_i

$$L_i = p_i 0.0385 E^{-0.6}$$



Fig. 7. – Scatter diagram of the counts/3 h at channel 41 (containing the 609.3 keV peak of ²¹⁴Bi) minus the counts/3 h of channel 50 vs. the counts/3 h at channel 183 (containing the peak of ²⁰⁸Tl) in units of the respective standard deviations, for all the 191 three hourly subintervals contained in the considered period. The low value of the correlation coefficient (r = 0.29) indicates the independence of radon daughters from thoron daughters.

corresponding to the resolution power (Knoll, 1979)

$$R_i = \text{FWHM}/p_i = 0.072 E^{-0.6}$$
.

The amplitude a_i can be measured in a first approximation as the counting difference between the *i*-th peak and the next right side local minimum (we call it "valley"), provided their distance is greater than $2L_i$.

In other words, to find the smaller peaks, it is sufficient to determine the position and the amplitudes of the best defined ones, and eliminate them by subtraction.

In fig. 4 we show how it is possible to separate the line corresponding to 137 Cs (661.6 keV) from the line of 609.3 keV of 214 Bi or two peaks of 214 Bi adjacent to the 40 K peak when the maximum quantity of radon daughters is present.

The half-width L_i of a peak as a function of energy can be determined by the differential effect produced by the presence of a certain radionuclide having a γ line in the energy interval we want to consider.



Fig. 8. – The total three hourly counting rate in the energy range 280-2800 keV. This appears to be the typical behaviour of the environmental radiation at ground. The gross variation depends on the radon daughters, as one could see by plotting the scatter diagrams of any two couples of given radon peaks of the pulse spectra. The time decay from local maxima is of the order of 9-12 hours.

In fig. 5 we can see the differential effect due to one kg of potassium hydroxide when laying over our detector for 30 min while atmospheric pressure and temperature were reasonably stable.

4. - The radioactive background and the ultrasoft cosmic rays

The radiation detected as described above, in the mentioned energy band, is essentially due to cosmic radiation and to the airborne radioactivity. The activity of radon daughters (²¹⁴Bi and ²¹⁴Pb) appears to be the most time variable.

As cosmic-ray variation, due to barometric effect or to solar activity has a time variation that should not exceed 20–25%, the large time variability we observe in the total counting in the mentioned energy range has to be attributed to the airborne radon daughters: up to a factor of two above the mean value and up to a factor of ten above the minimum value. In a first approximation we can consider the reference spectrum as the base over which the photoelectric peaks plus the continuum of radon are superposed. So, in principle, we can separate the radon contributions. In fact, when comparing a



Fig. 9. – Integral three hourly counting rate (dots) for low-energy secondary cosmic rays in the energy band 2.8–7 MeV (channels 200–500). The atmospheric pressure at Bologna 50 m a.s.l. (continuous line) is plotted with an inverted scale according to the pressure coefficient 0.48%/mb. Some of the oscillations of the counting rate around the pressure curve can be attributed to the diurnal solar effect.

radon photoelectric peak and the nearest "valley" to its right side, we can easily understand that the difference peak-to-valley would disappear if radon disappears and that its value is proportional to the radon abundance. Furthermore, when this difference disappears the counting level will be only the one due to cosmic rays plus the one of the remaining radioactive sources.

In fig. 6 we show the scatter diagram of three hourly counts of the channel containing the 609.3 keV peak of ^{214}Bi and the difference of this peak from the right side valley of the ^{137}Cs line (see again fig. 4).

The value of the limit of no-radon background can be estimated by the ordinate value reached by the best fit straight line to the cloud, when the mentioned peak minus valley is zero. By means of the other radon peaks, one can understand the possibility to obtain the radon correction over all the spectrum considered.

Figure 7 shows the scatter diagram of the peak-valley difference at 0.6093 MeV of 214 Bi and the counting peak of 208 Tl at 2.615 MeV. The correlation coefficient of r = 0.29 obtained with 191 points indicates the independence from radon daughters of thoron daughters.

Pulses with energies greater than 2.8 MeV in our detector are surely of cosmic origin. Their energy spectrum follows a power law of the type $E^{-\gamma}$, with $\gamma = 1.34$ according to results of measurements of balloons and satellites (Imhof *et al.*, 1976).

5. – The temporal variation

In fig. 8 the temporal variation of the counting rate in the channel interval 20–200 corresponding to gamma's in the energy interval 0.28–2.8 MeV is shown. The time record shows a number of peaks with decay time of 9–12 hours surely due to radon as one can understand from fig. 3 when comparing the times of minimum and maximum spectra.

In fig. 9 we can see the temporal behaviour of the secondary cosmic radiation with energy in the interval 2.8 to 7 MeV and its comparison with atmospheric pressure. As the pressure coefficient is negative and of about 0.5%/mb, and the pressure graph is drawn with inverted scale, the deviation of cosmic radiation counting rate in the mentioned interval from the pressure line should indicate the solar diurnal effect.

A deeper analysis of the pulses in the interval 0.2 to 2.8 MeV should show more details of the temporal variation of the ultrasoft component as a function of energy.

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