Journal of Environmental Radioactivity 100 (2009) 84–88

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

^{210}Po determination in urines of people living in Central Italy

M.A. Meli, D. Desideri*, C. Roselli, L. Feduzi

Institute of General Chemistry, University of Urbino "Carlo Bo", P.zza Rinascimento 6, 61029 Urbino, Pesaro, Italy

ARTICLE INFO

Article history:
Received 4 March 2008
Received in revised form
25 September 2008
Accepted 4 November 2008
Available online 9 December 2008

Keywords:
Polonium-210
Poison
Urine
Alpha spectrometry
Dose assessment

ABSTRACT

This paper presents the results of the monitoring programme on the urines of people living in an area of Central Italy (near the Republic of S. Marino) to evaluate the background level of the ^{210}Po excretion rate (mBq day^{-1}) in this region. The volunteers were subdivided in five age classes and in every age class groups of males and females, cigarette smokers and non-smokers were taken into account. The results indicated that the ^{210}Po excretion rate was widely distributed within each group of volunteers. The ^{210}Po excretion rate was <30 mBq day^{-1} for 93.2% of people. The obtained results are discussed and some conclusion, based upon the average values, was drawn.

1. Introduction

The death in London of the former secret service agent Alexander Livtinenko on 23 November 2006 due to a lethal intake of ^{210}Po, presumably via ingestion, sparked renewed interest in the field of ^{210}Po toxicity to humans (Editorial, 2007; Harrison et al., 2007; Stather, 2007).

^{210}Po occurs widely in nature and is an important component of man's natural radiation background (Blanchard, 1967; Parfenov, 1974; UNSCEAR, 1977). The main route of ^{210}Po intake by the human body is the ingestion with foodstuffs, other studies reported that smoking also represents a significant route (Al-Arifi et al., 2006; Khater, 2004; Little, 1965; Little and McGandy, 1966; Parfenov, 1974; Skwarzec et al., 2001). Ingestion with drinking water, especially of underground origin represents another route of ^{210}Po intakes. Inhalation of ^{222}Rn released from the soil also contributes in ^{210}Po body burden. The absorption coefficient of ^{210}Po into blood from the digestive tract was estimated to be 35%, and the excretion of this radionuclide from the body in the urine was 14–15 times less than faeces (Parfenov, 1974). However, the body burden of ^{210}Po in normal human body may differ from one person to another depending upon the mode life including diet habits, origin of drinking water, residence place which effects radon exposure rate and also smoking habits as smoking rate, smoking period and the type of the smoking material. Therefore, many factors may affect the ^{210}Po intake and lead to variations in the body burden in different individuals (Editorial, 2007; Harrison et al., 2007; Parfenov, 1974; Stather, 2007; UNSCEAR, 1988).

Due to the large variance in the estimates of ^{210}Po content among different populations, the prospective of this article is limited to the evaluation of the background level of the ^{210}Po excretion rate in the urines of people living in an area of Central Italy (near the Republic of S. Marino) where the Urbino University is located and to compare the results with data reported by other authors.

Different methods can be used for assessing ^{210}Po according to the objectives of the measurement. There are several different analytical techniques available, which are complementary, and fit for their purposes. Generally the greater the precision and sensitivity needed, the longer the process will take because of the need for chemical processing to separate and concentrate the polonium, and to allow time for a sufficient number of radioactive decays to take place.

For our purpose, to evaluate the low exposure of the members of the public, a sensitive method is needed. The method used has been adapted from one that is in routine operational use for measurements on environmental samples, e.g. food, water, sediments. It is therefore capable of measuring natural levels of ^{210}Po in many types of sample, including urine and it requires 2–3 days from receipt of a 24-h sample. This method was used by the Radiation Protection Division of the Health Protection Agency (HPA) after the poisoning incident in London to assess the level of ^{210}Po in urines of members of public and various employees who may contact with a contaminated person or location (HPA 2007-1, 2007; HPA 2007-2, 2007). It is

* Corresponding author. Tel.: +39 0722 303307; fax: +39 0722 303306.
E-mail address: donatella.desideri@uniurb.it (D. Desideri).

0265-931X/$ – see front matter © 2009 Elsevier Ltd. All rights reserved.
doi:10.1016/j.jenvrad.2008.11.004

also a rapid method for dose assessment purposes. In fact, from the amount of ^{210}Po excreted in the urine, by well-established methods, it is possible to calculate the amount of ^{210}Po that originally entered the body, and hence, using the appropriate dose conversion factors, the resulting radiation dose. The dose assessment methods generally adopted for use around the world are those developed by the ICRP over the last half-century or so (ICRP, 1994, 1995, 1996, 2007).

This is not the only method available and others laboratories are using different approach, as, for instance, a direct measurement of ^{210}Po by an ultra-low-level scintillation counter with an alpha/beta discrimination device (Anversa et al., 2007).

A validation check of the method was carried out by the participation to an international intercomparison organized by the International Atomic Energy Agency (IAEA, Vienna).

2. Material and methods

2.1. Sampling

132 samples of urine were analyzed of people living in the area of Central Italy (near to the Republic of S. Marino, Adriatic coast) where the Urbino University is located.

People were chosen taking into account the following parameters: age, sex and smoke. People age was dispersed along a wide range (from 5 to over 60 years) and the volunteers were subdivided in five age classes (5–15 years, 16–30 years, 31–45 years, 46–60 years and over 60 years).

59 males and 73 females had been participated in this study; 39 were cigarette smokers and 67 non-smokers who had never smoked. The continuous smoking individuals were identified as smokers whereas the irregularly smoking individuals were excluded from this study.

2.2. Pretreatment

The urine of 24 h was collected in a 2 l plastic bag and their volume was exactly measured. After addition to 0.5 l of urine of a known quantity of ^{209}Po as internal standard yield tracer and of 50 ml of conc. HNO_3, the sample was stirred and heated on a hot plate at 150 °C, evaporated until 25 ml and transferred to a suitable beaker. Sequential treatments with conc. HNO_3 and H_2O_2 were performed to destroy the organic material and release free polonium ions into the solution. Then the solution was evaporated to dryness and the residue was treated three times with conc. HCl at controlled temperature of 85–90 °C to ensure a complete nitrate removal. Finally the residue was dissolved with warming in 120 ml of 1 M HCl and filtered.

2.3. Radioanalytical method

^{210}Po is not a gamma emitter: it emits only alpha particles at 5.407 MeV, consequently, it is generally determined by alpha spectrometry.

Polonium was deposited at controlled temperature of 85–90 °C and at pH 1.5–2.0 in continuous for 4 h on a silver disk, placed in a syringe and immersed into a 200 ml of 1 M HCl solution containing 10 ml of 20% hydroxylamine hydrochloride and 10 ml of 25% sodium citrate. The silver disk was measured by α-spectrometry. No preliminary separation was required and essentially quantitative recoveries were obtained by using standard ^{209}Po tracer (Desideri et al., 2007; Murray et al., 2007; Jia et al., 2000).

The source was counted using high resolution alpha ray spectrometry system with silicon detectors (Canberra, USA) counting the source for 24 h. The mean counting efficiency was 31.7 ± 3.1% and the background was approximately 2×10^{-6} s^{-1} in the energy region of interest. A motor-driven vacuum pump provided adequate evacuation of the vacuum chambers of the system. The final result was reported as excretion rate (mBq day^{-1}) multiplying the ^{210}Po activity concentration (mBq l^{-1}) by the volume of urine excreted in a day.

The mean chemical yield was 60.1 ± 14.2% and the detection limit resulted to be 0.2 mBq day^{-1}. Fig. 1 shows an alpha spectrum of ^{210}Po separated of a urine sample.

The uncertainties given with the final results are one standard deviation resulting from the propagation of all random counting uncertainties occurred anywhere in the entire measurement process.

2.4. Analytical quality control

Analytical quality control of this radioanalytical method was carried out by different ways: a) addition of ^{209}Po (En. alpha = 4.976 MeV) as the yield tracer: this fact and the analysis of the relevant alpha spectrum permitted to obtain a precise figure for the recovery of any measure; b) participation to an international intercomparison to checked the accuracy and the reproducibility of the method: following the recent event of ^{210}Po poisoning, the Chemistry Unit of the International Atomic Energy Agency (IAEA) of Seibersdorf (Vienna) organized a proficiency test on the determination of ^{210}Po in acidified water. The aim of this interlaboratory exercise was to gather information on the current state of practice for ^{210}Po measurements at different activity levels in five liquid samples. Emphasis in this proficiency test was placed on both the accuracy and the evaluation of analytical uncertainty. In the final evaluation, both scores for trueness and precision were combined. A result was defined "acceptable" when it obtained "acceptable" score in both criteria. The samples 01–04 contained a ^{210}Po concentration activity in the range 1–10 Bq l^{-1}. Sample 05 was evaluated to check if any "false positive" was reported. If it presented a ^{210}Po concentration activity < 0.1 Bq l^{-1}, it obtained "pass" as final score. Table 1 shows the results.

3. Results and discussion

The results of ^{210}Po excretion rate in urine samples are illustrated in Figs. 2–4. 93.2% of samples resulted in <30 mBq day^{-1}. In the monitoring program of HPA (UK) of March 2007 on 741 urine samples tested, 81.0% were <30 mBq day^{-1}. HPA has judged this level of 30 mBq day^{-1} to be one at which there will certainly be some activity above the natural background ^{210}Po level, which typically falls in the range of about 5–15 mBq day^{-1} (HPA 2007-1,

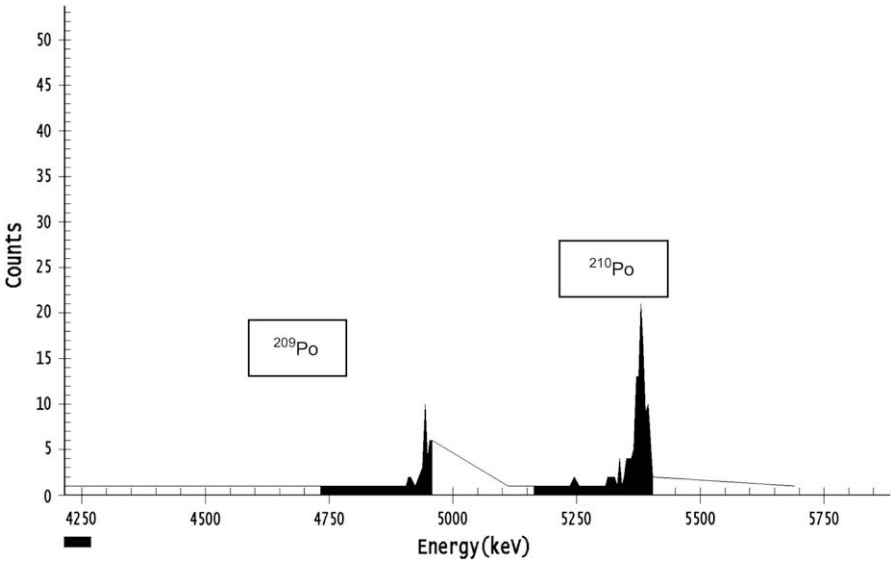

Fig. 1. Alpha spectrum of polonium separated from a urine sample.