Science of the Total Environment 472 (2014) 421–424

Contents lists available at ScienceDirect

Science of the Total Environment

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

Exposure to radionuclides in smoke from vegetation fires

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HIGHLIGHTS

- Natural radionuclides in vegetation are in low concentrations.
- Forest fires release natural radionuclides from vegetation and concentrate them in inhalable ash particles.
- Prolonged inhalation of smoke from forest fires gives rise enhanced radiation exposure of lungs especially due to polonium.
- Respiratory protection of fire fighters and members of public is highly recommended for radioprotection reasons.

ARTICLE INFO

Article history:
Received 30 July 2013
Received in revised form 4 November 2013
Accepted 13 November 2013
Available online 1 December 2013

Keywords:
Forest fires
Radioactivity
Polonium
Inhalation
Radiation dose

ABSTRACT

Naturally occurring radionuclides of uranium, thorium, radium, lead and polonium were determined in bushes and trees and in the smoke from summer forest fires. Activity concentrations of radionuclides in smoke particles were much enriched when compared to original vegetation. Polonium-210 (^{210}Po) in smoke was measured in concentrations much higher than all other radionuclides, reaching 7255 ± 285 Bq kg^{-1}, mostly associated with the smaller size smoke particles (<1.0 μm). Depending on smoke particle concentration, ^{210}Po in surface air near forest fires displayed volume concentrations up to 70 mBq m^{-3}, while in smoke-free air ^{210}Po concentration was about 30 μBq m^{-3}. The estimated absorbed radiation dose to an adult member of the public or a firefighter exposed for 24 h to inhalation of smoke near forest fires could exceed 5 μSv per day, i.e, more than 2000 times above the radiation dose from background radioactivity in surface air, and also higher than the radiation dose from ^{210}Po inhalation in a chronic cigarette smoker. It is concluded that prolonged exposure to smoke allows for enhanced inhalation of radionuclides associated with smoke particles. Due to high radiotoxicity of alpha emitting radionuclides, and in particular of ^{210}Po, the protection of respiratory tract of fire fighters is strongly recommended.

1. Introduction

Radionuclides of uranium (^{228}U), thorium (^{232}Th) and actinium (^{235}U) natural radioactive decay series, and non-series radionuclides such as ^{40}K, are present everywhere in the environment (Eisenbud and Gesell, 1997). These radionuclides occur in vegetation together with many other chemical elements absorbed by plants from soils and rocks. In addition to these naturally occurring radionuclides, artificial radionuclides originating in atmospheric nuclear weapon tests and nuclear accidents (UNSCEAR, 2000) are also present. Once released into the atmosphere, such artificial radionuclides were rapidly dispersed by atmospheric circulation and deposited onto vegetation and soils in vast areas around the globe, as recently observed again following the nuclear power plant accident in Fukushima, Japan (Masson et al., 2011; Carvalho et al., 2012). In Europe, this radioactive fallout was particularly noticeable after the nuclear accident of Chernobyl in April 1986, which caused heavy radioactive depositions particularly on forests in Belorussia and Ukraine (Bard et al., 1997; Paatero et al., 2009). Several years later, extensive forest fires in that region resuspended the radioactive cesium (^{137}Cs) from Chernobyl depositions into the atmosphere. This resuspension of ^{137}Cs was detected and measured in aerosols across western European countries and caused some alarm (Paatero et al., 2009).

Much less attention has been paid to the naturally occurring radionuclides present in vegetation, which are released also into the atmosphere by bush and forest fires, although the health impact of fine and ultrafine particle inhalation has been demonstrated (Wichmann and Peters, 2000; Lazaridis et al., 2008; Vos et al., 2009; Carvalho et al., 2011a; Bae et al., 2014). Combustion of plant biomass may be able to enhance the pathways and biochemical availability of those radionuclides to human beings. During the summer period, thousands of hectares are destroyed every year by forest and bushfires around the Mediterranean basin. The concentrations of radionuclides in surface air were determined in the vicinity of vegetation fires to assess their radiological impact on fire fighters and members of the public. A preliminary radiological assessment is presented herein.

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http://dx.doi.org/10.1016/j.scitotenv.2013.11.073

2. Materials and methods

Determination of radionuclides focused on a selection of the more common naturally-occurring alpha and beta emitters such as uranium (^{238}U, ^{235}U, ^{234}U) and thorium (^{232}Th, ^{230}Th) isotopes, radium (^{226}Ra), lead (^{210}Pb) and polonium (^{210}Po) that generally are the main contributors to the absorbed radiation dose in humans (UNSCEAR, 2000). Concentrations of these radionuclides were determined in vegetation samples and in smoke from vegetation fires, which were sampled in close collaboration with fire brigades in the Viseu region, Centre — North of Portugal, during late summer 2012.

Vegetation samples, such as tree trunk wood and leaves, were collected in the absence of fires, samples brought into the laboratory, oven-dried at 60 °C in air, milled, and the dry powder homogenized for analysis.

The sampling of smoke from wild fires was performed in the field, close to bush and forest fires. The temperature of flames, in each locale at the time of sampling, was measured with a hand held infra-red thermometer with laser pointer (Omega, UK). Air sampling was conducted near flames, to obtain freshly produced smoke particles. For this were used portable battery powered aerosol samplers (F&J Specialty, USA), with microfiber glass filters (Whatman, 50 mm in diameter), and a moderate air flux of 60 L min^{-1}. Larger smoke samples, were obtained using stand-alone large volume samplers (F&J Specialty, USA), powered by an electric generator (Yamaha, 10kVA), with microfiber glass filters 110 mm in diameter and an air flux of ca. 1400 L min^{-1}. Other large volume air samplers mounted on a tripod (Andersen), were used with microfiber glass filters 20 cm × 20 cm, and air flux of ca. 1600 L min^{-1}.

The concentration of radionuclides in six size classes of aerosol particles was determined on samples obtained with a Cascade Impactor air sampler (Andersen), filtering 1100 m^3 total volume per sample with an average flow rate of about 1370 L min^{-1}. The Cascade Impactor allowed the collection on Whatman filters of aerosol particles in size classes of] >7.6 μm];]7.6–3.2 μm],]3.2–1.6 μm],]1.6–1.0 μm],]1.0–0.5 μm], and] <0.5 μm].

All filters used for aerosol sampling were weighted before and after filtration, in a humidity and temperature controlled room, to determine the dry load. This was mostly composed of smoke particles from the vegetation fires. Afterwards, filters were used for determination of radionuclides.

The analysis of radionuclides was performed by radiochemistry and alpha spectrometry according to verified procedures (Oliveira and Carvalho, 2006; Carvalho and Oliveira, 2007). Briefly, isotopic tracers (^{232}U, ^{229}Th, ^{224}Ra, ^{209}Po, stable Pb) were added in known amounts to aliquots of samples, in order to be used as internal tracers for the determination of radiochemical yields. Plant and aerosol samples were dissolved in nitric and hydrochloric acids (3:1) and radioelements were separated and purified through ion exchange chromatography columns, both pre-packed (Eichrom) and prepared in the laboratory with Bio-Rad resins. Radioelements were then electroplated on either stainless-steel or silver disks. The alpha particle emission from the disks' surface was measured with ion-implanted silicon detectors in an alpha spectrometer (OctetePlus, EG&G Ortec). Uncertainties associated with the analytical results are the propagated uncertainties for the entire procedure and are given at 1σ significance level.

Quality assurance of the analytical methods was regularly checked by the analysis of IAEA certified reference materials and by participation in international analytical interlaboratory comparison exercises with good results (Carvalho and Oliveira, 2007).

3. Results and discussion

Determination of radionuclide concentrations in bush and tree samples showed that naturally occurring radionuclides are present in low concentrations, generally below 50 Bq kg^{-1} dry weight (dw) and often much lower than 10 Bq kg^{-1} (dw) in trunk wood, in leaves and other aerial plant structures (Table 1). Low radionuclide concentrations are generally expected in plants because these radioactive elements are neither essential elements nor oligo-elements with biochemical functions in plant metabolism and plant growth. Some plants are even classified as excluders due to their ability to prevent root absorption of some radioelements present in soils (Simon and Ibrahim, 1987). Notwithstanding, small amounts of radionuclides can be absorbed by root uptake and accumulated in plant tissues, such as verified particularly in uranium rich areas (Carvalho et al., 2009a, 2009b, 2011b; Gonçalves et al., 2010). Radionuclides formed in the atmosphere from the radioactive decay of atmospheric radon (^{222}Rn), also a member of the uranium series, are brought to the ground with dry and wet atmospheric depositions and thus are intercepted by plant aerial structures. These radionuclides in atmospheric depositions, such as the long lived radon daughters ^{210}Pb and ^{210}Po, can partly be accumulated in plants through foliar uptake (Simon and Ibrahim, 1987; Eisenbud and Gesell, 1997; IAEA, 2010). The low values of ^{210}Po/^{210}Pb activity concentration ratios, as often measured in plant leaves and other aerial plant structures reflect the concentration ratio of these two radon daughters in surface air and atmospheric depositions, typically around 0.1 (Carvalho, 1995; Carvalho et al., 2011a). However, in regions with frequent forest fires the ^{210}Po/^{210}Pb activity ratio in depositions intercepted by the vegetation may be much higher, as observed in results reported herein due to fires that took place in the region before our sampling (Table 1).

The naturally occurring radionuclides contained in vegetation, and in particular those in vegetables and fruits that are part of our diet,

Table 1
Radionuclide concentrations (Bq kg^{-1} dry weight) in plants, ashes from plant combustion, and in smoke free surface air aerosol for comparison, in Viseu district, Portugal, summer 2011.

Samples	^{238}U	^{235}U	^{234}U	^{230}Th	^{226}Ra	^{210}Pb	^{210}Po	^{232}Th
Cistus, bushes	0.56 ± 0.02	0.027 ± 0.003	0.56 ± 0.02	0.57 ± 0.03	2.1 ± 0.2	9.90 ± 0.35	12.0 ± 2.4	0.38 ± 0.02
Oak tree, trunk wood	24.5 ± 0.6	1.18 ± 0.04	24.2 ± 0.6	1.9 ± 0.1	4.9 ± 1.1	–	–	0.112 ± 0.008
Oak tree, trunk wood	22.6 ± 0.8	1.08 ± 0.05	21.8 ± 0.8	1.60 ± 0.07	5.4 ± 0.5	3.27 ± 0.16	5.51 ± 0.02	0.095 ± 0.007
Oak tree, leaves	1.68 ± 0.07	0.08 ± 0.01	1.56 ± 0.07	0.51 ± 0.04	7.4 ± 0.7	17.2 ± 0.4	30.8 ± 1.2	0.29 ± 0.02
Eucalyptus, trunk wood	0.114 ± 0.004	0.0050 ± 0.0006	0.115 ± 0.004	0.023 ± 0.002	0.96 ± 0.13	0.98 ± 0.03	1.68 ± 0.05	0.016 ± 0.001
Eucalyptus, bark	0.29 ± 0.01	0.014 ± 0.003	0.30 ± 0.01	0.089 ± 0.008	26.0 ± 2.4	1.88 ± 0.09	2.60 ± 0.06	0.049 ± 0.007
Eucalyptus, leaves	4.4 ± 0.1	0.21 ± 0.01	4.1 ± 0.1	0.20 ± 0.02	37.3 ± 2.6	10.3 ± 0.4	49.4 ± 2.3	0.08 ± 0.01
Acacia tree, trunk wood	0.020 ± 0.001	0.0014 ± 0.0004	0.018 ± 0.001	0.017 ± 0.001	1.5 ± 0.1	2.04 ± 0.05	4.05 ± 0.15	0.008 ± 0.001
Acacia tree, leaves	13.0 ± 0.3	0.58 ± 0.02	12.8 ± 0.3	5.3 ± 0.3	30.8 ± 1.6	20.27 ± 0.47	8.61 ± 0.33	0.16 ± 0.02
Pine tree, trunk wood	0.105 ± 0.006	0.006 ± 0.002	0.112 ± 0.006	0.067 ± 0.006	1.14 ± 0.06	1.98 ± 0.09	0.97 ± 0.03	0.065 ± 0.006
Pine tree, trunk wood	0.103 ± 0.013	0.0050 ± 0.0047	0.106 ± 0.013	0.014 ± 0.003	0.92 ± 0.09	1.43 ± 0.13	1.53 ± 0.06	0.009 ± 0.003
Pine tree, bark	0.42 ± 0.02	0.019 ± 0.003	0.42 ± 0.02	–	2.7 ± 0.5	2.80 ± 0.08	2.87 ± 0.06	–
Pine tree, leaves (needles)	1.99 ± 0.07	0.13 ± 0.01	1.91 ± 0.07	2.1 ± 0.1	4.9 ± 0.3	10.36 ± 0.31	3.10 ± 0.08	1.10 ± 0.08
Ashes from ground after forest fire	135 ± 4	6.2 ± 0.3	146 ± 4	259 ± 20	477 ± 54	402 ± 6	1115 ± 66	55.7 ± 4.5
Fly ashes (in surface air collected on filter F#6)	347 ± 19	18.0 ± 3.9	372 ± 20	209 ± 13	6144 ± 2908	2070 ± 88	7255 ± 285	203 ± 13
Fly ashes (in surface air collected on filter F#11)	224 ± 12	9.8 ± 2.6	227 ± 12	261 ± 14	5763 ± 1489	923 ± 53	3604 ± 148	95.7 ± 7.1
Aerosol (surface air in absence of fire smoke)	71.0 ± 3.7	2.8 ± 1.0	71.8 ± 3.7	42.4 ± 2.6	–	5895 ± 218	111 ± 7	42.1 ± 2.7