

## Forest Fires and Resuspension of Radionuclides into the Atmosphere

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**Abstract: Problem statement:** Forest fires are especially frequent around the Mediterranean Sea basin in the summer period and might be able to release naturally-occurring and man-made radionuclides from plant biomass and inject them into the atmosphere. The impact of this radioactivity on populations was not investigated before. **Approach:** Radionuclide analysis was performed in plants, in smoke from plant burning and in cigarette smoke to determine radionuclide concentrations by alpha spectrometry. **Results:** Concentrations of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in trees such as olive trees, showed low concentrations in roots, trunk and leaves and minor translocation of radionuclides from the root to aerial parts. Soil to plant transfer ratios for  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in several plants were in the range from  $10^{-4}$  to  $10^{-2}$ . Radionuclides from atmospheric depositions may be accumulated in plants by foliar uptake and for  $^{210}\text{Pb}$  this seems the main pathway, with plant aerial parts displaying  $^{210}\text{Po}/^{210}\text{Pb}$  ratios around 0.1, which is similar to the radionuclide ratios determined in atmospheric depositions. Experimental burning of wood from several tree species showed enhanced radionuclide concentrations in smoke compared to plant materials. Investigation of  $^{210}\text{Po}$  release from tobacco leaves used in cigarettes, showed especially enhanced concentrations of this radionuclide in the cigarette smoke particles. **Conclusion:** Radionuclide concentrations in cigarette smoke expose the lung tissues of regular smokers to high concentrations of  $^{210}\text{Po}$  that were considered carcinogenic. Although radionuclide concentrations in other plants analyzed were generally lower than in tobacco, globally the radionuclide activity in the plant biomass is elevated. Inhaled smoke particles from forest fires are likely to contribute to enhanced radiation doses in the human lung.

**Key words:** Naturally-occurring radionuclides, polonium, radioactivity monitoring, forest fires, atmospheric depositions, lung tissues, smoke particles, cigarette smoke, natural radionuclides, regular smokers

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### INTRODUCTION

Naturally-occurring and artificial radionuclides are present in the vegetation cover as elsewhere in the Planet (Eisenbud and Gesell, 1997; UNSCEAR, 2008). Natural radionuclides, such as those of the uranium radioactive decay series, are absorbed from soils and irrigation water by root uptake and incorporated into plant structures (IAEA, 2009; Carvalho *et al.*, 2009). Some natural radionuclides such as the radon daughters  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  may be absorbed also from surface air by foliar uptake. Their concentrations in plants have been extensively monitored in regions of normal and high radioactive background around the world and their contribution through the ingestion pathway to the internal radiation dose in humans has been assessed (UNSCEAR, 2000; 2008; 2009).

Artificial radionuclides were dispersed worldwide, especially after the nuclear weapon tests in the atmosphere during the 1950s and 60s and after the Chernobyl nuclear accident in 1986. Following the injection of radioactive particles into the atmosphere, the radioactive atmospheric fallout added artificial radionuclides, such as  $^{137}\text{Cs}$  and  $^{239+240}\text{Pu}$ , to the vegetation cover and surface soils in many regions of the globe. Once deposited on the Earth surface and incorporated in soils and plants, can human populations be exposed to these radionuclides? The answer to this question is of the utmost importance to the radiation safety of human populations, in particular when high radioactivity surface deposits do exist such as those in Ukraine and Belorussia near the Chernobyl area (UNSCEAR, 2008).

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Periodic atmospheric radioactivity monitoring performed at several western European countries detected  $^{137}\text{Cs}$  peaks in surface air in the summer of 2002 and afterwards (Masson *et al.*, 2008). In the absence of known accidental radioactivity releases, it was interpreted as re-suspension by summer fires of the  $^{137}\text{Cs}$  from Chernobyl accident deposited in Ukraine and Belorussia forests and soils (Amiro *et al.*, 1996; Paatero *et al.*, 2009). Besides the artificial  $^{137}\text{Cs}$ , vegetation also contains the common naturally-occurring radionuclides such as those of uranium and thorium decay series. In Europe, forest and vegetation fires are very frequent in summer, particularly around the Mediterranean Sea basin.

We hypothesized that the activity concentrations of these naturally-occurring radionuclides in surface air could be enhanced by the release of radionuclides from the vegetation cover by biomass fires. This study shows the first results of the investigation on the natural radionuclides released from plant material by fire.

## MATERIALS AND METHODS

To assess the concentration of the common naturally occurring radionuclides in the vegetation cover, smoke particles released by the combustion in the air of common plants were analyzed. Plant samples including trees (oak tree, pine trees, eucalyptus and acacia) and crops (pasture, maize) that were collected in the central region of Portugal in areas with normal background radioactivity levels. Alpha emitting radionuclides were analyzed in the smoke collected on pre-weighted micro fiber glass filters Whatman (average pore size 0.7  $\mu\text{m}$ ) using suction pumps.

In order to better understand the release of radionuclides from plants by the combustion and in particular of the radionuclide  $^{210}\text{Po}$ , we investigated the release of radionuclides from burning tobacco into cigarette smoke. Cigarettes were used to investigate in more detail the  $^{210}\text{Po}$  radioactivity partitioning and balance from the initial tobacco to smoke particles, residual ashes and to make an assessment of radioactivity exposure through smoke inhalation.

The filters with weighted amounts of smoke particles were analyzed starting with the addition of isotopic tracers for use as radiochemical yield tracers, followed by sample dissolution in HCl and  $\text{HNO}_3$  acids and radiochemical separation of radioelements. Each radioelement of interest was then electroplated on the surface of metal discs, made either of stainless steel or silver foil and discs counted by alpha spectrometry using an OCTECTEPlus (ORTEC-EG&G) spectrometer equipped with 450  $\text{mm}^2$  ion implanted

silicon detectors. Radiochemical procedures and alpha spectrometry detection techniques were validated and described in detail elsewhere (Oliveira and Carvalho, 2006; Carvalho and Oliveira, 2007).

Results are given in  $\text{Bq kg}^{-1}$  and wet to dry ratios are given to allow conversion of units.

## RESULTS AND DISCUSSION

Analyses of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ , for example in olive trees, showed their presence in roots, tree trunk and leaves, although the translocation of radionuclides from the root to aerial parts seems to occur in a very limited extent (Table 1). Transfer factors from soil to plants ( $\text{TF} = (\text{Bq kg}^{-1} \text{ in the plant}) / (\text{Bq kg}^{-1} \text{ in the soil})$ ) by root uptake typically were around  $10^{-3}$  (range  $10^{-4}$ - $10^{-2}$ ) for  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ , but may vary with soil and plant type (Bettencourt *et al.*, 1988; Carvalho *et al.*, 2009). Another route of radionuclide uptake by plants is the foliar uptake from atmospheric depositions, which seems the main pathway for most  $^{210}\text{Pb}$  accumulated in plants. In aerial vegetation (e.g., cabbage leaves and tree leaves)  $^{210}\text{Po}/^{210}\text{Pb}$  ratio is similar to the  $^{210}\text{Po}/^{210}\text{Pb}$  ratio in atmospheric depositions, about 0.1, while in soils  $^{210}\text{Po}/^{210}\text{Pb}$  ratio is about 1 usually (Carvalho, 1995).

These results showed the presence of measurable activity concentrations of key uranium daughter radionuclides in common plants. It was hypothesized that with combustion in the air the radioactivity in plants would be released from leaves and wood and likely carried with smoke particles released into the atmosphere.

Table 2 shows results of radionuclide activity concentrations measured in smoke particles obtained from the air combustion of several wood types. It may be noticed that there was a systematic enrichment of  $^{210}\text{Po}$  relative to  $^{210}\text{Pb}$  (1-3x) in smoke particles, reversing the radionuclide ratios in tree wood that displayed  $^{210}\text{Po}/^{210}\text{Pb} < 1$ .

Table 1: Activity concentrations of the natural uranium series in vegetation ( $\text{Bq kg}^{-1}$  wet weight) and in soil ( $\text{Bq kg}^{-1}$  dry weight). Average dry: wet weight ratios were 0.23 in the vegetables and 0.42 in the tree samples. Analytical uncertainty, as percent standard error, is about 5-10% of the activity concentrations given

Plant	$^{210}\text{Pb}$	$^{210}\text{Po}$	$^{210}\text{Po}/^{210}\text{Pb}$
Cabbage leaves	0.10	0.01	0.10
Maize, aerial parts	0.07	0.05	0.70
Olive tree, leaves	9.40	1.05	0.10
Olive tree, trunk wood	4.30	0.14	0.03
Olive tree, roots	1.40	1.10	0.80
Palm tree, leaves	5.40	1.07	0.20
Palm tree, bark	0.33	0.09	0.27
Tobacco leaves (cured)	5.00	4.70	0.90
Soil (0-30 cm)	100.00	100.00	1.00

Table 2: Radionuclide activity concentrations (Bq kg<sup>-1</sup> dry weight) in smoke from combustion of several wood qualities. Analytical uncertainty, as percent standard error, is about 5-10% of the activity concentrations given

Wood	<sup>238</sup> U	<sup>234</sup> U	<sup>230</sup> Th	<sup>226</sup> Ra	<sup>210</sup> Pb	<sup>210</sup> Po
Eucalyptus	65	88	40	318	52	125
Pine tree	<7	2	38	98	277	460
Oak tree	10	12	14	12	266	265
Acacia	11	36	68	505	440	610

Table 3: Polonium (<sup>210</sup>Po) in cigarette smoke and in ashes, for three commercial cigarette brands (UNSCEAR 2008)

Brand (g of tobacco per cigarette)	<sup>210</sup> Po activity per cigarette						Total mBq	210Po inhalation (mBq per 20 cigarettes)
	Main stream smoke		Residue (ash+butt)		Side stream smoke			
	mBq	Total (%)	mBq	Total (%)	mBq	Total (%)		
A (0.748 g)	1.52	5.2	2.1	7.2	24.4	84	28.9	30
B (0.793 g)	1.37	11.2	4.9	40.0	5.9	49	12.2	27
C (0.940 g)	0.97	37.3	0.3	11.5	1.3	51	2.6	19

Ra-226 was also high in smoke particles, but uranium and thorium isotopes were generally present in low concentrations. This suggests that <sup>210</sup>Po was more efficiently released from the plant material and condensed in the smoke particles, these ones becoming more enriched in this radionuclide than in <sup>210</sup>Pb.

Uranium and thorium are the least volatile elements and probably, larger activities would remain associated with the ash residues than with smoke particles. Comparing the activity concentrations of <sup>210</sup>Po and <sup>210</sup>Pb in the smoke particles (Table 2) with activity concentrations in plants (Table 1), results suggest that these radionuclides are enhanced by a factor of 10-100 times in the smoke in comparison with the initial material. Cigarette smoke was used as a test material. Most of the cigarette weight is made with tobacco plant leaves. In the cigarettes of several brands, it was verified that tobacco always contained naturally-occurring radionuclides, including <sup>210</sup>Po in activity concentrations averaging 19 (2.7-38.6) Bq kg<sup>-1</sup> dry weight (Table 1). In all cigarette brands, the cigarette combustion showed easy volatilization of <sup>210</sup>Po and transfer of this radionuclide to smoke particles. Po-210 was easily measured in the main stream smoke aspired through the cigarette filter or butt, in the side stream smoke dispersed into the atmosphere and in the combustion remains, i.e., ash and butts (Table 3). The efficient release of <sup>210</sup>Po from the tobacco leaf is due to the low polonium volatilization point (180°C) in comparison with the combustion temperature of cigarettes (600-800°C). The volatilization of polonium facilitates the release of this radionuclide from the tobacco and its association by condensation and transport with the smoke particles. It is suggested that volatilization of other radionuclides, all with volatilization points at much higher temperatures, is only partial and thus, their release from

the plant matrix and transfer to the smoke is smaller than with <sup>210</sup>Po.

Inhalation of naturally-occurring radionuclides with airborne particles (atmospheric dust mainly charged with radon daughters) occurs and gives a contribution to the radiation exposure of humans. Background levels of <sup>210</sup>Po and <sup>210</sup>Pb in the atmospheric particles inhaled and absorbed through the lungs, contribute to the absorption of <sup>210</sup>Po and <sup>210</sup>Pb into the blood through gut absorption of radio nuclides ingested with food. Average concentrations of <sup>210</sup>Po and <sup>210</sup>Pb in the surface air at Lisbon were determined at 31× 10<sup>-6</sup> Bq m<sup>-3</sup> and 181×10<sup>-6</sup> Bq m<sup>-3</sup>, respectively. Assuming a daily inhalation of 20 m<sup>3</sup> of air (Standard Man, as defined by the ICRP), the inhalation rate of these radio nuclides in this region is 6.2×10<sup>-4</sup> Bq d<sup>-1</sup> and 3.6×10<sup>-3</sup> Bq d<sup>-1</sup>, respectively. These are radionuclide inhalation rates at natural background levels in outdoor air.

Inhalation of cigarette smoke is a supplementary exposure to <sup>210</sup>Po radionuclide. The consumption of one pack of 20 cigarettes per day based on results shown above (Table 3), gives rise to a <sup>210</sup>Po inhalation rate up to 3×10<sup>-2</sup> Bq d<sup>-1</sup>, i.e., two orders of magnitude higher than background levels. Interestingly, this radioactivity inhalation rate is roughly similar for the main stream smoke (active smoker) and for the side stream smoke (passive smoker). The radiation dose imparted to the lung by <sup>210</sup>Po in the cigarette smoke was assessed to increase noticeably the risk of lung cancer (Carvalho and Oliveira, 2006).

The activity concentration of <sup>210</sup>Po in the cured tobacco leaves seems higher than in many other plant materials. Notwithstanding, the processes involved in the release of radionuclides by plant combustion are likely the same. The analyses of radionuclides in smoke from vegetation fires (Table 2) provided additional evidence on the enhancement of radionuclides in smoke

particles from combustion of common forest plants and trees. A significant amount of radionuclides of the natural radioactive decay series, is accumulated in plants and in spite of the low activity concentration of each radionuclide, altogether the activity in biomass is high. This activity may be released to the atmosphere by forest and vegetation fires.

It is hypothesized that repeated and prolonged exposure to smoke from forest and vegetation fires might also increase the risk of absorbed radiation dose from ionizing radiations to the general population.

### CONCLUSION

Forest fires definitely release naturally-occurring radionuclides accumulated in the vegetation to the atmosphere. Smoke particles from plant combustion contain radionuclides in activity concentrations much higher than in the original wood and leaves, likely due to volatilization and condensation of radionuclides onto the smoke particles. Concentrations of the alpha emitting radionuclide  $^{210}\text{Po}$  are especially enhanced in the smoke, in comparison with other radionuclides. To a certain extent, the inhalation of smoke from biomass burning can be compared to the inhalation of cigarette smoke, which is known to contribute to increased lung exposure to ionizing radiation. Further study is planned to assess this radiological risk in field conditions.

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