

Polonium in size fractionated mainstream cigarette smoke, predicted deposition and associated internal radiation dose



M. Tiwari, S.K. Sahu, R.C. Bhangare, G.G. Pandit*

Environmental Monitoring and Assessment Section, Health Safety and Environment Group, Bhabha Atomic Research Centre, Trombay, Mumbai, 400085, India

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ABSTRACT

In this study, size fractionated mass and ^{210}Po activity concentrations in mainstream cigarette smoke (MCS) were monitored for three popular cigarette brands. Size segregated collection of MCS was carried out using a cascade type impactor, while mass and ^{210}Po activity concentration were analyzed gravimetrically and alpha spectrometry (following the radiochemical separation) respectively. Multiple-Path Particle Dosimetry (MPPD V2.11) model is used for prediction of deposition fraction calculations for the MCS deposition in different compartment of human respiratory tract. The activity concentration of ^{210}Po is found 10.56 ± 2.46 mBq per cigarette for the tested cigarette brands. ^{210}Po size distribution indicates most of this associates with fine fraction ($D_p < 2.23 \mu\text{m}$) of cigarette smoke. The committed annual effective dose to smokers (smoking on an average 20 cigarette a day), considering the ^{210}Po and ^{210}Pb concentrations (assuming it is in secular equilibrium with ^{210}Po) in MCS, was estimated between 0.22 and 0.40 mSv, with mean value of 0.30 mSv for tested cigarette brands. Considering the risk factor of fatal cancer due to radiation exposure of lung (exposure time of 30 years); the average collective estimated fatal cancer risk is estimated as 1.5×10^{-4} due to ^{210}Po and ^{210}Pb exposure to smokers.

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1. Introduction

Polonium (^{210}Po) is a member of the ^{238}U decay series and one of the relatively long-lived radionuclides ($t_{1/2} = 138$ days) of radon decay products. ^{210}Po is an alpha-emitting ($E_{\text{max}} = 5.304$ MeV) radionuclide and is ubiquitously present in trace amounts in most plants and foodstuffs as well as in human tissues and environmental compartments. Almost 90% of pulmonary cancer death cases can be assigned to smoking, the carcinogenic effect of which is well-known (CDC, 2000). The concentrations of ^{210}Po in tobacco used in cigarette are in the range of $2.8\text{--}37$ Bq kg^{-1} and vary with the cigarette brand, probably due to the different varieties of tobacco used and to different manufacturing procedures (Khater, 2004). The relatively high concentration of ^{210}Po and ^{210}Pb in tobacco can be explained by uptake from soil through the roots and by the adsorption of aerosols of ^{210}Po and ^{210}Pb content through the surface of the tobacco leaves (Martell, 1974; Skwarzec et al., 2001). In case of uptake through the roots, the high ^{210}Po and ^{210}Pb concentration may be caused by the high radionuclide content of the

soil, but the high ^{210}Po and ^{210}Pb concentration of the applied phosphate fertilizers may also cause a significant increase in their levels (Hussein, 1994). The sticking of atmospheric aerosols on the surface of leaves is enhanced by the hairs covering the leaves, which further incline the wide surface of the tobacco leaves (Martell, 1974).

The finding of earlier studies indicate that ^{210}Po in cigarettes is volatilized at the temperatures characteristic for burning cigarettes and inhaled into the lung along with the mainstream cigarette smoke (MCS). It might effectively be a relevant factor in the increased incidence of lung cancer among cigarettes smokers (Radford and Hunt, 1964). Earlier investigators have also studied both the sources and behavior of ^{210}Po and ^{210}Pb in relation to smoking, and the biological effects of these on lung tissues and other organs (Batarekh and Teherani, 1987; Shabana et al., 2000). ^{210}Po is highly radioactive and chemically toxic element. Direct damage occurs from energy absorption into tissues from alpha particles. The biological half-time (the time for the level of ^{210}Po in the body to fall to half) is approximately 50 days. If taken into the body, ^{210}Po is subsequently excreted, mostly through feces but some amount is excreted through urine and other pathways. People who come into contact with a person contaminated by ^{210}Po will

* Corresponding author.

E-mail address: ggp@barc.gov.in (G.G. Pandit).

not be at risk unless they ingest or inhale bodily fluids of the contaminated person (Seiler and Wiemels, 2012; IAEA, 2006).

^{210}Po found in the lungs of smokers are approximately threefold higher than that found in non-smokers, the effect on lung cancer risk has been questioned after comparison of these data with studies conducted on miners (Smith et al., 2000). Human's contamination occurs through inhalation of the air and ingestion of water and foods (mainly fish and sea-foods). Another important self-contamination source results from the cigarette smoking, since ^{210}Po and ^{210}Pb are present in inhaled mainstream smoke (Martell, 1975). Since the 1960s investigators have reported (Kilthau, 1996) that ^{210}Po and ^{210}Pb are present in the gaseous and particulate phases of tobacco smoke and contribute to cancer risk due to their deposition in the lungs. The nuclides are absorbed into the blood and distributed to the different body organs, thus increasing the local radiation exposure. About 10% of the ^{210}Po of cigarettes was found in the main stream smoke filter and about 18% in the butt (Mussealo-Rauhamaa and Jaakkola, 1985).

According to NCRP Report No. 95, (1987), deposition of ^{210}Po in the lungs of cigarette smokers results in an average annual dose equivalent of 0.16 Sv (16 rem) to the segmental bifurcations of the bronchial epithelium. This would yield an annual effective dose equivalent to the average smoker of about 13 mSv (1300 mrem); the corresponding average population effective dose equivalent would be 2.8 mSv y^{-1} (NCRP, 1987). There are also other radionuclide activity concentrations of ^{137}Cs , ^{238}U , ^{232}Th , ^4K and ^{90}Sr were reported in cigarette tobaccos. It was concluded that the average annual effective dose due to inhalation for adults (smokers) for ^{238}U , ^{232}Th , ^4K , ^{137}Cs , and ^{90}Sr are 28.04 $\mu\text{Sv y}^{-1}$, 138.65 $\mu\text{Sv y}^{-1}$, 139.83 nSv y^{-1} , 29.86 nSv y^{-1} , and 415.29 nSv y^{-1} respectively (Sogut et al., 2014).

In this study concentrations of ^{210}Po in mainstream cigarette smoke are measured in popular cigarette brands from India. Mass and ^{210}Po size distributions for MCS of those cigarette brands are reported. Size dependent deposition fractions were calculated for MCS for different compartment of respiratory tract using a mathematical model. Then, deposition fractions are used in deposition calculation of ^{210}Po in respiratory tract and further estimation of associated inhalation risk.

2. Materials and methods

2.1. Cigarette samples

Three popular cigarette brands in India were chosen for this study. The tar content of the cigarettes was in the range of 11–17 mg per cigarette. The tar deliveries are pack tar values which are generated using ISO puffing conditions. For avoiding marketing use of our study the trade-marks of the cigarettes brands have been omitted. Although they belongs to well-known and appreciated brands. Cigarettes used for these experiments were conditioned in a humidified chamber at 65% relative humidity (RH) at room temperature (23 °C) for at least 24 h prior to smoking. The cigarettes were smoked under the ambient laboratory conditions (45% RH, 24 °C). Mainstream smoke generated under a standard smoking protocol (60-s puff interval, 2-s puff duration, and 35-mL puff volume) was collected on impactor (VCCI). All the cigarettes were fitted with filter tips approximately 20 mm in length. The cigarettes were smoked to a butt length of 23 mm or to the length of the filter overwrap plus 3 mm. For measurable quantity of MCS mass and subsequent analysis of ^{210}Po ; 10 cigarettes were smoked in single experiment. Each brands experiment was repeated thrice for quality assurance.

2.2. Experimental setup

Setup for collection of size fractionated MCS is depicted in Fig. 1. The mainstream cigarette smoke was passed through a dilution chamber prior to being sampled by the variable configuration cascade impactor (VCCI). Particulate free nitrogen was used as dilution media and it did not contribute any particulate signature in VCCI. The setup was checked for leaks and the mass flow meter was calibrated using master meter (flow meter) with high accuracy. All flows were controlled with mass flow controllers (Model 2179A; MKS, Andover, MA) with accuracies > 1% of their operating flow rate. The sampling flow rate for smoke was kept 1.1 lpm; which is equivalent to smoker's suction rate while smoking. As cigarette smoke is hygroscopic in nature so dilution is applied as aerosol drying technique. The dilution chamber was flushed with particulate free air (using HEPA filters) between the measurements to decontaminate dilution chamber.

2.3. Variable configuration cascade impactor

Size fractionated mainstream cigarette smoke was collected using variable configuration cascade impactor (VCCI) (Tiwari et al., 2013). Glass fiber filter paper discs (EPM2000, Whatman) were used as an impaction surface for particles as well as a backup filter in the impactor. Filters papers were baked at high temperature in furnace more than 450 °C to remove any volatile impurities. Smoke was drawn through the impactor at a flow rate of 10 lpm which is specified for to collect cutoff diameter of cascade impactor stages. The size ranges (μm) collected from the different stages of cascade impactor were as >21.3, 21.3–15.1, 15.1–11.2, 11.2–7.38, 7.38–5.47, 5.47–2.23, 2.23–1.13, 1.13–0.75, 0.75–0.50, 0.50–0.30, 0.30–0.10 and 0.10. The pressure at stage seven was continuously measure through the course of sampling so the flow rate and stage cut off can be monitored.

2.4. Radiochemical separation and instrumentation

After collecting the size fractionated MCS, filter papers were digested in Teflon beakers using HF: HClO_4 : HNO_3 cocktail at 85 °C up to dryness in a temperature controlled water bath. HF: HClO_4 : HNO_3 cocktail is used; as that is capable of dissolving glass fiber, removing the organic contamination and bringing metals in elemental form. Then residues of samples were dissolved in 0.5 N HCl and filtered through whatman filter paper and make up (50 ml). The sample solution was heated to 80 °C and ^{210}Po was spontaneously deposited on a brightly polished silver planchette for a period of 2.5 h under stirring conditions. Prior to plancheting, in digested samples a pinch of ascorbic acid is also added to reduce

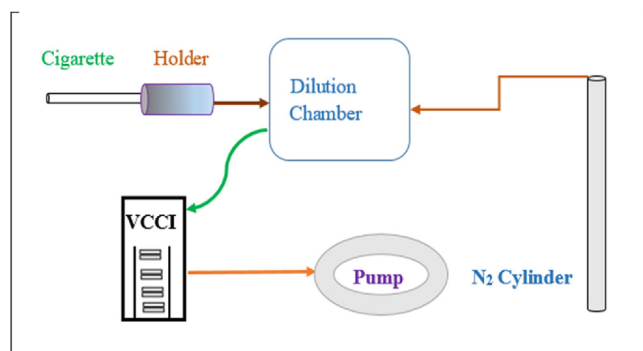


Fig. 1. Experimental set for collecting mainstream cigarette smoke (MCS) using variable configuration cascade impactor (VCCI).

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