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Effect of indoor-generated airborne particles on radon progeny dynamics

C. Vargas Trassierra[a], L. Stabile[a,*], F. Cardellini[b], L. Morawska[b], G. Buonanno[a,c]

[a] Department of Civil and Mechanical Engineering, University of Cassino and Southern Lazio, Cassino, FR, Italy
[b] National Institute of Ionizing Radiation Metrology (INMRI-ENEA), Rome, Italy
[c] International Laboratory for Air Quality and Health, Queensland University of Technology, Brisbane, Australia

HIGHLIGHTS

- Investigation of the interaction between particles and radon progeny dynamics.
- Measurements of particles emitted by different indoor sources.
- Tests performed in a controlled radon chamber.
- Particle size strongly influences the radon progeny dynamics.
- Particle surface area concentration is the key parameter of the radon-particle interaction.

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ABSTRACT

In order to investigate the interaction between radon progeny and particles, an experimental campaign was carried out in a radon chamber at the Italian National Institute of Ionizing Radiation Metrology, quantifying the amount of attached and unattached radon daughters present in air, as well as the equilibrium factor in the presence of particles generated through indoor sources. A fixed radon concentration was maintained, while particles were generated using incense sticks, mosquito coils and gas combustion. Aerosols were characterized in terms of particle concentrations and size distributions. Simultaneously, radon concentration and attached/unattached potential alpha energy concentration in the air were continuously monitored by two different devices, based on alpha spectroscopy techniques.

The presence of particles was found to affect the attached fraction of radon decay products, in such a way that the particles acted as a sink for radionuclides. In terms of sources which emit large particles (e.g. incense, mosquito coils), which greatly increase particle surface area concentrations, the Equilibrium Factor was found to double with respect to the background level before particle generation sessions. On the contrary, the radon decay product dynamics were not influenced by gas combustion processes, mainly due to the small surface area of the particles emitted.

1. Introduction

Indoor air quality represents a key aspect of the overall risk associated with exposure to airborne pollutants [1–4]. Indeed, indoor air quality is influenced by both (i) indoor generated pollutants not properly ex-filtrated through the building (due to the low air exchange rates [5,6]), and (ii) pollutants penetrating from outdoors. Therefore, indoor microenvironments cannot be considered safer than outdoor microenvironments, since indoor pollutant concentrations may reach higher levels than outdoor concentrations [7–9]. Moreover, people spend approximately 80–90% of their life in indoor microenvironments (houses, schools, workplaces, shopping centers, gyms), and therefore, the pollutant dose received by people in indoor microenvironments can be much higher than the dose received from time spent outdoors [10,11].

Radon (^{222}Rn) is a radioactive noble gas generated by the alpha decay of radium-226 (^{226}Ra) which is a radioactive element belonging to uranium-238 (^{238}U) decay chain. Uranium and its decay products are widely diffused in the earth crust and in building material such as tuff and “pozzolana” (or pozzolanic ash), a volcanic ash, both widely used in Central Italy. Radon has a relatively

* Corresponding author at: Department of Civil and Mechanical Engineering, University of Cassino and Southern Lazio, Via Di Biasio 43, 03043 Cassino, FR, Italy.
E-mail address: l.stabile@unicas.it (L. Stabile).

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long half-life (3.823 days) and it decays to polonium ^{218}Po (half life time 3 min) with the emission of an alpha particle. The other radon decay products (RDPs) are: lead-214 (^{214}Pb), beta emitter, half-life 26 min, bismuth-214 (^{214}Bi), beta emitter, half–life 19 min, and polonium-214 (^{214}Po), alpha emitter, half life 165 μs. Radon gas generated in soil or in building materials may diffuse through the soil and the wall and can accumulate in the air of dwellings and working places producing risk for human health [12].

The second most common radon isotope, ^{220}Rn, often referred as "thoron", belongs to the decay chain of thorium-232 (^{232}Th). This nuclide may be dangerous for human health too, but it has a very short half-life (55.8 s) and can reach the inhabited indoor environments only if ^{232}Th is present near the walls' surface.

Radon gas is known as the most important natural source of ionizing radiation and it is responsible for approximately 40% of the annual effective dose from all sources of radiation [13,14]. Epidemiological and toxicological studies have shown that long term exposure to radon increases the risk of lung cancer [15–19]. This risk is mainly associated with the abovementioned radon decay products ^{218}Po, ^{214}Pb, ^{214}Bi and ^{214} Po that are very reactive metallic ions [20,21]. RDPs dynamics in air may be summarized as follows: ^{218}Po generated by radon reacts in few seconds with water molecules and trace gases forming small clusters (whose diameter is in the range 0.5–1 nm) known as "unattached progeny" [22]. These clusters may either attach onto aerosol particles producing radioactive aerosol ("attached progeny") or, alternatively, plate out on walls and furniture. These two processes are in competition as take place on the time scale of few minutes. The rate of the two processes depends on several parameters such as aerosol concentration, room dimensions, and nature of the furniture surface. The permanence time of radioactive aerosol in air is of some hours like for normal aerosol. Attached and unattached progeny can be inhaled and then deposited in the deepest regions of the respiratory system, which is then exposed to alpha radiation emitted by radon daughters. Therefore, airborne particles represent one of the most important parameters affecting the dynamic of radon progeny, as well as its actual health effects [23,24]. In fact, the smaller the carrying particles the higher the probability that the progeny attached fraction will reach the more defenseless regions of the lungs [25]. To this end, ultrafine particles (UFPs, particles smaller than 100 nm), could represent a more efficient carrier of radon progeny into the alveolar and tracheobronchial regions.

In addition to their possible role as a carrier of radon decay products, airborne particles have also been classified as carcinogenic to humans (Group 1) [26–28] and responsible of cardiovascular deaths [29,30]. The scientific community identified a number of indoor sources emitting particles in the sub-micrometric and ultrafine ranges. The highest emitting sources were shown to be cooking activities [31–35], candle and incense burning [36], smoking/vaping activities [37–40], and residential heating (fireplaces). In particular, indoor exposure was found to be the highest contributor to the overall daily particle dose received by people [10,11,41].

In summary, indoor microenvironments can be characterized by high concentration levels of both radon and airborne particles. The effects of these two pollutants are usually studied separately. However, a synergic effect may also be present, which would enhance the health effects of particle-radon co-exposure, with respect to the separate exposures. In order to understand possible synergies, the dynamics of radon progeny in the presence of indoor generated particles should be investigated in-depth. The present paper aims to assess the effect of different indoor sources on radon progeny evolution, in order to identify the parameters which are likely to influence the radon progeny dynamics. To this purpose, experiments were performed in a controlled chamber at different radon and particle concentration levels, as well as different particle distributions, in order to monitor the variations of attached/unattached fraction dynamics.

2. Material and methods

2.1. Site description

This study was performed under the framework of a scientific co-operation agreement between the Italian National Institute of Ionizing Radiation Metrology (INMRI) and the Department of Civil and Mechanical Engineering of University of Cassino and Southern Lazio (Italy). The experimental campaign was carried out in a controlled 150-m^3 radon chamber ($7 \times 6.2 \times 3.5$ m) located at the ENEA Research Center in Casaccia (Rome, Italy). The chamber walls were made up of reinforced concrete with a thickness of 0.3 m. Radon gas emanated from the underneath soil entered the chamber through two vertical pipes. The chamber was under negative pressure since it has a lower pressure than adjacent areas: this improve the radon emanation from the underneath soil. Moreover, the radon concentration inside the chamber was also controlled by i) valves which regulated the flow of radon emissions into the chamber ii) a further ad-hoc mechanical ventilation system allowing to provide an increase of the air exchange rate of the room when lower radon concentration are needed. In particular, a further (minimum) exchange flow rate of 1 m^3 h^{-1} was set up for the experimental campaign since high radon concentrations were considered.

2.2. Experimental apparatus and quality assurance

Radon activity concentration was monitored with an AlphaGUARD PQ 2000 PRO Professional Radon Monitor (Genitron, Germany) which was calibrated using the INMRI Radon reference measurement system before the experimental study. The instrument, based on a 0.6 L ionization chamber, operated with 60 min integration time in diffusion mode, recording also temperature, pressure and relative humidity.

Radon Decay Products (RDPs) and concentrations in the air were monitored by a Radon Daughter Monitor (BWLM_PLUS_2S, Tracelab, Germany). The instrument has two independent acquisition units: one for total airborne RDP (attached and unattached fraction), and another one for the unattached fraction solely. In each unit air is driven through a sampling filter by a pump with a flux of 100 L h^{-1}. The filter of the first unit is made by cellulose, while a wire mesh is equipped on the second unit. An alpha spectrometer faces each unit. The spectrometers were made up of a surface barrier detector and an integrated multi-channel analyzer that collected the alpha-spectrum in 256 channels. The resolution is 22.6 channels per MeV, whereas the peak-width at half of the maximum is about 8 channels, which allows for separation of the ^{212}Po, ^{214}Po, and ^{218}Po peaks. The overall efficiency of the alpha spectrometer was 18%, including geometrical factor and detector efficiencies. The device was calibrated by Bundesamt für Strahlenschutz (BfS, Germany) within one year before the study.

Particle number distributions and total concentrations were measured through a Scanning Mobility Particle Sizer Spectrometer (SMPS 3996, TSI Inc.) and a Condensation Particle Counter (CPC 3775, TSI Inc), respectively. The CPC 3775 is a butanol-based condensation particle counter able to measure number concentrations up to 1×10^7 part. cm^{-3} with 1-s sampling time, for particles >4 nm in diameter. The SMPS is made up of an Electrostatic Classifier (EC 3080, TSI Inc.), which classifies particles according their electrical mobility diameter, and a further CPC 3775 was used to count the classified particles, thus providing particle number size distribution in the 6–220 nm or 14–700 nm mobility diameter range. Particle number distributions were measured based on a 135-s sampling