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Radon-222 related influence on ambient gamma dose

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ABSTRACT

Ambient gamma dose, radon, and rainfall have been monitored in southern Bucharest, Romania, from 2010 to 2016. The seasonal cycle of background ambient gamma dose peaked between July and October (100–105 nSv h^{-1}), with minimum values in February (75–80 nSv h^{-1}), the time of maximum snow cover. Based on 10 m a.g.l. radon concentrations, the ambient gamma dose increased by around 1 nSv h^{-1} for every 5 Bq m^{-3} increase in radon. Radon variability attributable to diurnal changes in atmospheric mixing contributed less than 15 nSv h^{-1} to the overall variability in ambient gamma dose, a factor of 4 more than synoptic timescale changes in air mass fetch. By contrast, precipitation-related enhancements of the ambient gamma dose were 15–80 nSv h^{-1}. To facilitate routine analysis, and account in part for occasional equipment failure, an automated method for identifying precipitation spikes in the ambient gamma dose was developed. Lastly, a simple model for predicting rainfall-related enhancement of the ambient gamma dose is tested against rainfall observations from events of contrasting duration and intensity. Results are also compared with those from previously published models of simple and complex formulation. Generally, the model performed very well. When simulations underestimated observations the absolute difference was typically less than the natural variability in ambient gamma dose arising from atmospheric mixing influences. Consequently, combined use of the automated event detection method and the simple model of this study could enable the ambient gamma dose "attention limit" (which indicates a potential radiological emergency) to be reduced from 200 to 400% above background to 25–50%.

1. Introduction

Radiological emergencies at nuclear facilities (e.g. Chernobyl in 1986; Fukushima in 2011) as well as unregulated or regulated weapons testing or detonations, have the potential to spread hazardous radioactive fallout over extensive regions of the globe (Lipscy et al., 2013; Steinhauser et al., 2014; Pravalie, 2014). Mitigation of the potential health effects from such incidents relies heavily upon early identification of the threat, and a rapid response (Steinhauser et al., 2014; UNSCEAR, 2014). Since gamma-emitting radionuclides are common to most forms of nuclear-related fallout, near-surface monitoring of the "ambient equivalent gamma radiation dose rate" has become widely adopted as a means of identifying such events. The ambient equivalent gamma radiation dose rate (hereafter "ambient gamma dose") is a measurable equivalent of the effective gamma radiation dose, which quantifies the human health risk associated with gamma radiation exposure (ICRU, 1993).

Continuous monitoring of the near-surface ambient gamma dose is performed routinely at major nuclear facilities, but also more generally on the country level at various locations around Europe. This monitoring network constitutes the main component of a country-level early warning system for radiological emergencies. The sensitivity and efficacy of this country-level warning system depends upon (i) the quality of observations (instrument performance and consistency of quality control/analysis procedures), and (ii) the level of understanding of the variability in the near-surface ambient gamma dose due to other contributing factors.

Natural factors contributing to the spatial and temporal variability in the near-surface ambient gamma dose include: cosmic radiation, local soil and rock characteristics (natural radionuclides in the soil), soil water content, atmospheric concentrations of short-lived naturally-occurring gamma emitting aerosols (e.g. progeny of ^{222}Rn and ^{220}Rn) that can change with atmospheric stability, and precipitation (snow or rain) that can concentrate gamma emitting aerosols at the surface (Bossew et al., 2017; Barbosa et al., 2017; Levin and Cotton, 2008).

In the absence of nuclear events, each monitoring station of the network provides useful information about background levels of the ambient gamma dose, which is site specific. Observations from each

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station can then be analysed to characterise the local terrestrial component of the ambient gamma dose and its natural variability (e.g. as a predictor of geogenic radon potential) (Bossew et al., 2017), information that is subsequently used to derive indicator thresholds for nuclear incidents. However, before reliable thresholds or indicators can be developed for radiological emergencies it is necessary to understand the natural short-term variability in ambient gamma dose resulting from meteorological factors that might otherwise be misinterpreted as an incident/event.

The majority of data reported from all local or national European ambient gamma dose monitoring stations are non-validated, implying that anomalies associated with changing meteorological conditions (including periods of atmospheric stability or heavy precipitation), and malfunctions of the equipment hardware or software, have not been identified and removed, so they can appear as erroneous high values. Consequently, isolated activity-threshold-alarms in these datasets cannot automatically be treated as a true indication of a dangerous increase in levels of radioactivity. Since significant ambient gamma dose variability can result from natural causes, this limits the current ability to generate prompt alerts of potential malfunctions of nuclear facilities or other external nuclear accident. In Europe, thousands national stations continuously monitor the ambient gamma dose contributing to EURDEP system, coordinated by the Joint Research Centre (JRC) of the European Community (see https://eurdep.jrc.ec.europa.eu/Basic/Pages/Public/Home/Default.aspx). There is a high degree of heterogeneity between those national networks regarding their architecture and design, instrumentation and evaluation, but their harmonization was recently possible (Bossew et al., 2017).

Currently, due to the combination of natural influences and site-to-site variations in equipment quality and data reduction methods, the "attention limit" for the European network observations is set at 2–4 times the long term average background value, and the automatic "alert limit" is set at around 10 times the average background. In the hope of optimising these limits to improve the sensitivity and reliability of the European-wide detection network there has recently been growing interest to increase the quality of ambient gamma dose data (Bossew et al., 2017), a goal that is contingent upon an improved characterisation of natural influences.

At the international level it has long been established that the most significant natural process contributing to short term variability of the near-surface ambient gamma dose is the precipitation-induced deposition of radon progeny (e.g. ^{214}Pb, ^{214}Bi) (Mercier et al., 2009; Voltaggio, 2012). A secondary natural influence is the nocturnal enhancement of ambient gamma dose due to radon build-up within the stable boundary layer. Lastly, considerable spatial variability in the ambient gamma dose can also arise due to radionuclides deposited as a result of past nuclear accidents/events.

Peaks associated with precipitation events are typically characterised by large, short-term increases of the ambient gamma dose followed by a rapid return to background levels, or temporarily below. Temporary below-background levels of ambient gamma dose following rainfall events sometimes result due to water-logging of the surface soil, which acts as a partial gamma shield for natural radioactivity in the soil as well as preventing radon emanation (Smetsers and van Lunenburg, 1994; Smetsers and Blaauboer, 1994; Smetsers, 1995; Blaauboer and Smetsers, 1997). In order to estimate the potential increase of ambient gamma dose to humans, the associated rainfall events need to be reliably identified, extracted, and have the estimated terrestrial background signal removed.

It is difficult to reliably predict the increase in ambient gamma dose that results from precipitation events. The concentration of radon progeny in precipitation is a complex function of atmospheric radon concentration (and distribution throughout the lower atmosphere) (Williams et al., 2011; Chambers et al., 2013), the history of cloud formation and the type of precipitation (rain or snow) (Greenfield et al., 2003). The corresponding increases in near-surface ambient gamma dose do not seem to be consistently correlated with the rain rate, rain event duration, precipitation volume or other meteorological parameters (Cortes et al., 2001; Fujinami, 1996; Greenfield et al., 2003). However, an inverse relationship between progeny concentration and the precipitation rate has often been reported (Fujinami, 1996; Paatero, 2000; Greenfield et al., 2003). In-cloud scavenging processes, where the radon progeny directly or indirectly attach themselves to other aerosols and form condensation nuclei for rain drops, have been shown to be much more efficient method of incorporating radon progeny in rainfall than below-cloud scavenging, where radon progeny are incorporated by, or attach themselves to, falling rain drops (Mercier et al., 2009). Consequently, the concentration of radon progeny in precipitation is believed to be more closely related to the in-cloud radon concentration (rainout) than the radon concentration in the air mass below the cloud (washout).

The aim of this contribution is to summarise our progress to date regarding the characterisation of contributions to the ambient gamma dose from radon in the lower troposphere. Most significantly, the performance of a simple model developed at the "Horia Hulubei" National Institute for Physics and Nuclear Engineering for estimating rainfall-related changes to the near-surface ambient gamma dose, is compared to that of a selection of simple and complex models developed for the same purpose.

2. Materials and methods

2.1. Site and measurements

The "Horia Hulubei" National Institute for Physics and Nuclear Engineering (IFIN-HH) is situated in a suburban area of southern Romania (44°21′2.72″N, 26°02′38.42″E), about 10 km southwest of the Bucharest central business district. The atmospheric monitoring system at IFIN-HH utilises a 60 m meteorological tower. Continuous measurements of wind speed, wind direction, temperature and relative humidity are made at 30 and 60 m above ground level (a.g.l.). Net radiation, solar radiation and rainfall are monitored at 30 m a.g.l., atmospheric radon concentration at 10 m a.g.l. (or 2 m from 2015), and the ambient gamma dose is monitored at 1.5 m a.g.l. (or 2 m from 2015). In the immediate vicinity of the site, within the measurement fetch, there is a heterogeneous mix of trees and buildings that vary in height from 10 to 15 m a.g.l.

All observations from the monitoring system are logged as 10-min averages of 10-s readings and subsequently integrated to hourly averages for analysis. In addition to the meteorological observations already described by Galeriu et al. (2011, 2014), atmospheric radon concentration is monitored using an "AlphaGUARD" (PQ2000 PRO, Saphymo, Germany; which also separately monitors atmospheric pressure, temperature and relative humidity). As described in Chambers et al. (2016), the AlphaGUARD was situated in a Stevenson's Screen (well-ventilated, weatherproof enclosure) to protect it from precipitation, operated in diffusion mode, and set for hourly integration. The ambient gamma dose is measured using a GAMMATRACER (XL2-2-RS232).

2.2. Rainfall and ambient gamma dose

When the short-lived ^{222}Rn progeny ^{214}Pb and ^{214}Bi are deposited on the ground by rain, they significantly increase the near-surface (above ground) ambient gamma dose. Within a short period of time the rainwater containing ^{214}Pb and ^{214}Bi can migrate into the soil and form a volume source. For a short, low-intensity, rainfall event the contaminated soil layer is thin, and the depth of penetration depends on the initial soil water content. For a prolonged rainfall event, contaminated water from the start of the event is pushed deeper into the soil, resulting in a radioactivity profile with a quasi-exponential shape. If the total radioactivity per unit soil surface area is considered, the dose