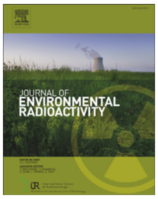
Journal of Environmental Radioactivity 154 (2016) 68–82

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

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



Atmospheric stability effects on potential radiological releases at a nuclear research facility in Romania: Characterising the atmospheric mixing state

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ARTICLE INFO

Article history:
Received 10 November 2015
Received in revised form
13 January 2016
Accepted 17 January 2016
Available online 6 February 2016

Keywords:
^{222}Rn
Atmospheric stability
Mixing depth
Radon flux
Radioactive releases
Tritium

ABSTRACT

A radon-based nocturnal stability classification scheme is developed for a flat inland site near Bucharest, Romania, characterised by significant local surface roughness heterogeneity, and compared with traditional meteorologically-based techniques. Eight months of hourly meteorological and atmospheric radon observations from a 60 m tower at the IFIN-HH nuclear research facility are analysed. Heterogeneous surface roughness conditions in the 1 km radius exclusion zone around the site hinder accurate characterisation of nocturnal atmospheric mixing conditions using conventional meteorological techniques, so a radon-based scheme is trialled. When the nocturnal boundary layer is very stable, the Pasquill–Gifford "radiation" scheme overestimates the atmosphere's capacity to dilute pollutants with near-surface sources (such as tritiated water vapour) by 20% compared to the radon-based scheme. Under these conditions, near-surface wind speeds drop well below 1 m s^{-1} and nocturnal mixing depths vary from ~25 m to less than 10 m above ground level (a.g.l.). Combining nocturnal radon with daytime ceilometer data, we were able to reconstruct the full diurnal cycle of mixing depths. Average daytime mixing depths at this flat inland site range from 1200 to 1800 m a.g.l. in summer, and 500–900 m a.g.l. in winter. Using tower observations to constrain the nocturnal radon-derived effective mixing depth, we were able to estimate the seasonal range in the Bucharest regional radon flux as: 12 mBq m^{-2} s^{-1} in winter to 14 mBq m^{-2} s^{-1} in summer.

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1. Introduction

Nuclear facilities are commonly required to monitor their emissions of radioactive gases and aerosols to the environment, in order to gauge the integrated environmental impacts of routine releases of pollutants, and to help in the forecasting of potential health risks associated with accidental releases (Galeriu et al., 2014; IAEA, 2011a,b; EURATOM TREATY, http://www.euratom.org/). An important component of regulatory monitoring programs is the routine measurement of meteorological quantities that can be used to characterise the state of the atmosphere and its ability to dilute the emitted pollutants and transport them away from the area.

Near-surface concentrations of hazardous air-borne pollutants to which workers, residents, wildlife or crops may be exposed, are primarily a function of the source strength, the volume of the atmosphere into which they mix, and entrainment processes (e.g. Pal, 2014). For pollutants with sources typically below the height of the nocturnal inversion layer, concentrations will usually peak in calm pre-dawn conditions, when the lower atmosphere is most poorly mixed (e.g. Avino et al., 2003; Baciu, 2005; Galmarini, 2006; Pearce et al., 2011; Crawford et al., 2016; Grundström, 2015; Chambers et al., 2015a,b). On the other hand, emissions from elevated sources (e.g. tall stacks) frequently result in peak concentrations shortly after dawn (so-called "fumigation events"; Oke, 1987), when turbulence erodes the nocturnal inversion and incorporates the overlying air into the developing convective boundary layer.

Since establishing and maintaining dense regional monitoring networks is logistically and economically prohibitive, efforts to understand the impact of radioactive releases into the atmosphere

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http://dx.doi.org/10.1016/j.jenvrad.2016.01.010
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tend to rely mainly upon plume dispersion modelling and our ability to accurately assess the "stability" (degree of mixing) of the lower atmosphere. The most accurate conventional techniques to characterise atmospheric stability (e.g., Richardson number, Obukhov length; Foken, 2008) are based on surface and boundary layer similarity theory. However, since these require sophisticated (research quality) instruments that are expensive and hard to maintain routinely, they are not widely adopted. Consequently, less accurate categorical techniques derived from routine meteorological measurements, such as the Pasquil-Gifford turbulence and radiation schemes (Pasquil, 1961; Turner, 1964; Pasquill and Smith, 1983), tend to be more commonly adopted.

A new approach to stability classification that is gaining acceptance relies upon observations of the naturally-occurring radioactive trace gas Radon-222 (e.g. Perrino et al., 2001; Perrino, 2012; Chambers et al., 2015a,b, and references therein). Because radon is emitted steadily by all soils and rocks, it has a surface source function that varies slowly in both space and time. In particular, diurnal radon source variations can be considered to be constant at most inland sites. Once in the atmosphere, radon is unreactive, poorly soluble and decays at a rate that is small in comparison to vertical mixing time scales. As a result, near-surface radon concentrations represent a direct measure of the intensity and vertical extent of nocturnal atmospheric mixing, and perform better than meteorological proxies in characterising the outcomes of vertical mixing on surface-emitted passive scalar quantities (IAEA, 2012; Williams et al., 2013; Chambers et al., 2015b).

Horia Hulubei National Institute for Research and Development in Physics and Nuclear Engineering (IFIN-HH) is the largest research institute in Romania. Close to Bucharest, the site is located in a topographically-complex mixed urban-rural landscape. In order to better assess the fate of the radioactive gases and aerosols that are exhausted from this facility via a 40 m stack, a comprehensive suite of meteorological measurements have been made since 1995 (when the reactor and radiopharmaceutical production facilities were operational) from a nearby 60 m tower (Galeriu et al., 2014). The effective release height at IFIN-HH site is similar to that at the nearby Cernavoda Nuclear Power Plant (CNPP), which currently produces around 20% of Romania's electricity. CNPP is also situated in relatively complex terrain, and is the subject of ongoing research on the influence of atmospheric stability on pollutant concentrations using conventional meteorological techniques.

Since 1997, when reactor operations at IFIN-HH ceased, the meteorological tower has served as a real-time data provider for the RODOS (Real-time On-line DecisiOn Support) decision support system for nuclear emergencies (Galeriu et al., 2011; Ehrhardt, 1997). In particular, FDMH (Food and Dose Module Hydrogen), a component of RODOS, was used to investigate the radiological impact of accidental releases of tritiated water and transfer from the atmosphere to the food chain or inhalation pathways (IAEA, 2014; Galeriu et al., 2000). Since the start of the reactor decommissioning process in 2007 it has become necessary to increase the levels of mandatory meteorological surveying, and tower observations also began to be made available for use in land–atmosphere interaction studies.

At night, when atmospheric conditions are stable, near-surface concentrations of tritiated water (HTO) increase, thereby increasing the radiological hazard. Under these conditions HTO more readily makes its way into soil water or enters the crops in the surrounding rural areas and is converted to organic forms. If tritium from HTO binds to carbon within the crops, organically bound tritium (OBT) is formed. Since OBT is a very stable organic form the radiological hazard (through risk of subsequent ingestion and incorporation into DNA) is increased. At present crop uptake of HTO at night and subsequent formation of OBT is not well understood (Melintescu et al., 2015; Galeriu and Melintescu, 2011, 2015). Of the few models used to estimate the OBT:HTO ratio that consider nocturnal uptake, parameterisations of the process are very simplistic (Melintescu and Galeriu, 2015; IAEA, 2014; Melintescu et al., 2015; Galeriu and Melintescu, 2015).

Nocturnal atmospheric stability classification by conventional meteorological approaches has proven to be problematic at sites with significant surface roughness heterogeneity such as IFIN-HH (Galeriu et al., 2014). In light of this issue, the current study aims to: (i) develop a versatile, continuous (i.e., not categorical) classification scheme for the atmospheric mixing state at the IFIN-HH site based on near-surface atmospheric radon measurements, (ii) characterise the behaviour of key meteorological quantities within the defined mixing states, (iii) compare the efficacy and consistency of the Pasquil-Gifford and radon-based approaches for nocturnal atmospheric stability classification by assessing their ability to distinguish changes in key parameters (including near-surface wind speed and gradients, temperature gradients, and net radiation) between identified stability categories, and (iv) characterise changes in atmospheric mixing depth across the diurnal cycle for a range of atmospheric stabilities in summer and winter. Detailed investigation of identified stability influences on nocturnal tritium concentrations, as well as improvement of parameterisations of tritium uptake by crops, will be the subject of a subsequent investigation.

2. Materials and methods

2.1. Site and measurements

IFIN-HH is situated approximately 10 km SSW of the Bucharest central business district (44°21′2.72″N, 26°02′38.42″E) in a mixed urban-rural landscape (Fig. 1). Locally, the surface is quite inhomogeneous and, in places, roughness elements (including trees and buildings) reach 10–15 m above ground level, a.g.l. When the reactor and radiopharmaceutical production at this site were in operation, radioactive gases and aerosols were exhausted from this facility via a 40 m stack, with the effective source height ranging from 44 to 55 m. However, since operations at this facility ceased, radioactive emissions from this site have been minor and often hard to distinguish from background.

A 60 m instrumented tower is located on the premises of this research facility. Wind speed, wind direction, temperature and relative humidity are monitored at 30 and 60 m a.g.l., solar radiation is monitored at 10 m, net radiation and rainfall at 30 m agl. All observations are logged as 10-min averages of 10-s readings and subsequently integrated to hourly averages for analysis. In addition to the meteorological observations, atmospheric radon concentration is monitored at 10 m a.g.l., using an "AlphaGUARD" (PQ2000 PRO, Saphymo, Germany; which also monitors atmospheric pressure, temperature and relative humidity). 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.

Information regarding cloud cover, cloud height, atmospheric mixing depth and aerosol properties is recorded nearby using a vertically-oriented modified CHM 15k Nimbus ceilometer (Nimbus Jenoptik, Germany). The ceilometer's measurement range is 5 m–15 km, with a 15 m resolution and full overlap range of 300–400 m. Its principle of measurement is similar to that of a lidar, using light detection and ranging to target aerosols and cloud. For the purpose of this investigation the ceilometer's 30 s output was averaged to hourly resolution, and mixing depth was taken to be the height of the first identified inversion (PBL_0). Further details regarding the tower instrumentation are provided in Galeriu

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