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RADON-BASED ASSESSMENT OF STABILITY EFFECTS ON POTENTIAL RADIOLOGICAL RELEASES

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Abstract: Eight months of climatology and ^{222}Rn observations from a 60 m tower at a nuclear research facility in Romania are analysed. Heterogeneous surface roughness conditions in the site's 1 km radius exclusion zone hinder accurate characterisation of atmospheric stability using 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 below 1 ms^{-1} and nocturnal mixing depths vary from ~25 m to less than 10 m above ground level (agl.). Daytime mixing depths at this flat inland site range from 1100 to 1800 m agl. in summer, and 500 to 900 m 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.

Key words: *^{222}Rn; nocturnal stability; stable nocturnal boundary layer; urban pollution; mixing height.*

Near-surface concentrations of air-borne pollutants are primarily a function of source strength and mixing depth. Nuclear facilities are required to monitor their emissions and local meteorology to gauge the environmental impacts of routine or accidental releases (Galeriu et al., 2014). Here, plume dispersion modelling is usually employed in lieu of a dense monitoring network to assess regional impacts, with the accuracy of forecasts closely related to how well meteorological observations can be used to characterise the atmospheric mixing state ("stability"). Historically, categorical techniques such as the Pasquil-Gifford schemes (e.g. Pasquill and Smith, 1983) have been used for stability classification rather than techniques based on similarity theory. Here we demonstrate a recently-developed radon-based technique for stability classification (Chambers et al., 2015a,b) that can be easily implemented in models for regulatory reporting. Radon is an ideal atmospheric tracer; it is unreactive, poorly soluble, has a relatively consistent terrestrial source, and is approximately conservative over the course of a single night ($t_{0.5}$ ~ 3.8 days). Consequently, 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 (Williams et al. 2013).

Horia Hulubei National Institute for Research and Development in Physics and Nuclear Engineering is situated 10 km SSW of Bucharest, Romania, in a mixed urban-rural landscape. Since operations at this facility ceased in 1997 radioactive emissions from its 40 m stack have been minor. Hourly meteorological monitoring is routinely conducted from a nearby tower at 30 and 60 m agl. to assess the fate of emitted radioactive gases and aerosols. Observations from this tower have 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). In addition to the tower observations, 10m radon concentrations were recorded hourly (AlphaGUARD, PQ200 PRO), and daytime mixing depths were provided by a CHM 15k Nimbus ceilometer.

In the vicinity of the tower surface roughness elements (including trees and buildings) reach 10-15 m agl., which has made nocturnal stability classification by conventional meteorological approaches problematic. Here we use 8 months of hourly radon and meteorological observations in 2012 to develop a versatile classification scheme for the atmospheric mixing state, characterise the behaviour of key meteorological quantities within the defined mixing states, compare the efficacy and consistency of the Pasquil-Gifford and radon-based approaches for stability classification, and characterise seasonal changes in mixing depth across the diurnal cycle for a range of atmospheric stabilities.

Unlike the Pasquil-Gifford or bulk Richardson number techniques, which can provide stability classifications on an hourly basis, the radon-based approach assigns stability classifications to whole nights, based on observations within a 10-hour nocturnal window. Stability classification of the following daytime period is then inferred, with a high success rate, based on atmospheric "persistence".

A radon time-series (Figure 1a) represents variability on both long (> diurnal) and short (≤ diurnal) timescales. While long timescale variability is mostly related to air mass fetch effects, diurnal variability is mostly related to changes in the atmospheric mixing state (or "atmospheric stability").

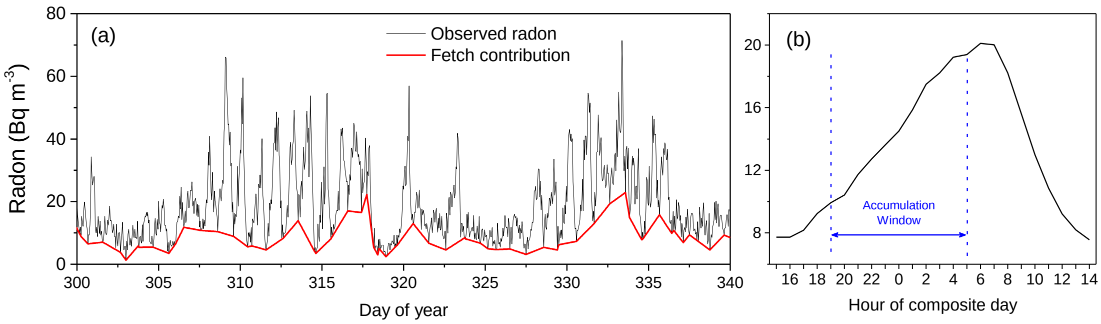

Figure 1. (a) Hourly radon time series with fetch-related component shown in red, and (b) composite diurnal cycle of radon at 10m agl., with 10-hour nocturnal accumulation window indicated.

Once the diurnal component has been isolated (i.e. by subtracting the fetch contribution from the observed concentrations), the mean nocturnal radon accumulation from 1900h – 0500h (Figure 1b) can be calculated each night and related to the mean atmospheric mixing state (e.g. Williams et al. 2016). After plotting the cumulative frequency histogram of nocturnal radon accumulation values (Figure 2a) thresholds (such as quartile ranges) can be assigned to define stability categories which are applied to 24-hour periods. Diurnal composites of radon for days assigned to one of the 4 stability categories (Figure 2b) show that "stable nights" result in the greatest diurnal amplitude of radon, whereas near-neutral (well mixed) nights result in the lowest amplitude chances. Since stable nights are usually associated with the clearest sky conditions and calmest winds, corresponding daytimes are usually the most unstable, and vice versa.

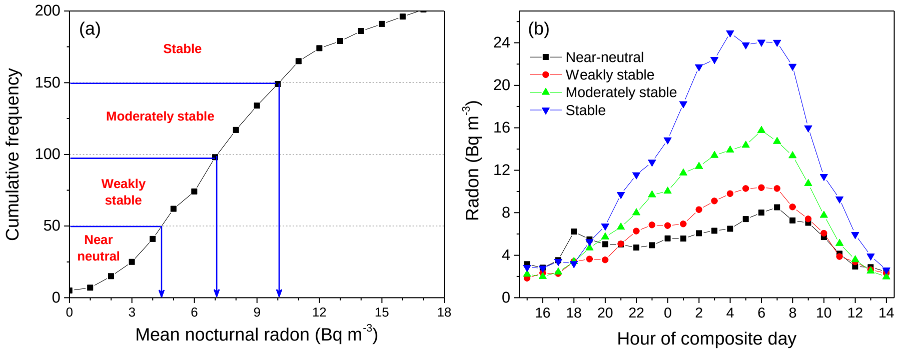

Figure 2. Cumulative frequency plot of nocturnal radon accumulation with assigned stability categories, and (b) diurnal composite radon concentration in each stability category.

As a "sanity check" of the radon-based scheme, we performed an hourly stability classification of the entire dataset using the Pasquil-Gifford radiation scheme (PGR), and bulk Richardson number (Ri_B) approach. We then formed diurnal composites (Figure 3) of the hourly PGR and Ri_B stability classifications according to the 4 daily radon-based stability categories. Most clearly seen in the seasonal Ri_B composites (Figure 3c,d), despite being defined for 24-hour periods based on only 10-hours of observations, the radon-based scheme reliably assigns hourly Ri_B estimates across the entire diurnal cycle (large positive values for stable nights, and increasingly large negative values for daytime instability); near neutral and weakly stable cases even fall either side of the "critical" Richardson number (Ri_C=0.25).

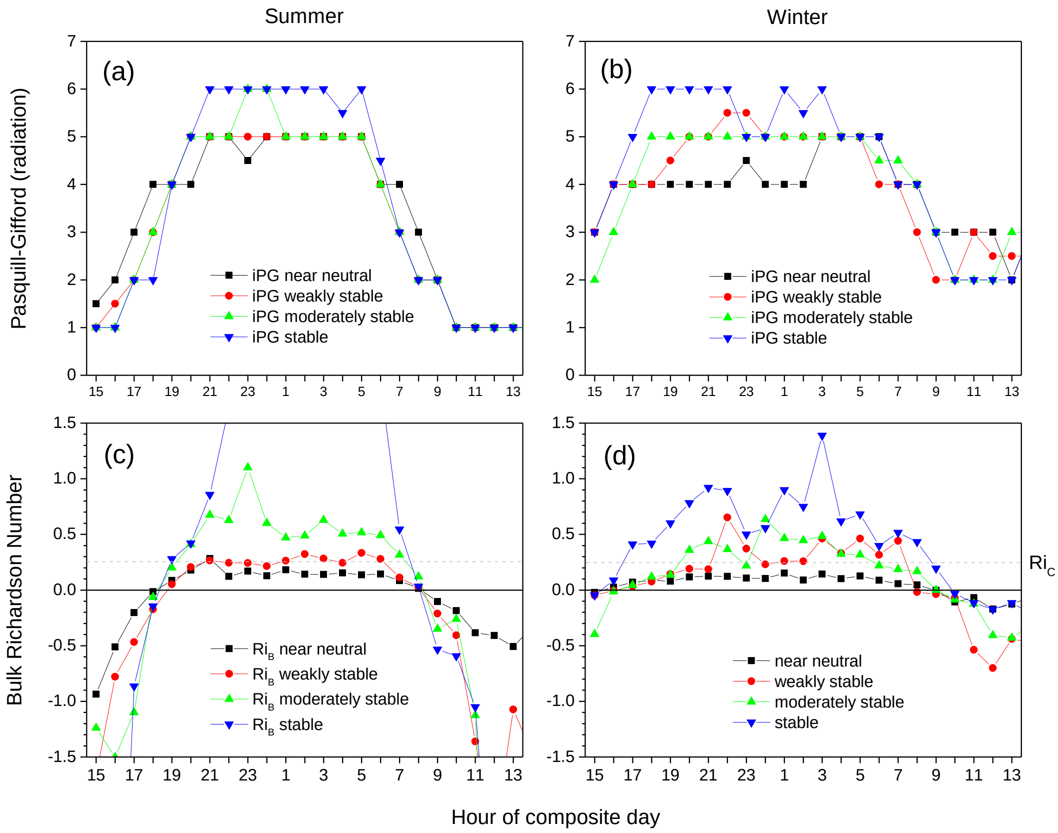

Figure 3. Composite PGR and Ri_B stability classifications for each radon-based stability category (PGR: 1 – unstable; 4 – neutral; 6 – stable).

Looking at hourly distributions of diurnal composite plots of evenings classified as stable by the radon-based and PGR schemes (Figure 4), the radon-based scheme yields median diurnal amplitudes ~20% greater than that of the PGR scheme. Since the behaviour of radon is representative of the behaviour of tritiated water vapour (or other gaseous pollutants with sources beneath the nocturnal inversion layer), it is evident that the choice of stability classification technique can have serious implications for exposure estimates in the event of a release from the facility.

When climatological observations are grouped by radon-based stability categories clear and consistent differences are apparent (Figure 5). Near-neutral conditions were associated with stronger winds and more cloud cover and small near-surface temperature gradients, whereas "stable" nocturnal conditions were associated with near-calm conditions, clear skies and strong near-surface temperature gradients.

Average daytime mixing depths reported by the ceilometer ranged from 600m in winter to 1200m in summer. Nocturnal mixing depths estimated from the 10m radon observations and a box model (Griffiths et al., 2013) varied from <10m on stable nights to ~60m on weakly stable nights. By constraining radon box model estimates of nocturnal effective mixing depth with tower observations we were able to well-constrain the seasonal variability in the Bucharest regional radon flux: 12 mBq m^{-2} s^{-1} in winter to 14 mBq m^{-2} s^{-1} in summer; values which were in close agreement with recently published European radon flux maps (e.g. Karstens et al., 2015).

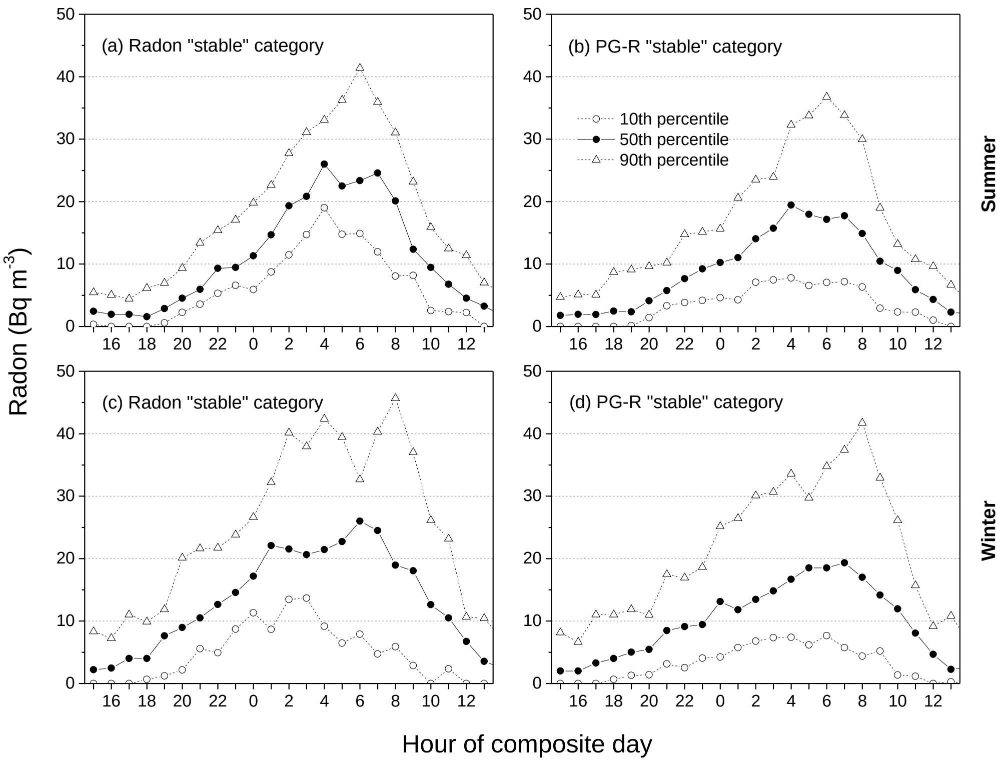

Figure 4. Comparison of extreme pollution events (stable nocturnal conditions) in summer and winter as predicted by the (a,c) radon-based scheme, and (b,d) PGR scheme.

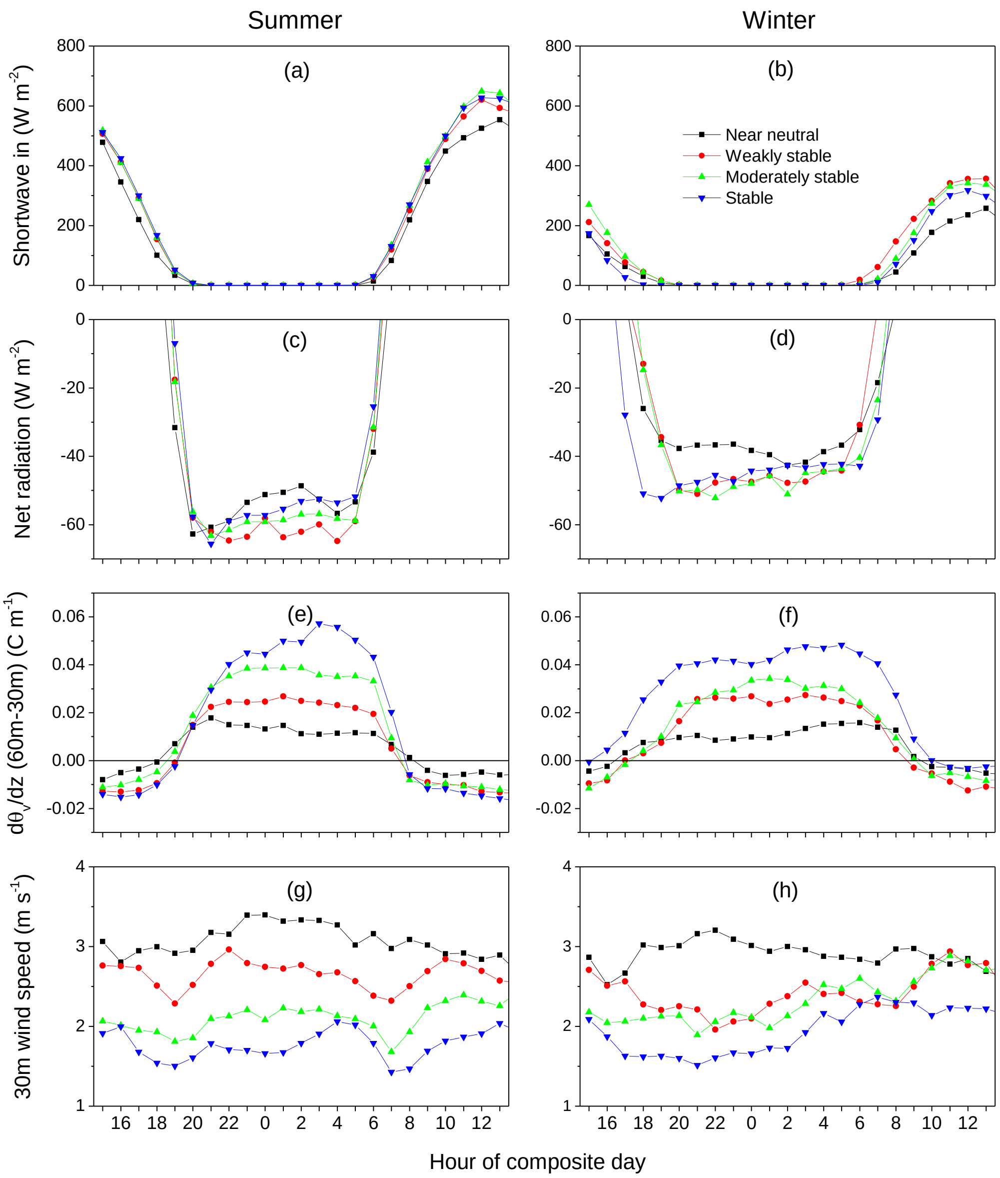

Figure 5. Diurnal composites of climatological parameters by season and radon-based stability category.

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