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Radon and radioactivity at a town overlying Uranium ores in northern Greece

K. Kourtidis [a, *], A.K. Georgoulias [a], M. Vlahopoulou [a], N. Tsirliganis [b], N. Kastelis [a], K. Ouzounis [a], N. Kazakis [b]

[a] Department of Environmental Engineering, School of Engineering, Democritus University of Thrace, 12 Vas. Sofias Str., 67100 Xanthi, Greece
[b] ATHENA Research and Innovation Center in Information, Communication and Knowledge Technologies, ISLP Xanthi Branch, 67100 Xanthi, Greece

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ABSTRACT

Extensive measurements of ^{222}Rn in the town of Xanthi in N Greece show that the part of the town overlying granite deposits and the outcrop of a uranium ore has exceptionally high indoor radon levels, with monthly means up to 1500 Bq m^{-3}. A large number of houses (40%) in this part of the town exhibit radon levels above 200 Bq m^{-3} while 11% of the houses had radon levels above 400 Bq m^{-3}. Substantial interannual variability as well as the highest in Europe winter/summer ratios (up to 12) were observed in this part of the town, which consist of traditional stone masonry buildings of the late 19th-early 20th century. Measurements of ^{238}U and ^{232}Th content of building materials from these houses as well as radionuclide measurements in different floors show that the high levels of indoor radon measured in these buildings are not due to high radon emanation rates from the building materials themselves but rather due to high radon flux from the soil because of the underlying geology, high radon penetration rates into the buildings from underground due to the lack of solid concrete foundations in these buildings, or a combination thereof. From the meteorological variables studied, highest correlation with indoor ^{222}Rn was found with temperature ($r^2 = 0.65$). An indoor radon prognostic regression model using temperature, pressure and precipitation as input was developed, that reproduced indoor radon with $r^2 = 0.69$. Hence, meteorology is the main driving factor of indoor radon, with temperature being the most important determinant. Preliminary flux measurements indicate that the soil-atmosphere ^{222}Rn flux should be in the range 150–250 Bq m^{-2} h^{-1}, which is in the upper 10% of flux values for Europe.

1. Introduction

Radon (^{222}Rn) is a nuclide originating from the uranium (^{238}U) decay chain. ^{238}U is occurring naturally in granitic, metamorphic, and non-metamorphic sedimentary rocks. Hence the underlying geology together with soil type influence radon flux from the surface to the atmosphere (Ball et al., 1991; Gillmore et al., 2005; Nazaroff, 1992; Tanner, 1980). Regarding indoor radon levels, building foundations and building type also influence indoor radon concentrations (e.g. Kropat et al., 2014). Indoor radon is also influenced by a variety of other, meteorology-related factors (e.g. Rowe et al., 2002).

Measurements of radon in houses in Greece are, as related to other countries in Europe, rather sparse. While measurements have been performed in spas (Danali et al., 1986), caves (Papachristodoulou et al., 2004), active faults (Papastefanou et al., 2001) and mines (Georgiou et al., 1988), literature on indoor levels in dwellings is not very extensive (Clouvas et al., 2007, 2011; Geranios et al., 2001; Ioannides et al., 2000; Nikolopoulos et al., 2002). Measurements of Rn emanation have been performed in Greek cement constituents (Maraziotis, 1987) and building materials (Papastefanou et al., 1984; Siotis and Wrixon, 1984).

In NE Greece, where our measurements were done, radon exhalation is expected to be around 75–150 Bq m^{-2} h^{-1}, which is on the upper 10% of the values for Europe (López-Coto et al., 2013). Especially in and near Xanthi, the underlying geology, also with the existence of a Uranium ore (see Section 2.1 below for details), may

* Corresponding author.
E-mail addresses: kourtidi@env.duth.gr (K. Kourtidis), argeor@env.duth.gr (A.K. Georgoulias), tnestor@ipet.gr (N. Tsirliganis), nkastei@env.duth.gr (N. Kastelis), ouzounis@env.duth.gr (K. Ouzounis), nikkazak@ipet.gr (N. Kazakis).

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result in high ^{222}Rn emanation rates and hence indoor concentrations.

Clouvas et al. (2007) measured indoor radon levels in 19 out of the 52 administrative regions (Prefectures) of Greece. Among the 19 studied Prefectures, the Prefecture of Xanthi had the highest mean radon concentration (229 Bq m^{-3}) and the highest percentage (42%) of schools with ^{222}Rn concentrations above 200 Bq m^{-3}. In a follow-up study, Clouvas et al. (2009) measured indoor radon concentrations in 77 schools of the prefecture of Xanthi in winter. The arithmetic mean radon concentration was 231 Bq m^{-3} with a range between 45 and 958 Bq m^{-3}.

The facts outlined above, prompted us to study in more detail radon in the town of Xanthi. The results presented here come from an extensive dataset of indoor radon and ancillary measurements in Xanthi, consisting of:

i. Three long-term (6 months or more) indoor radon datasets in three houses with a temporal resolution of around 48 h, allowing for the seasonal study of indoor radon fluctuations. One of these datasets was obtained concurrently with T, P and precipitation measurements, allowing for the study of the influence of meteorology on indoor radon levels.
ii. One dataset of extensive radon surveying in different houses in the town, with short-term radon measurements in around 25 houses in two different seasons;
iii. Outdoor γ-radiation measurements in the old part of the town with a spatial resolution allowing the mapping of γ-radiation levels;
iv. Indoor measurements of radionulcides in one house in the old part of the town;
v. Radioactivity measurements in building materials of traditional stone masonry houses;
vi. ^{222}Rn indoor flux determinations in the basements of a traditional stone masonry house.

2. Experimental methods

2.1. The site

The measurements were performed in the town of Xanthi, NE Greece. The northern part of the town, which is classified as historic and hence preserved (named, hereafter, Old Town), with the majority of the houses being stone masonry buildings of the late 19th to the early 20th century, is built on the mountain slope to the north and lies at the foothills of the Rodopi mountain range, overlying tertiary granite deposits (Fig. 1, top). The rest of the town, which is also its modern part, with the majority of the houses being 4–6 storey modern concrete structures build after 1970, lies at the alluvial plains, formed by Quaternary alluvial deposits (Fig. 1, top). The region hosts large tertiary granite/granodiorite deposits, and very near the study region is where the most important Uranium ores of Greece have been localized by the Greek Institute of Geology and Mineral Exploration IGME (Pergamalis et al., 1998, 2010). These deposits have an outcrop that appears to be underlying the northern part of the town of Xanthi (Pergamalis et al., 1998, 2010; Tsokas et al., 1996). The area belongs to the Rhodope Mountain complex. It hosts late Cambrian metamorphic rocks, Meta-, or Middle- Hercynian age granite bodies included inside them and an Oligocene age volcanic cover of rhyolites, quartz diorites, aggregated tuffs and tuffites gradually changing in sediments of the underlying volcano sedimentary series (Pergamalis et al., 1998, 2010; Tsokas et al., 1996). Further, as mentioned in Section 1 above, it is located in the region where the highest indoor radon concentrations in Greece were measured (Clouvas et al., 2011).

Due to the building type (no concrete foundation etc), the underlying geology and the building materials (stones, to a large part granodioritic ones, from a nearby riverbed), the Old Town could be prone to high indoor radon levels.

2.2. The instrumentation

Radon was measured with an electronic radon dosimeter (Ramon 2.2, GT-Analytic SARL, France) with a silicon semiconducting sensor. The sensor detects alpha particles emitted from the decay of Polonium-218 (^{218}Po), the latter originating from ^{222}Rn decay. The Ramon 2.2 was factory-calibrated against a reference AlphaGuard Radon detector, traceable to the Austrian National Radon Standard and to the European Primary Radon Standard of the PTB (Physikalisch-Technische Bundesanstalt) in Braunschweig, Germany. As of 2011, the instrument had very successfully participated at six validation tests for radon detectors, two of which were conducted by the Austrian National Institute of Metrology (BEV) (2005 and 2009), three were conducted in Switzerland by the Paul Scherrer Institute (2005, 2006 and 2010) and one was conducted by the National Radiation Protection Institute of the Czech Republic (2010). Due to the successful participation at these validation tests, the Ramon 2.2 complies with the general requirements for the competence of calibration and testing laboratories according to DIN EN ISO/IEC 17025. The precision of the measurement is 5% and the monitor has a temporal resolution of 48 h.

Outdoor measurements of ionising radiation were made with a dose rate meter (AUTOMESS 6150AD4, Automess GmbH, Germany). The sensor was calibrated with a ^{137}Cs source and its precision is 5%. The dose rate meter measures photon radiation (gamma and X) and provides average values as well as maximum values over a period of measurements. A built-in Geiger–Mueller (GM) counting tube serves as the detector. The device can be also operated with external probes. An external probe (AUTOMESS 6150 AD-17, Automess GmbH, Germany) was used for measuring alpha, beta and gamma radiation.

Indoor measurements of radionuclides were made using a portable γ-spectrometer with a NaI crystal (SafeSPEC, Eurisys Mesures, France). The detected radionuclides were potassium-40 (^{40}K), thallium-208 (^{208}Tl) and bismuth-214 (^{214}Bi). For the identification of ^{40}K the 1461 keV gamma ray line from its decay to ^{40}Ar by electron capture was used., while for ^{208}Tl the 2615 keV gamma ray line (supported also by the 860 keV line) from its β-decay to ^{208}Pb, and for ^{214}Bi the 1764 keV gamma ray line from its β-decay to ^{214}Po were used.

The alpha counting system used for the assessment of building material samples was the ELSEC 7286 Low Level Alpha Counting System. Up to four scintillation-photomultiplier assemblies can be controlled from one control unit. There is an adjustable high voltage supply and discriminator unit for each photomultiplier tube (PM). The output pulses from the discriminator circuit are fed into an internal microprocessor which keeps track of the total number of fast pairs from the decay of ^{219}Rn followed by the decay of ^{215}Po (half-life 1.8 ms) in the ^{235}U series which occur within 4 ms and slow pairs from the decay of ^{220}Rn followed by the decay of ^{216}Po (half-life 0.15 s) in the ^{232}Th series, which occur between 20 and 400 ms apart. These pair counts allow the user to estimate the uranium and thorium contents according to the method described by Aitken (1985). Counting time was 1 h, and for each sample there were 42–77 repetitions. The samples were crushed, and in the system, a fine powdered layer of the sample is placed on top of a ZnS-coated Mylar film scintillation screen. The range of the most energetic alpha particles from thorium and uranium is around 50 μm. When these particles hit the ZnS-coated Mylar surface, it produces light and this light produces photoelectrons from the