REGIONAL CALIBRATION OF EROSION RADIOTRACERS (^{210}Pb AND ^{137}Cs): ATMOSPHERIC FLUXES TO SOILS (N SPAIN)

SUPPORTING INFORMATION

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Study areas

Carrion Basin

The Carrion River, with a mean annual discharge of 0.66 km^3, is a tributary of the Duero River (Northern Castilian Plateau) through the Pisuerga River. Its catchment drains an area of 3351 km^2 mainly in the province of Palencia. In the upper basin, the river flows through alkaline soils developed on a geological substrate of alternating limestone and shale. In the middle and lower basin, the river flows through soils developed from clay-rich Tertiary and Quaternary deposits. The upper basin is a mountainous region where coal mining activities have been taking place for over a century, although they have now drastically decreased. The climate in the riverhead region is continental Mediterranean and in the lower basin the continental characteristics are stronger, with long, cold winters and hot, dry summers.

Noguera Pallaresa Basin

The Noguera Pallaresa Basin in the province of Lleida (Figure 1) is part of the Ebro Basin (NE Spain), draining a total area of 2821 km^2. The Noguera Pallaresa River has an average annual discharge flow of about 1.5 km^3·yr^{-1}, displaying seasonal fluctuations. There are strong climate and landscape contrasts between the northern and southern basins. The upper basin is characterized by a Mediterranean climate under high mountain continental influence with annual rainfall ranging from 700 to 1500 mm yr^{-1}. The rest has a typical Mediterranean climate under low mountain continental influence with an annual rainfall ranging from 500 to 700 mm yr^{-1} in the central basin, and less than 500 mm yr^{-1} in the lower basin. The geological units present in the region are mainly metamorphic and granitic rocks in the Pyrenees mountains (Paleozoic period), and calcareous and dolomiteous rocks (Mesozoic period) in the pre-Pyrenees range and the lower basin.

Sample pre-treatment and analysis

In the laboratory samples were dried at 50ºC over a 3 day period in a mechanical convection incubator and then sieved using a 2 mm mesh. Bulk dry density was determined and homogeneous aliquots were sub-sampled for ^{210}Pb, ^{137}Cs and ^{226}Ra analysis.

The soil cores from the Carrion Basin were measured by γ-ray spectrometry at the Low-Level Radioactivity Laboratory of the University of Valladolid (LIBRA). All measurements

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were carried out with a Canberra n-type HPGe detector, with 25% relative efficiency, resolutions of 1.1 keV at 122 keV and of 2.0 keV at 1.33 MeV. Samples were placed in Petri dishes, sealed and stored for at least 3 weeks in order to ensure secular equilibrium of ^{226}Ra and ^{214}Pb. ^{210}Pb and ^{137}Cs activities were calculated from their photopeaks at 46.5 and 662.1 keV, respectively. To determine ^{226}Ra activity, the 351.9 keV photopeak from ^{214}Pb was used. The efficiency calibration curve for that geometry was obtained following the procedure described by Daza and co-workers (1), using several calibration sources prepared by labelling soil samples with a multi-gamma mixture (DAMRI 9ML01-ELMH05/4) and a ^{210}Pb solution provided by CIEMAT. Nevertheless, at 46.5 keV self-absorption effects are pronounced and differences in bulk density or chemical composition between the calibration samples and the sample under study could result in significant variations in the efficiency. Hence, the efficiency value at 46.5-keV was corrected to account for matrix effects by using a Monte Carlo-based method (2).

The soil cores from the Noguera Pallaresa Basin were analyzed by the Environmental Radioactivity Laboratory at the Autonomous University of Barcelona. Determination of ^{210}Pb activities was accomplished through the measurement of its daughter nuclide ^{210}Po in equilibrium (3). Briefly, aliquots of 100-200 mg were totally digested by using an analytical microwave oven after addition of ^{209}Po as an internal tracer. Plated onto high pure silver discs, polonium isotopes were counted by α-spectrometry using low background silicon surface barrier (SSB) detectors (EG&G Ortec). ^{226}Ra and ^{137}Cs were determined by γ-spectrometry using calibrated geometries on an n-type coaxial HPGe detector. ^{226}Ra was quantified from ^{214}Pb (351.9 keV line) after secular equilibrium in the radioactive chain was attained.

In both laboratories, excess (atmospheric) ^{210}Pb was determined by calculating the difference between total ^{210}Pb and ^{226}Ra activities. For all radionuclide activities (including both alpha and gamma spectrometry) and calculated values (such as excess ^{210}Pb, core inventories and fluxes) uncertainties were calculated by standard propagation of the 1 sigma counting and all analytical uncertainties of samples and blanks.

Interlaboratory study

In order to verify that the results provided by the two laboratories were consistent, an intercomparison exercise was carried out. Two soil samples from the province of Lleida were characterized for ^{210}Pb, ^{226}Ra and ^{137}Cs (Table 1). The Student-t test was used to compare the results from both laboratories and showed that they were not different ($t<1.96$) at the 0.05 confidence level.

Theoretical ^{222}Rn exhalation

Extensive measurements of ^{222}Rn exhalation rates, levels in soils or concentrations in buildings are available worldwide. Many countries have created maps of radon risks in their territory, which are based on direct measurements or on indirect indicators of indoor radon, such as geology, concentration of radium and radon in the ground and permeability. In Spain, a predictive map of ^{222}Rn concentrations indoors (4,5) has been developed and validated with experimental measurements. It is based on a theoretical model to estimate radon exhalation rates coupled with a simple model to simulate accumulation in a typical dwelling. The map that we propose for ^{222}Rn exhalation rates is built on the same basis as the map of indoor ^{222}Rn.

The ^{222}Rn exhalation caused by diffusive transport can be calculated according to:

$$J = E\sqrt{\lambda D}\ \rho\ C_{Ra-226}\tanh(\sqrt{\lambda / D}\ H), \qquad \text{(Eq. 1.)}$$

E being the emanation coefficient, λ the radon decay constant, D the diffusion coefficient for radon in the soil, ρ the soil density, $C_{Ra\text{-}226}$ the specific activity of ^{226}Ra in the soil, and H the height of the emanating object. For soils ($H \geq 2$ m), the term *tanh* can be considered equal to unity.

Average values of 0.25 to E and of 2.0 10^{-6} m^2s^{-1} to D were assigned for soils across the whole country, although both parameters vary widely with the moisture content of the medium (6). The above values are representative of soils composed of silty and clayish sands with a low moisture content. For soil density we have considered a value of 1400 kg m^{-3}.

The measurements from the Natural Gamma Radiation Map of Spain (MARNA) were used as a surrogate for concentrations of ^{226}Ra in soils. First, the exposure rate (T_{total}) due to gamma radiation from all emitters in the soil, measured in μR h^{-1}, was related to the gamma exposure rate due to ^{226}Ra alone ($T_{Ra\text{-}226}$), by means of the following equation, determined by a regression fit of experimental measurements (7):

$$T_{Ra-226} = 0.2703\, T_{total} - 0.1219 \qquad \text{(Eq. 2.)}$$

Next, to convert $T_{Ra\text{-}226}$ to ^{226}Ra concentration in the soil, the Microshield code v. 5.0.5. was applied to obtain the exposure rate 1 m above an infinite homogeneous soil with a ^{226}Ra content of 1 Bq kg^{-1}:

$$T_{Ra-226} = 0.051\, C_{Ra-226} \qquad \text{(Eq. 3.)}$$

Using Eq. 2 and Eq. 3, the 1.500.000 plus measurements of the MARNA map were transformed into ^{226}Ra concentrations in soils and these, in turn, into ^{222}Rn exhalation rates using Eq. 1. The resulting map is shown in Figure 1.

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Table 1: Interlaboratory study of ^{137}Cs, ^{210}Pb and ^{226}Ra in two soil samples. UVA: Universidad de Valladolid. UAB: Universitat Autònoma de Barcelona. The Student-t test values (t<1.96) indicate that all compared values do not differ at a 0.05 confidence level.

		A_{UVA} (Bq kg^{-1})	A_{UAB} (Bq kg^{-1})	t
Soil 1	^{210}Pb	126 ± 4	124 ± 5	0.31
	^{226}Ra	38.0 ± 1.7	39 ± 3	0.29
	^{137}Cs	29.3 ± 2.1	34.5 ± 1.7	1.92
Soil 2	^{210}Pb	52.6 ± 2.1	53.0 ± 2.2	0.13
	^{226}Ra	19.3 ± 1.5	15 ± 2	1.72
	^{137}Cs	24.7 ± 1.4	23.6 ± 1.1	0.62

Figure 1. Map of radon exhalation rates (*J*). The isolines shown correspond to levels of 0.016 Bq m^{-2} s^{-1} and 0.037 Bq m^{-2} s^{-1}, respectively. The red dots indicate the sampling locations of the soil cores.