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European ^{222}Rn inventory for applied atmospheric studies

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ABSTRACT

The radioactive noble gas ^{222}Rn, naturally emitted from land surfaces, is widely used as a tracer in characterising atmospheric transport and mixing processes. A constant and homogenous ^{222}Rn source of 1 atom cm^{-2} s^{-1} is generally assumed, sometimes less in northern latitudes. It is well known that ^{222}Rn flux varies in space and time but a robust description of it on a continental scale has not been possible before. Here, we present for the first time a spatially and temporally resolved ^{222}Rn source map for the European Continent (European Union, Norway, former Yugoslavia and Switzerland). It is based on the correlation between the ^{222}Rn flux and terrestrial γ-dose rate. Total γ-dose rate is monitored in this area at nearly 3600 stations continuously and the terrestrial component can be extracted from these measurements. On a resulting 0.5° × 0.5° map, mean annual values of ^{222}Rn flux ranged from 0.03 to 1.76 atom cm^{-2} s^{-1}, half of the values being between 0.40 and 0.70 atom cm^{-2} s^{-1}. The source strength was patchy but exhibited a decreasing trend with increasing latitude. Large values were mainly found on the Iberian Peninsula, small values along coasts and in northern and eastern parts of Europe. The seasonal amplitude in ^{222}Rn flux south of 55°N was small in 2006 with weekly averages deviating less than ±15% from the annual mean. Between 65°N and 70°N, weekly means are 2.5 times larger in summer than in winter.

1. Introduction

The radioactive noble gas ^{222}Rn has been widely used in the evaluation of atmospheric chemistry and transport models (Jacob et al., 1997; Dentener et al., 1999; Bergamaschi et al., 2006). Information on location, extent and strength of sources and sinks of greenhouse gases can be obtained when such models are run in an inverse mode (Gurney et al., 2002; Rödenbeck et al., 2003; Bousquet et al., 2006; Hirsch et al., 2006). Also, the accumulation of ^{222}Rn and other gases in the nocturnal boundary layer has been extensively used to estimate the emissions of CO_2, CH_4, N_2O, Hg° and other species by mass balance approach (Moriizumi et al., 1996; Schmidt et al., 1996, 2001, 2003; Wilson et al., 1997; Levin et al., 1999; Biraud et al., 2000, 2002; Conen et al., 2002; Obrist et al., 2006; Sturm et al., 2006). Currently, the effective use of ^{222}Rn in these contexts is limited by the poor accuracy of the ^{222}Rn inventory. Mostly, tuning and validation of atmospheric chemistry and transport models have relied on the simple assumption of a ^{222}Rn flux from land surfaces of 1 atom cm^{-2} s^{-1} between 60°S and 60°N, and 0.5 atom cm^{-2} s^{-1} between 60°N and 70°N (Rasch et al., 2000, e.g.). A modification of this inventory proposed a linear decrease from 1 atom cm^{-2} s^{-1} at 30°N to 0.2 atom cm^{-2} s^{-1} at 70°N (Conen and Robertson, 2002). A more differentiated inventory was suggested by Schery and Wasiolek (1998), based on porous media transport theory and calibrated with direct flux measurements from Australia and Hawaii. This inventory has so far remained in a preliminary state because of a lack of required input data. Gupta et al. (2004) have shown that the widely used assumption cited in Rasch et al. (2000) tends to overpredict atmospheric ^{222}Rn concentrations in both hemispheres. Robertson et al. (2005) provided evidence that using a northwards decreasing flux rate may improve model simulations sometimes but not always. Therefore, a more detailed and robust ^{222}Rn inventory description is desirable (WMO, 2004). Direct measurements of ^{222}Rn flux over large and heterogenous areas are expensive. However, ^{222}Rn flux can be described as a function of terrestrial γ-dose rate (Szegvary et al., 2007b). Total γ-dose rate is measured continuously at over 3600 stations in national emergency monitoring networks in Europe. Knowledge about type and make of detectors used in the contributing countries, their elevation above sea level and the possible contribution of artificial radiation (^{137}Cs), enable the extraction and spatial modelling of the terrestrial natural component of the γ-dose rate (Szegvary et al., 2007a). The area for which terrestrial γ-dose rates can currently be described includes the European Union, Norway, former Yugoslavia and Switzerland (from here onward more briefly: Europe).

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2. Constraining the European ^{222}Rn inventory

2.1. Approach

Terrestrial γ-dose rate is a good proxy for ^{222}Rn flux (Szegvary et al., 2007b). It is mainly determined by contributions from the decay of ^{40}K, ^{238}U and ^{232}Th including their progeny. The proportional contribution of ^{238}U and its progeny, one of which is ^{222}Rn, to the terrestrial γ-dose rate is more or less constant throughout Europe. It is on average 29% in northwest Italy (Chiozzi et al., 2002), 27% in Spain (Quindos Poncela et al., 2004) and in Germany (Franke, 2002), and 30% in Cyprus (Tzortzis et al., 2003). Continuous monitoring of the total γ-dose rates in Europe has intensified after the Chernobyl powerplant accident. Data from these national monitoring networks are collected at the Joint Research Center of the European Commission in Ispra (Italy). It is made available and stored on the European Radiological Data Exchange Platform (EURDEP) (de Cort and de Vries, 2005). Reported values are total γ-dose rates. They include a terrestrial component (mainly from ^{40}K, ^{238}U and ^{232}Th and their progeny), a cosmic component (mainly muons), an anthropogenic component (mainly from ^{137}Cs) and an inherent background of the measurement devices. To derive the terrestrial natural γ-dose rate, all other components have to be subtracted. This was done for all data of the year 2006 in the EURDEP database to generate maps of weekly means of the terrestrial γ-dose rate by ordinary kriging (Szegvary et al., 2007a). These maps were transformed into maps of mean weekly ^{222}Rn flux according to an empirical regression equation ($r^2 = 0.55$) based on parallel measurements of γ-dose rate and ^{222}Rn flux at 58 locations in Switzerland, Germany, Hungary Finland and Scotland (Szegvary et al., 2007b). The regression equation (Eq. (1)) reads:

$$^{222}\text{Rn flux}\left[\text{atom cm}^{-2}\ \text{s}^{-1}\right] = a\gamma\text{dose rate}\left[\mu\text{Sv h}^{-1}\right] - b \qquad (1)$$

$a = 11.8(\pm 1.3)$; $b = 0.15(\pm 0.11)$.

The measure of uncertainty given in the parentheses are ±1 standard error of the dimensionless factors a and b.

We applied a correction factor of 0.3 to peat soils as they are defined in the European Soil Database (European Soil Bureau Network and the European Commission, March 2004), because only these soil types were found to be significantly overestimated by Eq. (1). The correction factor was determined by comparing measurements made on peat soils with measurements on mineral soil in the same region. The correction factor of 0.3 is again an empirical factor, derived from measurements made in Finland and Switzerland (Szegvary et al., 2007b). Blank rocks were assigned a zero ^{222}Rn flux because the emanating surface is very small, leading to negligible exhalation rates.

In the following, we discuss ^{222}Rn flux maps with a spatial resolution of 0.5° × 0.5° and a temporal resolution of 1 week. While we have confidence in Eq. (1) to reliably predict differences in ^{222}Rn flux between regions (Szegvary et al., 2007b, Table 2), its application to predict temporal changes is uncertain. First because of the small number of longer term measurements we have made so far. Secondly, their duration of a few weeks in most cases, and thirdly, because they have been limited to a small area in the center of Europe. We may provide a tentative analysis for Switzerland. There are four stations, where we have at least four weeks of continuous measurements. Fitting Eq. (1) to weekly means at each of these stations separately, provides us values for a in Eq. (1) of 26.6, 15.5, 11.9 and 10.4. Fitting Eq. (1) to the ensemble the other 18 stations where we have short term measurements only, gives a value of 11.5 (standard error = 0.3) for a in Eq. (1). Hence, it may well be that temporal changes in γ-dose rate are associated with larger changes in ^{222}Rn flux than spatial changes in γ-dose rate. Yet, current evidence is not strong enough to justify or provide a separate model for temporal changes.

2.2. Inventory description

First, we will describe the mean annual ^{222}Rn flux, focusing on its spatial heterogeneity. Second, we will look at temporal variations, and third, we will compare our new ^{222}Rn inventory to previous inventories. The regional distribution of mean annual ^{222}Rn fluxes in Europe during the year 2006 exhibits a considerable heterogeneity (Fig. 1). Also, flux values are mostly below the commonly assumed 1 atom cm^{-2} s^{-1}. Small values in the northern parts of Europe may be caused by high soil moisture contents shielding parts of the terrestrial γ-dose as well as restricting diffusion of ^{222}Rn from soil to atmosphere. A further cause is certainly the abundance of organic soils with a small mineral component and resulting low γ-dose and ^{222}Rn flux rates. Some coastal regions towards the Atlantic and the North Sea are most likely weak ^{222}Rn sources because of the low concentrations of radionuclides in the quarternary sand deposits (Kemski et al., 2001; Franke, 2002) on which soils in some of these regions have formed. Large ^{222}Rn fluxes can be found on the Iberian Peninsula, where dry soil conditions and soils developed on crystalline rocks generate high γ-dose and ^{222}Rn flux rates (Quindos Poncela et al., 2004). But since no ^{222}Rn flux measurements were available for the Iberian Peninsula, these measurements should be used with caution. Even in the northern parts of Europe, in Sweden and Southern Finland large values can be found. They are well correlated with the prevalence of crystalline rocks (e.g. granite) (Asch, 2005). The 'hot spot' region near Rome, Italy, may be explained by strong volcanic influence as described in Voltaggio et al. (2006). Half of the 0.5° × 0.5° grid values (Fig. 1) are between 0.40 and 0.70 atom cm^{-2} s^{-1}. The distribution is close to normal with a mean (0.55 atom cm^{-2} s^{-1}) only slightly larger than the median (0.51 atom cm^{-2} s^{-1}).

2.3. Temporal variation

Being continuous, the γ-dose rate measurements allowed us to estimate the temporal variation in ^{222}Rn flux in Europe. To somehow structure the data for presentation, we produced time series of weekly averages for 0.5° latitudinal and 0.5° longitudinal bands (Fig. 2). The graphs indicate not only a generally decreasing trend of the ^{222}Rn flux rate with increasing latitude but also a growing amplitude of the seasonal cycles at northern latitudes. Seasonality in ^{222}Rn flux was found mostly in northern regions,

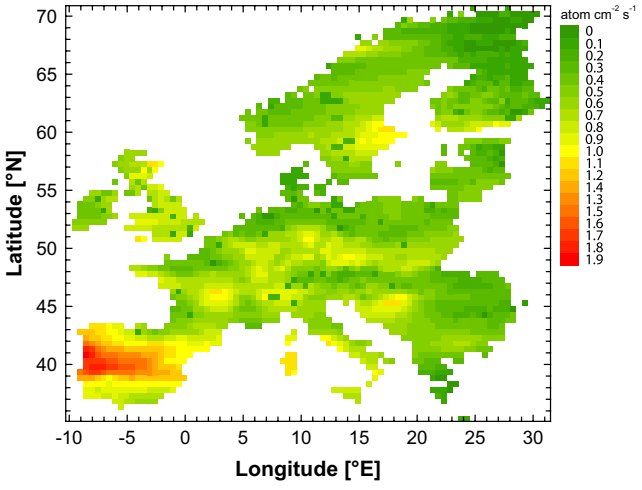

Fig. 1. Estimated mean annual ^{222}Rn flux in Europe for the year 2006 (data available at: http://radon.unibas.ch).