EUROPEAN ^{222}RN FLUX MAP FOR ATMOSPHERIC TRACER APPLICATIONS

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Abstract

^{222}Rn is commonly used as a natural tracer for validating climate models. Generally, a constant and homogenous ^{222}Rn source term of 1 atom cm^{-2} s^{-1} is assumed as a standard, sometimes reduced in northern latitudes. A tendency to overestimate measured ^{222}Rn concentrations by simulations with this standard assumption has often been found. To improve current models of atmospheric chemistry and transport a better source term for ^{222}Rn than currently used is necessary. This work aimed to establish a method for mapping the ^{222}Rn source term by using a commonly measured proxy, the terrestrial γ-dose rate. A relatively stable fraction ($\approx$20%) of the total terrestrial GDR originates from the ^{238}U decay chain, of which ^{222}Rn is a member.

In this study a regression model could be established by simultaneous measurements of ^{222}Rn flux and terrestrial GDR at locations in Switzerland and Germany. This model was validated on a regional scale by measurements in Finland and Hungary, at locations covering wide ranges of γ-dose rates. The predictions were within the error margin of measurements, and therefore considered to suffice to produce regional means of ^{222}Rn flux by using γ-dose rate as a proxy. To be able to develop a ^{222}Rn flux map for Europe, a base map for the γ-dose rate was necessary. For this instance, we used the large number of national γ-dose rate measurements, established after the nuclear reactor accident in Chernobyl in 1986. These data are composite values of terrestrial, cosmic and anthropogenic contributions and instrument background (self-effect). We extracted the terrestrial part of the total γ-dose rate provided by the EUropean Radiological Data Exchange Platform (EURDEP), which continuously udates and stores the data. Subsequently we produced annual, seasonal and weekly γ-dose rate maps for Europe (European Union, Norway, former Yugoslavia and Switzerland) with geostatistical methods.

The regression model was then used to transform the terrestrial γ-dose rate maps into ^{222}Rn flux maps, using also additional information (organic/mineral soil, bare rock surface). Spatially and temporally resolved ^{222}Rn source maps for the European Continent resulted, with a spatial resolution of 0.5° x 0.5°. Previously made studies could be confirmed, and even more information was available now: modeled ^{222}Rn flux ranged from 0.03 to 1.76 atom cm^{-2} s^{-1}, with a coefficient of variation of 51% and half of the values were between 0.40 and 0.70 atom cm^{-2} s^{-1}. The weekly ^{222}Rn flux maps were applied in a simulation with the atmospheric transport model TM5, as well as the standard assumption of 1 atom cm^{-2} s^{-1} (with 0.5 atom cm^{-2} s^{-1} between 60°N and 70°N). The results from TM5 showed that our spatially resolved ^{222}Rn source term can improve predictions of atmospheric ^{222}Rn concentrations. In a case study in Gif-sur-Yvette (France) one week of ^{222}Rn concentrations were observed. The air mass trajectories turned (a) from areas with large (0.61 atom cm^{-2} s^{-1}) to (b) areas with small (0.30 atom cm^{-2} s^{-1}) ^{222}Rn fluxes. The standard assumption overpredicted atmospheric concentrations by (a) 70% and (b) 260%, while the simulation based on the new inventory followed the observation closely.

On the basis of our approach we also produced ^{222}Rn flux maps for the United States of America and the Russian Federation territory, which are still preliminary and await verification.

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Chapter 1

Introduction

Information about regional sources and sinks of greenhouse gases are increasingly derived from variations in their atmospheric concentrations by means of inverse modelling with atmospheric transport models (Gurney *et al.*, 2002). Calibration and validation of such models is generally performed with an inert tracer, such as ^{222}Rn (Genthon and Armengaud, 1995; Li and Chang, 1996; Jacob *et al.*, 1997; Stockwell *et al.*, 1998; Dentener *et al.*, 1999; Rasch *et al.*, 2000). The natural radioactive noble gas ^{222}Rn is part of the ^{238}U decay series and produced by the decay of ^{226}Ra, a trace element present in all mineral soils. Its emission from ice-free land surfaces is about 2-3 orders of magnitude larger than from oceans. Unlike the biogenic trace gases CO_2, CH_4 or N_2O, which are very reactive in the atmosphere, geogenic ^{222}Rn has much simpler source and sink functions. This makes ^{222}Rn, together with its source distribution and short half life ($t_{1/2}$=3.82 days), a useful tracer in atmospheric transport studies, an application first proposed by Israël (1951). Therefore, it is important to have good knowledge of its source term.
Direct measurements of ^{222}Rn flux carried out on local scales, as summarised in Wilkening *et al.* (1975); Turekian *et al.* (1977); Conen and Robertson (2002), have indicated large spatial and temporal variations. The current practice is to assume a spatially and temporally uniform flux rate of 1 atom cm^{-2} s^{-1} from all ice-free land surfaces, with a lower ^{222}Rn flux rate above 60°N (mainly due to higher soil moisture content). It is generally acknowledged that this undifferentiated source term description is limiting the validation of atmospheric transport models. Generally, a more detailed source term is highly desirable to improve the validation of atmospheric transport models since the quality of validation is directly proportional to the quality of the ^{222}Rn source term used (WMO, 2004).
This study suggests and describes the European ^{222}Rn source term. The approach is to calibrate ^{222}Rn flux in a number of direct measurements against terrestrial γ-dose rate, a widely measured proxy, and to use the high density of European γ-dose rate measurements, established after the nuclear reactor accident in Chernobyl in 1986, to produce a full description of the European ^{222}Rn source term.
γ-dose rate is related to factors also affecting ^{222}Rn flux, such as soil ^{226}Ra concentration and soil moisture. Increasing soil moisture, for example, increases the attenuation of γ-rays emitted from soil, reducing γ-dose rate above ground. This correlation has been exploited to track changes in soil moisture with airborne γ-dose meters (Peck *et al.*, 1992). Simultaneously, large soil moisture contents reduce soil diffusivity (Lehmann *et al.*, 2000) and thus ^{222}Rn flux. Yet, γ-dose rates are also affected by other factors, for example ^{40}K and ^{232}Th concentrations, which have no influence on ^{222}Rn flux, and vice versa. Nevertheless, of all widely measured environmental parameters, γ-dose rate has been found

to be most closely related to ^{222}Rn flux and able to explain around 60% of its spatial variability (Schery *et al.*, 1989; Nielson *et al.*, 1996). Correlation coefficients presented in these studies are lower estimates as they are based on parallel spot measurements of γ-dose rate and ^{222}Rn flux. Short term fluctuations resulting from rain are asynchronous for both parameters and larger correlation coefficients are expected over longer observation periods.
The research plan consisted of three major parts:

Part 1 Description of ^{222}Rn flux as a function of γ-dose rate based on direct measurements of both parameters and verification of the description, or predictive model, at locations where ^{222}Rn has not been measured during the establishment of the model(chapter 2).

Part 2 The development of a spatial and temporal European terrestrial γ-dose rate map, which provides a basis for the ^{222}Rn flux map (chapter 3).

Part 3 Application of the predictive model to European terrestrial γ-dose maps and their translation into a ^{222}Rn flux map of the region (chapter 4). Furthermore, the application to an atmospheric transport model, and the results compared to the commonly used method of 1 atom cm^{-2} s^{-1} are shown.

Part 1 involves simultaneous measurements of ^{222}Rn flux and γ-dose rate at various locations to establish a regression model. The predictive quality of this model for ^{222}Rn flux based on γ-dose rate data was validated at locations not visited previously. Part 2 investigates an important issue, as the accuracy of the resulting ^{222}Rn flux map, which should be achieved in part 3, is directly related to the preciseness of the extraction of the terrestrial γ-dose rate from the total measurements of the γ-dose rate. The total measured γ-dose rate consists of dose rates deriving from cosmic rays, artificial radiation, a terrestrial part and an inherent background from material of the measurement device. Before part 3 can be attempted, several iterations between parts 1 and 2 might be necessary and lead to the inclusion of other parameters for which Europe-wide data is available. Candidate parameters are climate and pedological features. We are aware of intricacies in this approach, including the possible short-term asynchrony in fluctuations of γ-dose rate and ^{222}Rn flux caused by precipitation events. However, unlike other methods that have been tried to describe ^{222}Rn flux, the one proposed here would allow the creation of a European flux map and the approach could also be applied to other continents.

Chapter 2

Predicting terrestrial ^{222}Rn flux using γ-dose rate as a proxy

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2.1 Abstract

^{222}Rn is commonly used as a natural tracer for validating climate models. To improve such models a better source term for ^{222}Rn than currently used is necessary. The aim of this work is to establish a method for mapping this source term by using a commonly measured proxy, the γ-dose rate (GDR). Automatic monitoring of GDR has been networked in 25 European countries by the Institute for Environment and Sustainability at the Joint Research Centre (JRC IES) in Ispra, Italy, using a common data format. We carried out simultaneous measurements of ^{222}Rn flux and GDR at 63 locations in Switzerland, Germany, Finland and Hungary in order to cover a wide range of GDR. Spatial variations in GDR resulted from different radionuclide concentrations in soil forming minerals. A relatively stable fraction (20%) of the total terrestrial GDR originates from the ^{238}U decay series, of which ^{222}Rn is a member. Accordingly, spatial variation in terrestrial GDR was found to describe almost 60% of the spatial variation in ^{222}Rn flux. Furthermore, temporal variation in GDR and ^{222}Rn was found to be correlated. Increasing soil moisture reduces gas diffusivity and the rate of ^{222}Rn flux but it also decreases GDR through increased shielding of photons. Prediction of ^{222}Rn flux through GDR for individual measurement points is imprecise but un-biased. Verification of larger scale prediction showed that estimates of mean ^{222}Rn fluxes were not significantly different from the measured mean values.

2.2 Introduction

^{222}Rn is commonly known as a hazardous radioactive (noble) gas in indoor air. Yet, ^{222}Rn is also often used as a natural tracer of air transport. Observations of atmospheric ^{222}Rn have been very useful in the evaluation of climate models simulating transport, transformation and removal processes of gases and aerosols (Rasch *et al.*, 2000, e.g.). Used in inverse mode, these models can provide information on location, extent and strength

of sources and sinks of greenhouse gases based on the measurement of changes in their atmospheric concentrations (Chevillard *et al.*, 2002; Gupta *et al.*, 2004). Currently, the effective use of ^{222}Rn in this context is limited by the poor accuracy of the ^{222}Rn source function (WMO, 2004). Current practice is to assume a spatial and temporal uniform flux rate of 1 atom cm^{-2} s^{-1} from all ice-free land surfaces. Improvement of the source term was attempted by Schery and Wasiolek (1998), who created a global ^{222}Rn flux map based on porous media transport theory and calibrated with experimental ^{222}Rn flux data from Australia and Hawaii. It predicted regional variations of a factor of three not to be uncommon. However, current lack of detailed data on input parameters in large parts of the world results in the proposed map still being preliminary and depending on more data becoming available. Furthermore, additional flux measurements over a greater variety of conditions are needed for robust validation and eventual verification of the model. A different interpretation of the flux term was proposed by Conen and Robertson (2002), based on atmospheric profile measurements integrating over larger areas and indicating a decline in ^{222}Rn flux from ice-free land surface from 1 atom cm^{-2} s^{-1} at 30°N to 0.2 atom cm^{-2} s^{-1} at 70°N. This source term was found to improve predictions but it was speculated that ^{222}Rn flux might begin to decline well north of 30°N (Robertson *et al.*, 2005). A more detailed source term is highly desirable to improve validation of atmospheric transport models since the quality of validation is directly proportional to the quality of the ^{222}Rn source term used. Therefore, we are proposing a new method to describe the ^{222}Rn source term, initially focusing on the European continent. Our approach is to calibrate direct measurements of ^{222}Rn flux against terrestrial γ-dose rate (GDR). We made use of the high density of European GDR measurements, established after the nuclear reactor accident in Chernobyl in 1986, to produce a full description of the European ^{222}Rn source term.

2.3 Basic Concept

The source of ^{222}Rn is ^{226}Ra, a member of the ^{238}U decay chain. Gamma spectroscopic analysis of soil surface samples (0-20 cm depth) in geologically diverse regions of Switzerland showed that ^{238}U contributes an almost constant proportion to the terrestrial GDR (Figure 2.1a) and that ^{226}Ra activity is closely related to the ^{238}U activity (Figure 2.1b). Large radioactive disequilibria of the uranium decay series have been found in the limestone Karst soils in the Jura mountains (Von Gunten *et al.*, 1996). Selective migration of individual members of the ^{238}U decay chain could lead to an over- or underestimated GDR-based ^{222}Rn flux in such locations. However, such cases seem to be rare, as seen in the close correlations in Figure 2.1a & b. The proportion of the contribution of the ^{238}U series to total γ-dose rate is also reported for North-West Italy in Chiozzi *et al.* (2002), for Spain in Quindos Poncela *et al.* (2004) and for Cyprus in Tzortzis *et al.* (2003). Contributions of the ^{238}U series for individual types of rocks reported in these four studies range from 12% to 90%. However, the average for each country or region ranges from 27% (Spain), 29% (North-West Italy) to 30% (Cyprus, Switzerland). Thus, in the context of our objective to predict larger scale averages for ^{222}Rn flux, it seems justified to assume a constant contribution of the ^{238}U series to the total γ-dose rate. Therefore, we assume that ^{222}Rn flux resulting from the decay of ^{226}Ra is directly related to terrestrial GDR. This assumption is probably a good first approximation but not entirely correct as indicated by the relatively large scatter in the ratio of ^{222}Rn flux to ^{226}Ra activity (Figure 2.1c). Firstly, only part of the produced ^{222}Rn emanates into air filled pore space from

where it might escape into the atmosphere and the fraction emanating may depend on grain size (Nazaroff, 1992). Secondly, differences in grain size and soil moisture modulate gas diffusivity and thus the fraction of emanated ^{222}Rn that may reach the atmosphere before decay. Thus, the proportion of ^{222}Rn produced that escapes into the atmosphere is variable and depends on factors other than ^{226}Ra content. Indeed, the emanation coefficient for radon can vary by a factor of 10. The magnitude of this variation is a question of scale. Greeman and Rose (1996) determined emanation coefficients for each horizon in 12 contrasting soil profiles in the North-East of the United States. Emanation coefficients ranged from 5.5% to 33% for individual horizons. However, average emanation coefficients for entire soil profiles only ranged from 13% to 29% and two-thirds of the soil profiles were in the narrow range between 18% and 22%. Hence, despite large differences at the small scale, emanation factors at larger scales seem to be within a narrow range.

2.4 Methods

2.4.1 ^{222}Rn flux measurement techniques

A barely modified closed chamber method as described in Lehmann *et al.* (2000, 2003) was used to measure the ^{222}Rn flux. The main modification consisted in air from the chamber not being pumped through a series of two but only one alpha-decay detector (Alphaguard 2000 Pro, Genitron Instruments Frankfurt, Germany). The flow rate was 0.5 litres min^{-1}, a delay volume of 1.5 litres was used to remove most of the ^{220}Rn with its half-life of 56 s (Lehmann *et al.*, 2003) used the second detector, which was installed before the delay volume to evaluate also the ^{220}Rn flux). From there, the air passed to the detector where only ^{222}Rn was measured. The ^{222}Rn flux was estimated from the increase in ^{222}Rn activity measured in 10 min intervals over about 1.5 hours. Remaining ^{220}Rn may have affected the absolute value of measured ^{222}Rn activity but not its increase over time, as ^{220}Rn concentrations reach a steady state between production and decay after about 7 min and we always rejected the first 10 min measurement interval. Due to radioactive decay of ^{222}Rn with a half-life time of 3.82 days the assumption of a linear increase of ^{222}Rn in the chamber must be corrected by a factor of +0.38%. Two types of chambers were used: an automatically closing and opening chamber which measured autonomously the ^{222}Rn flux from soil over a longer time period. This flux chamber, a cylindrical box with a diameter of 20 cm and 25 cm height had a flap, which closed automatically 6 times a day for 1.5 hours to accumulate ^{222}Rn and was then opened for 2.5 hours prior to the next measurement. A second analytical system was a manually closable chamber (a plastic box with the dimensions 35 cm x 27 cm and 13 cm height) which was used for spot measurements. The instrument we used in our study was compared in 2003 (Robertson, 2005, PhD Thesis) to an instrument which has been widely used in East-Asia. The mean flux determined at six locations was 52 Bq m^{-2} h^{-1} (standard error 9 Bq m^{-2} h^{-1}) with our instrument and compared well with the mean flux of 49 Bq m^{-2} h^{-1} (standard error 8 Bq m^{-2} h^{-1}) measured with the instrument described in Iida *et al.* (1996). Long-term measurements of ^{222}Rn fluxes were made at 7 different field sites of the Swiss Meteorological Service (MétéoSuisse). Normally, measurements took place for a duration of 3-4 weeks, except at the field site in Basel-Binningen, where continuous measurements were made over a year in order to estimate seasonal variations. Soil moisture at that location was measured with 4 TDR two-rod probes (rod length: 18 cm), connected with a multiplexer to a Tektronix 1502B (Tektronix, Inc., Wilsonville, USA). The signal was evaluated and logged with a

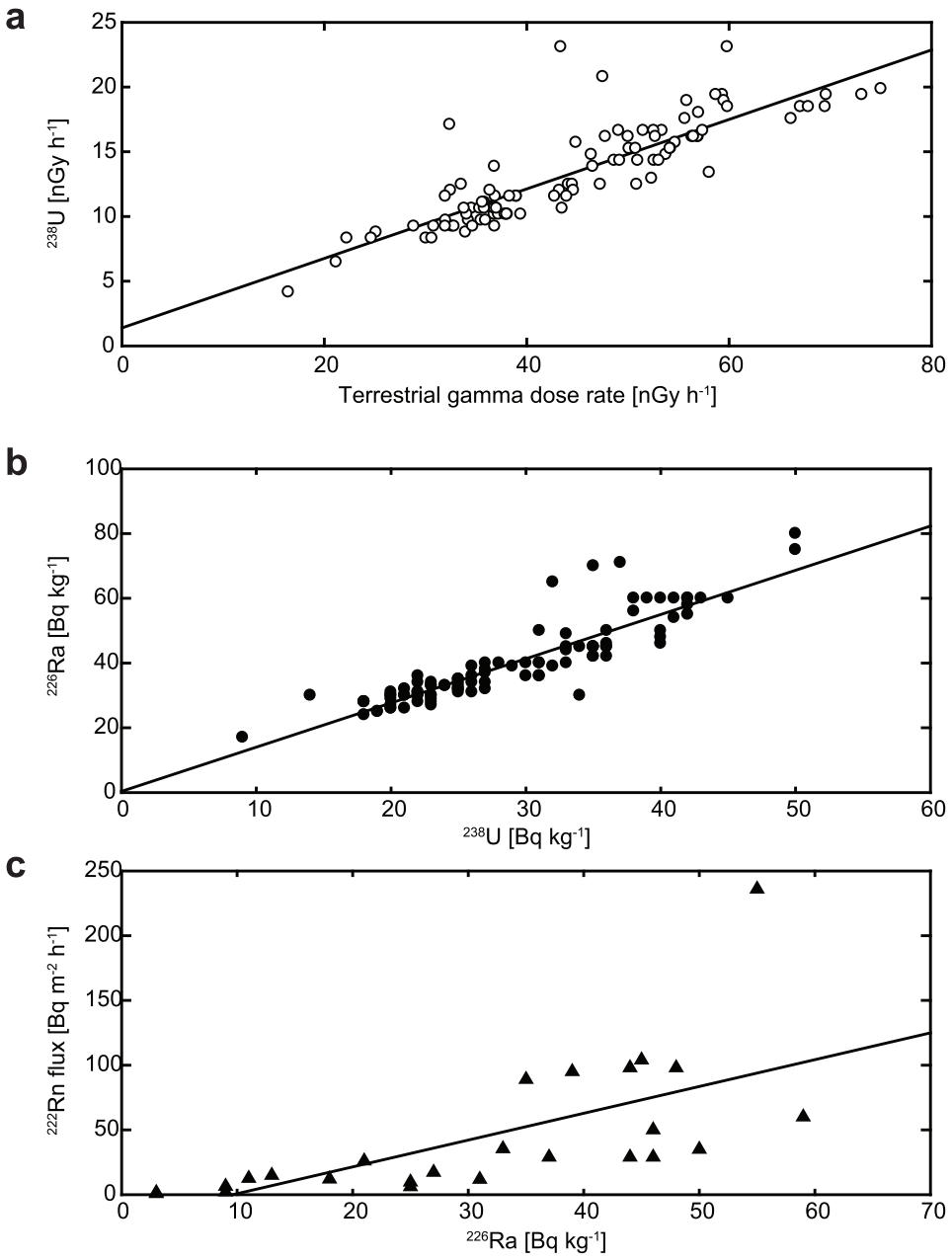

Figure 2.1: Correlations between (a) the contribution of GDR originating from the ^{238}U decay series and total terrestrial γ-dose rate; (b) ^{226}Ra activity and ^{238}U activity and (c) ^{222}Rn flux at the soil surface and soil ^{226}Ra activity. Data for (a) and (b) was kindly provided by SUER (Section of Surveillance of Radioactivity, Switzerland)

data logger (CR10, Campbell Scientific, Inc., USA). The manually closing chamber was used for in situ measurements of ^{222}Rn flux at 29 sites in Switzerland and South-West Germany, at 8 sites each in Southern (Helsinki region) and Northern Finland (Rovaniemi region) and at 12 sites in Hungary. Supplementary data from Scotland (Robertson *et al.*, 2005) was included. These measurements (n=9) were done with the same analytical ^{222}Rn system. The difficulty of spot measurements of ^{222}Rn flux and GDR is to get representative values for the specific location. Especially precipitation has been found to have significant effects on GDR because of the deposition of Rn daughters associated with aerosols, but also on short-term variations in ^{222}Rn flux. Therefore, we avoided spot measurements during or immediately (4-8 h) after precipitation events. Additionally we studied on small scale spatial variability in a woodland in Basel (Lange Erlen) using a nested sampling design with lag distances of 0.5 m, 5 m and 50 m.

2.4.2 γ-dose rate

An autonomous gamma probe (Gammatracer, Genitron Instruments Frankfurt, Germany) for continuous surveillance of the environmental gamma radiation was used for measuring GDR (10 H*). The gamma probe was placed 1m above ground during the measurement. Since most of the measurements took place at locations of the national gamma monitoring networks, where GDR is continuously measured, the gamma probe was used as a reference probe. This allowed inter-comparison of different probes at the network sites. The terrestrial component of the γ-dose rate was obtained by subtracting the cosmic part (which depends on altitude above sea level and can be calculated) from the measured total GDR (Murith and Gurtner, 1994). A correction was made for the artificial radiation, which is mainly derived from ^{137}Cs from the Chernobyl powerplant accident in 1986, based on the "Atlas of Caesium deposition on Europe after the Chernobyl accident" (de Cort *et al.*, 1998).

2.5 Results and discussion

2.5.1 Correlation of ^{222}Rn flux and terrestrial GDR at different locations

The results of the measurement campaign are shown in Table 2.1, containing field site information and soil properties (all data concerning this research can also be found on the website `http://radon.unibas.ch`). There is a linear relationship between ^{222}Rn flux and terrestrial GDR (Figure 2.2), though the effect of heteroscedasticity is observed, i.e. the variability described by standard deviation depends on the mean value. This means high GDR values are associated with higher variability (an effect, which is often observed in nature).
The measured data covers a range from almost 0 to 200 nSv h^{-1} respectively 0 to 250 Bq m^{-2} h^{-1}. Most soils in Europe have γ-dose rates between about 40 to 140 nSv h^{-1} well within this range. Very low GDR (40 nSv h^{-1} and a ^{222}Rn flux less than 15 Bq m^{-2} h^{-1}) can be found at locations which have either a high water content and/or low or no mineral content like peat soils. Overall, almost 60% of the variation in ^{222}Rn flux can be described by the spatial variation of terrestrial GDR. Still, there is a lot of variation, which may also be caused by the gamma probe and the ^{222}Rn measurement chamber integrating over different soil volumes. The measurement of GDR is mostly influenced by the variability of radionuclides and soil moisture near the soil surface (0 to 0.1 m)

Field site information			^{222}Rn flux and GDR		Particle size fractions [%]			
Long.	**Lat.**	**Elevation asl**	**^{222}Rn flux**	**Terr. GDR**	**Sand**	**Silt**	**Clay**	**Moisture**
[°E]	[°N]	[m]	[Bq m^{-2} h^{-1}]	[nSv h^{-1}]	(63-2000μm)	(2-63μm)	(0-2μm)	[wt%]
Switzerland								
7.58	47.54	316	66*	88	5.9	71.4	22.7	21.3
7.88	47.43	610	14	57	25.7	55.4	18.9	17.6
7.88	47.43	610	14*	64	25.7	55.4	18.9	17.6
6.67	46.51	461	91*	93	30.9	54	15.1	
7.74	47.29	453	87*	84				
7.42	46.93	565	84*	83	45.2	37.1	17.7	
7.42	46.93	565	66	63	45.2	37.1	17.7	
6.92	46.33	381	37	96	5.5	82.4	12.1	
6.92	46.33	381	13	92	5.5	82.4	12.1	
7.84	46.3	640	50*	108	44.3	52.6	3.1	
6.58	46.84	1202	49	65	37.2	47.2	15.6	21.8
6.79	47.08	1018	67	73	6.3	70.3	23.4	25.8
6.23	46.4	430	92	89	25.2	49.3	25.6	15.3
9.84	46.81	1590	18	65	54.2	33.8	12	29.1
9.88	46.53	1705	37	77	39.6	46.5	13.9	25.5
10.07	46.34	1201	98	92	48.5	34.4	17.1	23.4
7.79	47.26	422	83	66	33	41.8	8.8	23.9
8.31	46.5	1345	61	100	62.1	34.3	3.6	18.4
8.9	47.48	536	96	69	28.6	49.4	22	24.9
9.4	47.43	779	44	61	33.5	45.7	20.8	34.1
9.07	47.03	515	74	59	8.7	52.8	38.5	41.3
9.52	47.13	460	39	66	20.5	70.1	9.4	26.8
8.46	47.06	1040	7	33	17.3	39.5	43.3	
7.64	47.59	268	109	105				
Germany								
7.81	47.76	850	157	155				
8.00	47.66	700	30	82				
8.14	47.59	300	33	61				
7.95	47.56	280	58	62				
7.78	47.56	350	98	116				
7.82	47.65	500	89	127				
Finland								
25.29	60.39	11	189	132	2.6	39.6	57.8	20.3
26.22	60.46	30	84	191	70.8	23.4	5.8	8.2
26.05	60.44	6	124	166	87.3	7.4	5.3	12.5
23.79	61.51	112	55	135	9.1	48.9	42	24.7
24.04	61.27	94	60	94	23	36.2	40.9	16.2
24.29	60.89	110	51	100	25.9	61.6	12.5	17.4
22.37	60.45	37	134	104	17.4	40	42.7	23.9
23.98	60.47	37	108	124	1.5	42.5	56	16.9
28.14	66.14	250	12	80	67.5	25.1	7.5	53
26.76	66.37	118	95	82	8.5	69.2	22.3	18.8
25.79	66.51	61	49	81	58.2	37.2	4.5	11.1
26.91	65.4	118	6	53	33.9	41	25.2	76.4
26.47	65.95	160	6	50	42	49.5	8.6	48.9
26.64	67.41	173	35	45	52.9	42.7	4.4	26.7
27.33	66.72	162	2	43	35.4	58.1	6.5	54.9
24.85	66.12	27	53	73	38.4	51.5	10.1	8.4
Hungary								
17.67	47.71	121	29	86	54.5	37.1	8.4	3.2
18.41	47.56	182	108	100	60.1	25	14.9	4.3
19.14	47.94	227	80	108	12.9	60.9	26.2	9.6
19.54	48.1	163	44	89	52.5	31.7	15.9	9.6
19.79	48.05	225	47	94	30.7	44.4	24.9	2.4
17.89	47.1	260	79	93	25.5	57	17.6	5.2
17.47	47.35	144	25	70	48.6	31.2	20.2	13.8
20.27	47.73	127	23	70	82.6	9.5	7.9	3.6
20.77	48.1	230	111	91				11.8
20.26	48.23	176	73	92	27.7	47.2	25.1	7
18.61	47.76	112	50	88	44.9	45.4	9.6	8
18.80	47.55	203	52	92	12.6	68.1	19.3	21

* Longterm measurements in Switzerland

Table 2.1: Measurement results of field sites in Switzerland, Germany, Hungary and Finland

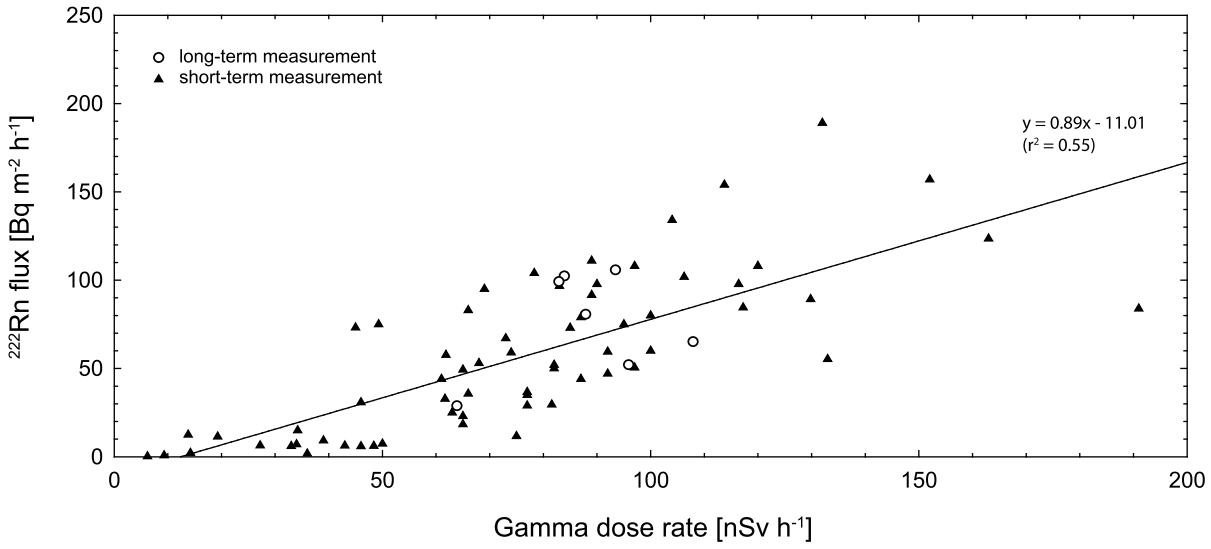

Figure 2.2: Correlation of ^{222}Rn flux and terrestrial γ-dose rate measured at field sites in Switzerland, Germany, Scotland, Finland and Hungary

within a radius of about 10 m around its location. In contrast, measured ^{222}Rn flux is mostly influenced by ^{226}Ra content and soil moisture in 0 to 1 m soil depth but a three to four orders of magnitude smaller area. Thus, inhomogeneities in radionuclide and moisture distribution on this scale will affect both parameters to a different extent. The scatter in Figure 2.2 is unlikely to be caused by short-term fluctuations in either parameter. Not only the short-term measurements (triangles) show the scattering effect, but also the long-term measurements (circles), which would smoothen out such short term effects. The nested sampling near Basel revealed that the coefficient of variation between measurements separated by a distance of 0.5 m was 19%, increasing to 21% and 36% for 5.0 and 50.0 m distances, respectively. The large coefficient of variation at the smallest distance may to a large part be caused by the error in our ^{222}Rn measurement, which we estimate to be around $\pm$15% of the mean. For atmospheric tracer applications, regional information on the ^{222}Rn flux is required. The variability in the correlation between GDR and ^{222}Rn flux, which can be found on the local scale, seem to counter balance on the regional scale, as discussed later (see chapter 4.4).

2.5.2 Correlation of ^{222}Rn flux and terrestrial GDR over time

Temporal variations in ^{222}Rn flux can be observed in GDR at the long-term measurement in Basel (Figure 2.3) during the period from June to November 2006, where soil moisture and precipitation was also measured. At the beginning of July a prolonged dry period began without nearly any precipitation and soil moisture decreased almost constantly. During this period the ^{222}Rn flux was observed to increase by about 100% until the beginning of August. Simultaneously, GDR increased from 82 nSv h^{-1} to 98 nSv h^{-1}, which is nearly 20%. Decreasing soil moisture increases the air filled pore volume and with it the diffusivity of soil. Therefore, ^{222}Rn flux is larger when soils are dry and less ^{222}Rn decays before it may reach the soil surface (Grasty, 1997). At the same time, low soil moisture leads to reduced shielding of gamma-rays and a larger proportion of them can be detected in the atmosphere above the ground. Diurnal changes in the amplitude

of GDR during periods without precipitation are supposed to be influenced by changes in Rn and Rn-progeny concentrations in the near surface air, where they accumulate during atmospherically stable conditions at night (Greenfield *et al.*, 2002, 2003a).

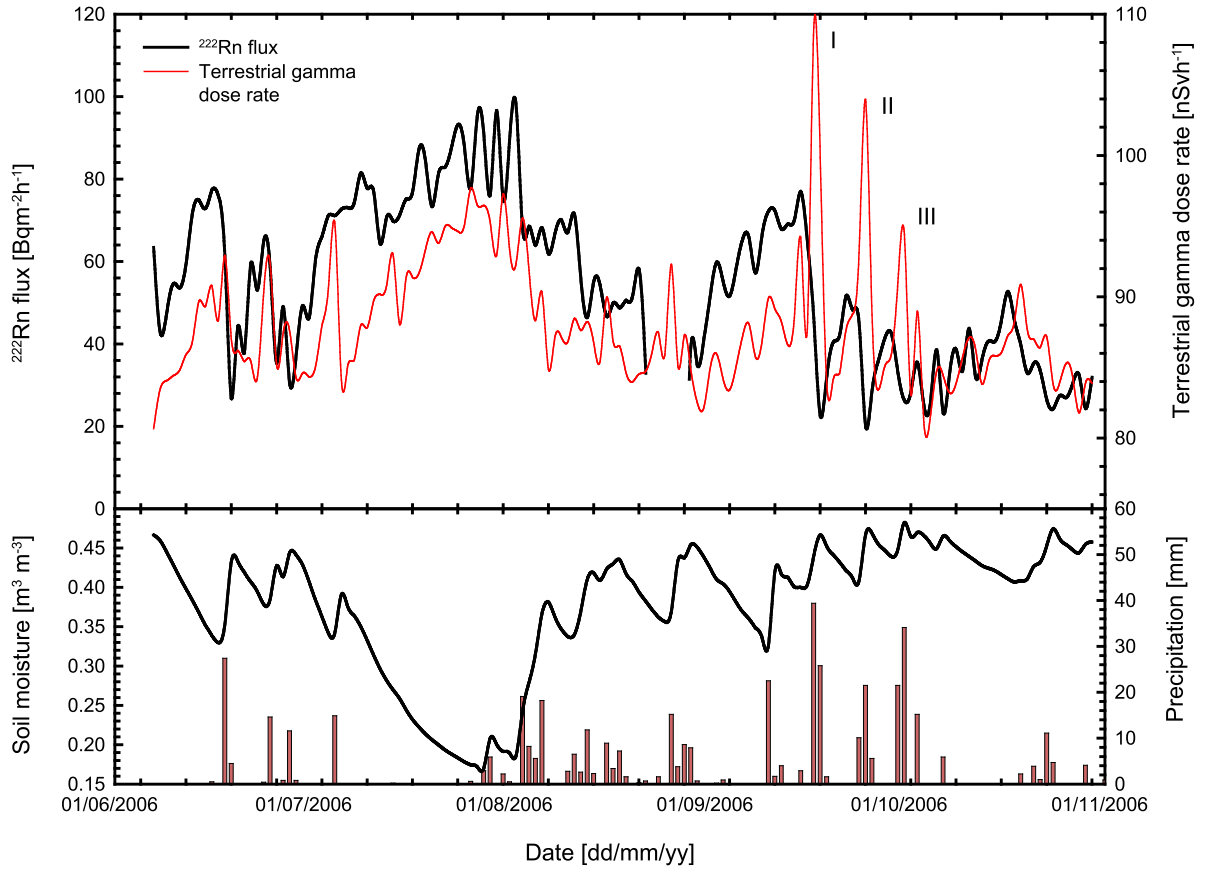

Figure 2.3: ^{222}Rn flux, terrestrial γ-dose rate, precipitation and soil moisture time series from June to November 2006 in Basel (Switzerland). Heavy rain events are marked with I, II and III

At end of September through the beginning of October three intense rain events were recorded (Figure 2.3). These were days within a period of otherwise stable weather conditions, where during a short time period between 60 mm and 80 mm of rain fell, approximately the same amount for all three rain events. After each of the three events, the ^{222}Rn flux decreased immediately with the beginning of precipitation, probably because of the wet soil surface severely inhibiting ^{222}Rn diffusion into the atmosphere. The reaction of GDR was initially to the contrary. It suddenly increased after the first rain event from 85 nSv h^{-1} to 110 nSv h^{-1}, an increase of 29%. This effect is caused by outwash of particles from the lower atmosphere, carrying previously absorbed ^{222}Rn progeny back to the soil surface (Greenfield *et al.*, 2002, 2003a). The cummulative half-life of the short-lived ^{222}Rn progeny is about 50 minutes. Thus, the GDR decreased within a few hours once rain had stopped and was lower than it was before the rain event (8-10%). The second and third rain event showed the same effect. The only difference between the three rain events was the amplitude of the peak at the start of each rain fall, which was smaller for the second and third compared to the first one because the atmosphere was getting increasingly cleaner of particles carrying ^{222}Rn progeny.

2.5.3 Factors affecting ^{222}Rn flux but not GDR

Our analysis of the correlation between ^{222}Rn and terrestrial GDR showed that both parameters are affected similarly by the radionuclide content of the soil and by soil moisture. However, there are also factors affecting ^{222}Rn flux without having a similar effect on GDR which we have not evaluated so far. Total pore space and tortuosity are important variables that affect ^{222}Rn flux (Nazaroff, 1992) but not GDR. A larger proportion of ^{222}Rn produced within the soil profile will escape to the atmosphere from coarse grained soils with a large total pore volume than from compacted fine grained soils, whereas the escape of γ-rays is unlikely to be affected by this. There already exist models for ^{222}Rn flux prediction based on geological and pedological factors, but such models require numerous parameters which are not well known due to the complicated interactions between different geological and pedological units influencing the ^{222}Rn flux (Ielsch *et al.*, 2002). Temperature differences between air and soil have also been found to be a factor influencing ^{222}Rn flux (Nazaroff, 1992), which is driven by diffusion and possibly mass flow. As for other possible correlations between environmental parameters and ^{222}Rn flux, we have tested for correlations with air temperature, atmospheric pressure, soil temperature and difference between air and soil temperature. If one of these parameters was correlated with ^{222}Rn flux, it was very weak. We do not think these parameters have a strong direct effect on ^{222}Rn flux but rather coincide with precipitation events or dry spells. In principle, diurnal pressure variations may cause mass flow through periodic expansion and contraction of the soil gas volume and influence the otherwise mainly diffusion-driven exchange of radon between soil pore space and atmosphere. We would expect this to be a major factor in deeply weathered dry soils with large air volumes. In the commonly humid regions in Europe we studied, it might not be a major issue.

2.5.4 Verification on a regional scale

As mentioned in the introduction, our interest in describing the ^{222}Rn flux term is because of its application in the validation of atmospheric transport models. We therefore would like to be able to correctly predict regional averages of ^{222}Rn flux. To test our approach of using GDR as a proxy, we split our data in one part to produce the correlation function between ^{222}Rn flux and GDR (Switzerland, Germany, Scotland) and another part to verify the correlation (N- and S-Finland, Hungary). The correlation function derived was:

$$y = 0.995(\pm 0.10) \cdot x - 14.97(\pm 8.11) \; (r^2 = 0.66) \tag{2.1}$$

, where y is the ^{222}Rn flux in Bq m^{-2} h^{-1} and x is the GDR in nSv h^{-1}. The measured regional means differed by a factor of up to 3, as considered not to be uncommon by Schery and Wasiolek (1998). Still, predicted means were within the error margin of the respective measured mean (Table 2.2), strongly supporting the effectiveness of our approach.

	^{222}Rn flux measured	**^{222}Rn flux predicted**	**n**
S-Finland	100 ±17 Bq m^{-2} h^{-1}	102 ±13 Bq m^{-2} h^{-1}	8
N-Finland	32 ±11 Bq m^{-2} h^{-1}	41 ±06 Bq m^{-2} h^{-1}	8
Hungary	60 ±09 Bq m^{-2} h^{-1}	68 ±03 Bq m^{-2} h^{-1}	12

Table 2.2: Verification of the model in Finland and Hungary for regional mean values of measured and predicted ^{222}Rn flux.

2.6 Conclusion

Most of the spatial variation in ^{222}Rn flux may be explained by the variation in radionuclide activity in soils derived from different parent material. Soil moisture has been shown to have similar effects on ^{222}Rn flux as it has on GDR, except for short time periods during precipitation events. Considering additional parameters besides GDR, e.g. soil type, might further improve the prediction of ^{222}Rn fluxes on the small scale. However, it may also unnecessarily complicate prediction, especially if we are going to extend it to areas where required data may not be available. To predict average regional ^{222}Rn flux, the empirical correlation with GDR seems to suffice to produce regional means of ^{222}Rn flux within the error margin of measurements.

2.7 Acknowledgments

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Chapter 3

Mapping terrestrial γ-dose rate in Europe based on routine monitoring data

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3.1 Abstract

After the nuclear reactor accident in Chernobyl in 1986, most countries of the European Union (EU) established monitoring networks measuring outdoor γ-dose rates for early warning. The data is composite values γ-dose rate due to terrestrial, cosmic and artificial radiation sources, and in most cases also include some instrument background. While EURDEP is mainly designed for exchanging and stocking data during radiological emergencies, the data it is storing in its database may potentially contain valuable information about spatio-temporal variations of the ^{222}Rn source term which can be used for the validation of atmospheric transport models and other atmospheric tracer applications. The use of γ-dose rates as a proxy for outdoor ^{222}Rn concentrations is indeed possible if one can extract the terrestrial γ-dose rate contribution from the values reported in EURDEP. It is the purpose of this paper to discuss the preparation of the terrestrial γ-dose rates using EURDEP data and to present seasonal maps of terrestrial γ-dose rates in Europe. Such maps could be used for the preparation of ^{222}Rn source term which can be used for the validation of atmospheric transport models as well as for exploring variations in soil moisture content, an important parameter in flood prediction. These applications are the focus of the ongoing studies, but beyond the scope of this paper. In this paper, we show how the terrestrial γ-dose rate can be derived from the emergency monitoring data and two seasonal maps of γ-dose rates at the European scale are produced using geostatistics.

3.2 Introduction

A lot of effort and financial input is usually required to gather the necessary data for modelling any parameter of scientific interest on a large scale such as Europe. Using existing

networks providing data about a variable that is a proxy of a desired parameter can therefore be an interesting solution. For investigating spatio-temporal variations of the ^{222}Rn source term for the validation of atmospheric transport models and other atmospheric tracer applications - a variable that is measured only at few locations-, other sources of data were explored, among which total γ-dose rates that are monitored intensively in most European countries. In 1986, after the Chernobyl accident, most European countries extended their γ-dose rate monitoring networks or established new ones. The data they collect are transmitted regularly in the situation of routine to the EUropean Radiological Data Exchange Platform (EURDEP, http://eurdep.jrc.it). This activity initiated by the European Commission is designed to store and exchange data from the national networks in situations of routine monitoring and in emergencies (with higher temporal resolution). Because of the continuous monitoring, temporal variability in terrestrial γ-dose rates can be observed and potentially used for scientific purposes. Terrestrial γ-dose rates are indeed closely related to factors affecting ^{222}Rn fluxes (Schery *et al.*, 1989) and could enable the large-scale modelling of the ^{222}Rn source term (Szegvary *et al.*, 2007b). This term, in combination with observations of atmospheric ^{222}Rn concentrations, should allow the evaluation and calibration of climate models simulating transport, transformation and removal processes of gases and aerosols. Used in inverse mode, these models can further provide information on location, extent and strength of sources and sinks of greenhouse gases based on the measurement of changes in their atmospheric concentrations. Currently, the effective use of ^{222}Rn in this context is limited by the accuracy of the ^{222}Rn source term (WMO, 2004). Changes in the terrestrial γ-dose rates also indicate changes in soil moisture (Jones and Carroll, 1983), a central parameter in streamflow forecasting. Aubert *et al.* (2003) showed the potential of sequential assimilation of soil moisture and streamflow data in forecasting of flood events. The currently central limitation to the operational implementation is the availability of soil moisture data. This information might be derived at a sufficiently high temporal resolution and with little effort from the existing radiological emergency monitoring network. To gain more information about the terrestrial γ-dose rate to be used as a proxy for such applications, the objectives of this study were twofold: (I) to extract the natural terrestrial component from reported data on total γ-dose rate, considering all factors of different networks, (II) and interpolate the data for generating seasonal maps of the natural terrestrial dose rates at the European level.

3.3 The European Radiological Data Exchange Platform (EURDEP)

The EURDEP network is currently used by 30 European countries for the continuous exchange of data from their national radiological monitoring networks while several other European countries applied for participation and are in the process of interfacing to the network. During routine operation, monitoring data is made available by the participating organizations at least once a day. During an emergency, data has to be made available at least once every two hours. In practice, more and more organizations make data available on an hourly basis as a routine.
The status of June 2007 is that thirteen countries make their data available during routine once per hour, four countries each two hours and the remaining countries between three and 24 hours. As a result of this network of networks, measurements of more than 4000

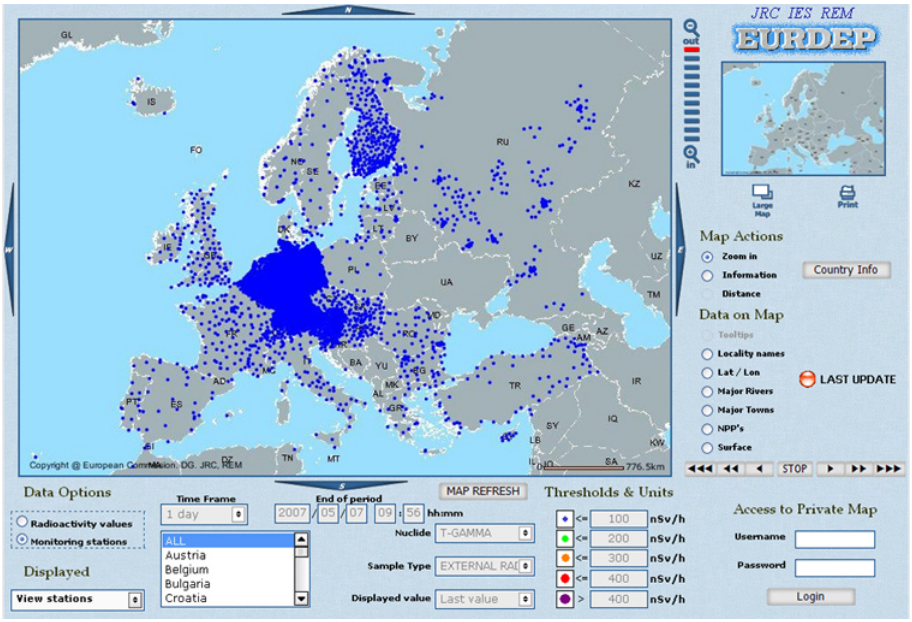

Figure 3.1: Monitoring stations within EURDEP (screenshot from http://eurdep.jrc.it, 07.05.2007)

monitoring stations (Table 3.1) are made accessible to the competent authorities in almost real-time and - with some restrictions and delays - to the public.

The total number of measurements listed in Table 3.1 is not always closely matching the theoretical number of measurements that is obtained by multiplying the number of stations of a country with the number of measurements per day for the 365 days in a year. A generic reason is that the indicated values for the number of stations and the measuring period are a snapshot that is only accurate for the end of 2006, that many stations did not have a regular and continuous operation throughout the entire year and that there were stations added or discontinued during the year. For some countries however the difference between the theoretical and the received amount of measurements is very significant and therefore deserves a more detailed explanation: Bulgaria and Russia have a measuring period of 10 minutes but they only sent a single 10 minute average per day. Slovenia sent a single 30 minutes average per hour for most stations. Switzerland has more than doubled the participating γ-dose stations since October 2006. Slovakia changed from one 24 hour average to sending 24 distinct measurements per day in September 2006. Finland, Hungary and Lithuania had long periods of time in which very few data were sent. Portugal sent mostly some six 20 minutes averages per day for many stations and from Norway there were very few measurements received during the first nine Months of the year. In order to further harmonize the exchange of monitoring data, the EURDEP Workshop of June 2006 therefore adopted an hourly measuring period and hourly data-transmission as the future standard. In addition the Joint Research Centre (JRC, a Directorate General of the European Commission) was asked to implement a watchdog system that will detect and report irregular data contributions. The central node of the EURDEP network is the Institute for the Environment and Sustainability of the JRC which is based in Ispra, Italy. The Federal Radiation Protection Agency (BfS) in Freiburg, Germany and the Directorate General for Transport and Energy of the European Commission (DG TREN) in Luxemburg act as mirror-sites for most of the data. Figure 3.1 shows a screenshot of

Country	Nr. of stations	Nr. of stations /2'500 km^2	Total nr. of measurements	Measuring period	Transmission interval during routine
Austria	334	9.96	166'414	24 h	24 h
Belgium	130	10.65	1'086'046	1 h	1 h
Bulgaria	27	0.61	9'228	10 min	24 h
Croatia	12	0.53	166'925	30 min	6 h
Czech Republic	54	1.71	427'079	1 h	1 h
Denmark	10	0.58	79'643	1 h	2 h
Estonia	10	0.55	83'106	1 h	3 h
Finland	256	1.90	1'414'803	1 h	1 h
France	168	0.77	9'821'873	10 min	12 h
Germany	2'068	14.48	780'992	24 h	24 h
Great Britain	91	0.93	751'413	1 h	1 h
Greece	24	0.45	183'171	1 h	24 h
Hungary	86	2.31	283'024	1 h	1 h
Iceland	1	0.02	33'713	15 min	2 h
Ireland	14	0.50	112'943	1 h	1 h
Italy	38	0.32	206'043	1 h	24 h
Latvia	15	0.58	110'901	1 h	2 h
Lithuania	11	0.42	45'104	1 h	1 h
Luxembourg	17	16.43	131'420	1 h	1 h
Netherlands	165	9.93	7'073'751	10 min	1 h
Norway	43	0.33	111'743	10 min	1 h
Poland	13	0.10	106'830	1 h	2 h
Portugal	13	0.35	80'055	20 min	24 h
Romania	37	0.39	13'120	24 h	24 h
Russia	294	0.04	41'788	10 min	24 h
Slovakia	23	1.18	33'997	1 h	1 h
Slovenia	37	4.56	199'335	30 min	1 h
Spain	32	0.16	1'285'935	10 min	24 h
Sweden	35	0.19	862'605	15 min	24 h
Switzerland	55	3.33	153'266	1 h	1 h
Averages	137	2.82	861'876	3 h	9 h
Totals:	4'151		25'856'266		

Table 3.1: Overview of the γ-dose stations that contributed to EURDEP in 2006

the web interface of the EURDEP website. This graphical user interface allows the user to select a number of variables as well as a time period of interest for which measurements can be displayed on a map of Europe or downloaded. Table 3.2 shows the total number of received measurements per year since 2003. The national networks participating to EURDEP present a large diversity in their densities, topologies (e.g. clustered around critical facilities and cities) or distributed regularly over the whole territory. Sweden, for example, has only about 35 stations, whereas Finland operates more than 250 monitoring sites. These discrepancies can be explained by the variety in the priorities and objectives set by the European countries for what concerns the radiation protection. Typically, while countries without nuclear power plants will focus on the detection of possible releases from nearby countries, those who have nuclear plants will want to monitor their territory with a higher accuracy. To be able to distinguish increased dose rate measurements that could have an origin in artificial radiation caused by an accident in a nuclear power plant for example from the natural background, this background must be known. Different techniques are used to asses the background: Whereas, as examples among many, Sweden uses airborne γ-spectrometry to determine its natural radiation background (Åkerblom *et al.*, 2005), Finland uses mostly its monitoring system consisting of permanently installed and continuously operating -probes. This diversity in the monitoring strategies has a number of consequences, some of which will be discussed later in the context of how to produce maps of γ-dose rates for whole Europe, i.e. covering regions with different monitoring procedures. In addition to the network structure, the variable monitored varies between countries. Thus, the type of detectors in use (Geiger-Müller counter, proportional counters, scintillation detectors, ionisation chambers) and aspects of data handling affect the readings. Both are information which is difficult to get, because some of the data is not public. To explore the variety of sensors (among other aims), in April 2004 the Euro-

pean Commission launched a study project for "the Evaluation of existing standards of measurement of ambient dose-rate; and of sampling, sample preparation and measurement for estimating radioactivity levels in air", or AIRDOS. The AIRDOS final report (Bossew *et al.*, 2007) includes a compilation and evaluation of site criteria, sampling and measurement methods and data handling procedures for national ambient dose rate monitoring networks and nuclear site-specific environmental monitoring stations in the EU and other European countries (e.g. Switzerland, Norway). A questionnaire was sent to the participating countries, and the local authorities were invited to supply relevant information on their networks, including technical details, data handling and definition of alert conditions. As a complementary activity, an intercomparison of detector systems of seven European countries was carried out by Thompson *et al.* (2000) within the EURADOS Action Group on monitoring of External Exposure, which is part of the European Research Council's Fourth Framework Programme. Although comparing dose rate detectors leads to better understanding of the pathways, which can be different between countries, from the physical quantity which is intended to be measured, to a reported number, there is still a lack of a common data handling procedure as evident from the replies in the AIRDOS questionnaires. Within our study, we focus only on γ-dose rate detectors and measurements; air sampling networks which are also discussed in AIRDOS, are not considered.

Year	Nr. of stations	Total measurements
2003	3'747	1'552'979
2004	3'740	3'707'402
2005	3'936	15'861'222
2006	4'151	25'856'266

Table 3.2: Number of participating γ-dose stations and received measurements in the last years

3.4 Extracting natural terrestrial γ-dose rate of data from different national networks and detectors

3.4.1 Inherent Background

Inherent background, or self-effect, is the value which a dose rate detector reports in a zero-radiation field, or more technically, the mean response of the detector to the true variable being zero. The self-effect of a γ-dose probe can be determined in underground laboratories like the underground laboratory for dosimetry and spectrometry (UDO) at the Asse salt mine (Neumaier *et al.*, 2000), supported by the Physikalisch-Technische Bundesanstalt (Germany). At that time, this salt mine was located more than 900 m below ground at a depth, where the cosmic muon intensity is reduced by more than 5 orders of magnitude compared to surface, and where natural radioactivity of rock as well as ^{222}Rn concentrations are very low. For these reasons, the ambient dose equivalent rate at UDO was reported to be less than 1 nSv h^{-1} (we always refer to ambient dose equivalent, H*(10)), a value which is low enough to estimate the self-effect of -probes. A comparison of over 20 detectors (Sáez-Vergara *et al.*, 2003) showed that the inherent background differs between type of detector as well as between models of the same type, but made by different producers. In addition, slight differences of the zero effect between instruments of the same

model (i.e., same type, same producer), or within a detector "population", have been observed, which may be considered as a residual random variability. Ionisation chambers present typically background levels as low as 0.2 - 2.0 nSv h^{-1}, while proportional and Geiger-Müller counters present higher levels and larger ranges of variations, respectively up to 10 nSv h^{-1} for proportional, and 7 to 41 nSv h^{-1} for Geiger-Müller counters. Considering that environmental background radiation levels reported in Europe typically range from 30 nSv h^{-1} to 200 nSv h^{-1}, the contribution of the self-effect to the reported measurements can be very important. Differences in the reported handling of the self-effect may lead to inconsistencies of reported dose rate values across national borders. Therefore, as a first step, the self-effect must be subtracted from the measured value. However, detectors of the same population show a certain variability of the self-effect between individual probes of 5-10%. This is a rough estimate, since, unfortunately, little is known about the variability of the self-effect in a population of detectors of same type and producer. In order to correct the inherent background for monitoring networks (i.e many detectors, depending on network), a mean value is used for instruments of same type and producer, but the residual random variability of the self-effect adds to the uncertainty of the difference gross value minus self-effect. At this point, also the data handling procedures become important because when using EURDEP data it must be known how the self effect is handled by different national authorities. Unfortunately only a few countries subtract the inherent background from their measurements, some are "considering" this effect (according to the replies to the AIRDOS questionnaire), but most networks report gross data, i.e. including self-effect. As AIRDOS provides also information on detector type and producer, the inherent background can be estimated and subtracted.

3.4.2 Cosmic ray subtraction

Measurements cleared of the inherent background provide a measure of the combined contributions of natural terrestrial, artificial and cosmic γ-dose rates. The natural terrestrial part originates from the gamma radiating progenies of the ^{238}U and ^{232}Th decay series and from ^{40}K. The artificial component is caused mainly by ^{137}Cs deriving from nuclear weapons tests and fallout from the Chernobyl accident in 1986 (other gamma radiating radionuclides contained in fallout contribute very little, because they have largely decayed due to their short half life, or because their initial concentration was low). The cosmic radiation mostly consists of muons. Because the cosmic radiation is mainly a function of the air pressure (Wissmann *et al.*, 2007), and γ-dose rate field sites in Europe are obviously not situated at the same barometric altitude, the cosmic contribution to the total γ-doses is not the same for all the stations. The higher the altitude at which the γ-probe is located, the lower the air pressure and the stronger the cosmic radiation. There is also a local variation of the cosmic part, which is mainly induced through local fluctuations of the air pressure. This fluctuation may cause variations of ± 3 nSv h^{-1}. The dependence on altitude was examined in airplane experiments (Wissmann *et al.*, 2007) over Braunschweig and the Baltic Sea, where two detector types measured radiation at different altitudes. Another method was used by Murith and Gurtner (1994), who measured the terrestrial radionuclides with in situ gamma ray spectrometry.
The cosmic part of the total γ-dose rate resulted from the difference between calculated terrestrial γ-dose rate from the gamma ray spectra and measured total γ-dose rate at field sites at different altitudes in Switzerland. Results from both studies showed very similar

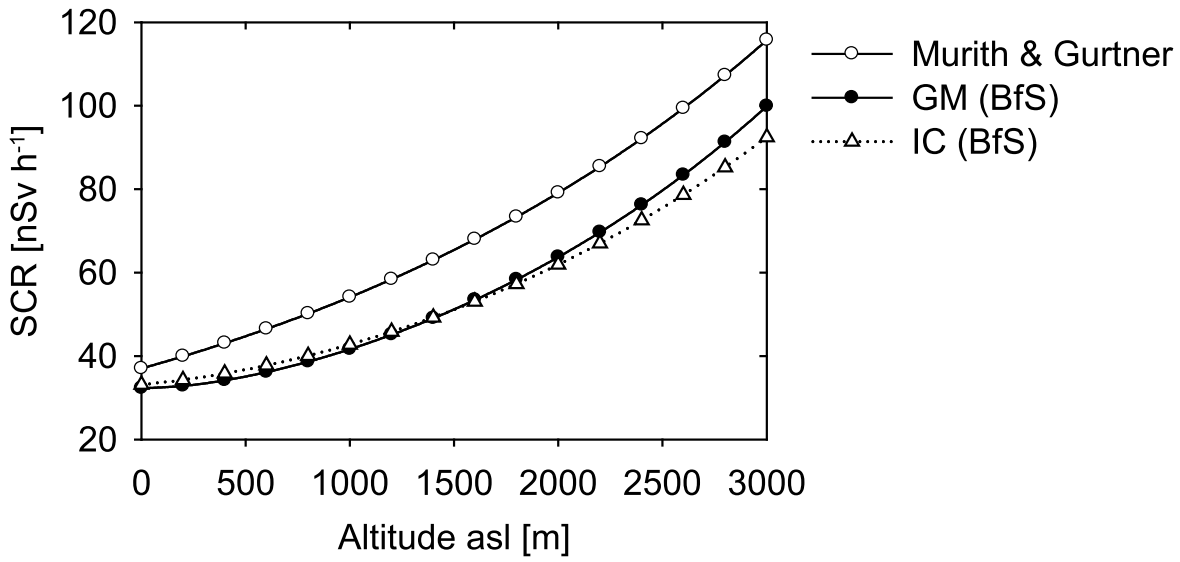

Figure 3.2: Dependence on secondary cosmic rays (SCR)

correlations of the cosmic radiation depending on altitude (Figure 3.2) and can be used to subtract the cosmic background at EURDEP field sites, when the altitude is known. We used both approaches to the dependence of altitude, but the model from Murith and Gurtner (1994) seemed to overestimate measured values at low altitudes. One reason might be that this study was carried out in Switzerland, where almost half of the field sites are situated between 500 m and 1000 m asl and 20% are between 1000 and 2000 m asl. Because 85% of the detectors were found to be located at altitudes below 500 m asl at the European scales, the approach of Wissmann *et al.* (2007) was preferred for the corrections. Here again, not all detectors will show identical responses to cosmic radiations as shown in Sáez-Vergara *et al.* (2007), and a correction factor is necessary. Plastic scintillation detectors seem to be insensitive to secondary cosmic radiations, other detectors have an over response, depending on the detector type. Since not all detector types have the same response, the calculated cosmic radiation with the formula of Wissmann *et al.* (2007) has to be corrected depending on the detector type. The latitudinal and longitudinal distribution of the cosmic ray flux was not taken into account as it was considered to be negligible. The effect of the geomagnetic latitude effect, i.e. the incident flux density, is minimal near the geomagnetic equator and increases with latitude. It is reported to be about 10% lower at the geomagnetic equator than at high latitudes (Bouville and Lowder, 1988). For European latitudes (35°N to 70°N) this effect is considered to be less than ±5% at ground level.Also fluctuations of the cosmic ray intensity depending on solar wind and the magnetic field were considered negligible. AIRDOS provides data of the altitudes of most stations of the national γ-dose rate networks. Missing information could be added with altitudes picked from digital elevation models (Google Earth, where national elevation models are used). However, in high altitude and steeply sloping regions, imprecise coordinates of the measurement stations might lead to large errors in estimating altitude and thus cosmic radiation.

3.4.3 Artificial Radiation

Deriving from nuclear weapons tests and the nuclear power plant accident in Chernobyl, ^{137}Cs is present in almost all soils in Europe, where it is the main source of artificial -radiation. Radiocaesium is still present today because of its relatively long half-life of 30.2 years and its strong fixation in most soils. The residual level of ^{137}Cs due to the atmospheric testing of nuclear weapons prior to the Chernobyl accident was estimated to range between 1-4 kBq m^{-2} (de Cort *et al.*, 1998) and we therefore will here only consider further the deposition of ^{137}Cs over Europe after the Chernobyl accident which varied from a few kBq m^{-2} to several thousands of kBq m^{-2} (de Cort *et al.*, 1998). With a few spots showing levels of ^{137}Cs deposition exceeding 100 kBq m^{-2}, parts of Finland, Sweden, Norway and Austria were among the most contaminated regions in Western Europe. Thus, in order to estimate the purely natural terrestrial γ-dose rate, the contribution of the ^{137}Cs must also be subtracted, in particular in those regions that were most affected by Chernobyl. Due to the migration of ^{137}Cs into soil, the external γ-dose rate above surface is reduced as a result of the attenuation of γ-rays by the soil layer between the source and the soil surface. Therefore, the external dose rate decreases more quickly then could be expected from physical decay of ^{137}Cs only. However, vertical migration rates are low in most soils (typically about 0.1 - 0.2 cm y^{-1}, Bossew and G.Kirchner (2004); Timms *et al.* (2004)), and not too different, as the resulting γ-dose rate above ground is concerned. Therefore, we believe that using only one single value for the conversion of ^{137}Cs inventory (Bq m^{-2}) into external γ-dose rate, does not induce an intolerable uncertainty. Moreover, the contribution of anthropogenic radiation to the total dose rate can be considered as rather low, except in a few cases. For the conversion we thus used the assumption from Bossew *et al.* (2001), who found a dose rate conversion factor for ^{137}Cs of 1 nSv h^{-1} per kBq m^{-2} by regression analysis of measured dose rate against activity concentration values. This seems to be a rough, but reasonable estimate for "old" fallout (ca. 10 years or more after deposition). The value can be approximately confirmed by calculations based on a physical model of tracer transport in soil and considering attenuation by "usual" surface roughness. The estimation of the ^{137}Cs inventory at the sites of the dose rate monitors involves another source of uncertainty. Because only very few measurements of ^{137}Cs deposition levels have been made at the detector field sites, and because no digital (gridded) version of the Atlas of Caesium deposition in Europe after the Chernobyl accident (de Cort *et al.*, 1998) exists, only the isoline levels of the last reference could be used to derive some estimates of the ^{137}Cs levels at all EURDEP monitoring stations. Practically, monitoring stations falling between two isoline levels were receiving values equal to the mean value of the two levels, while those located close to an isoline were receiving the value indicated by the line. We recognize that this approach may generate considerable uncertainties regarding the estimated contribution of the ^{137}Cs. Still, for most of the detectors, contributing to EURDEP, the impact of this correction will not show a significant impact and the contribution of Chernobyl falls within the range of the natural fluctuations caused by ^{222}Rn in near surface air or changes in air pressure. For 83% of the values (between 0 and 2 nSv h^{-1}), no correction of the total γ-dose rate was deemed necessary. Only 17% of the values were found to fall between values ranging from 2 to 20 nSv h^{-1} and were therefore corrected to avoid a further bias in determining the natural terrestrial component of γ-dose rates. Only one field site was found to be located in a very highly contaminated region (between 40 and 50 nSv h^{-1}).

3.4.4 Dose rate aggregation interval

Dose rate values reported to EURDEP by different countries correspond to different temporal aggregation times, ranging from 10 minutes to 1 day. The aggregated time, i.e. the interval between the reference times of subsequent data transmitted to EURDEP, may be equal to the measuring time (also called counting, sampling, collect or integration time), but in most cases represents the period over which the results of individual measurements are averaged before being transmitted to EURDEP. The counting interval can be 1 min, e.g. for the TechniData® detector, but the output value may be a mean calculated over 1 h. The counting time can also be variable: some detectors count for a fixed time, others until a preset number of impulses has been collected. This has a number of consequences on the data used to map the reported γ-dose rates: while small sampling times, corresponding to high temporal resolution of the original data, will reveal increased dose rate due to rainfall events which lead to increased concentrations of gamma radiating Radon progenies near ground due to washout (Greenfield *et al.*, 2003b), daily means may "smooth out" such, in most case short-term episodes below statistical detectability. The data selected in this study are thus chosen using the least common denominator between all these measurements, which is the daily mean of the γ-dose rate. In order to exclude anomalies, a filter has been applied: a value is excluded as outlier if it is a factor 100 above the mean of the preceding two days and above a threshold of 400 nSv h^{-1}. Such anomalies occur, as experience shows, from time to time as result of spurious electronic responses, i.e. equipment malfunction, or as "real" readings due to detector calibration or the use of radioactive sources near a monitor, e.g. for material testing. If one intends to monitor soil moisture or other parameters related to γ-dose rates, including such anomalies would lead to increased uncertainty in estimating the temporal and spatial distribution of the variable of concern.

3.4.5 Site criteria

Unfortunately there are no common standards for detector siting and set-up among the networks which participate in EURDEP. For emergency purposes, when the interest is mainly the detection of values above a critical threshold, this is not an essential issue. However, for our purposes, the detector siting may have some influence on the reported values. In order to be able to map the terrestrial component of the dose rate for the purpose of estimating the ^{222}Rn source term or soil moisture, the monitor must be sited such as to be able to measure the quantity of concern, i.e. be located above natural soil; measurements must be comparable, i.e. the responses of different detectors to a given phenomenon not only must be known, but also a physical model of the response must be available, so that the responses can be recalculated into each other by changing the model parameters. As an example, the response of a dose rate detector to ^{222}Rn emanating from the ground depends on the height of the detector above ground. Also the response of a detector to a given ratio of ^{222}Rn/^{220}Rn source terms depends on the height of the detector above ground because of the different diffusion lengths of the two isotopes, in turn due to their different half lives. The detector response to these physical phenomena in dependence of detector height is not an easy problem - to our knowledge no parameterization is available for this. Therefore, a standardized height of the detectors above ground is wishful, like 1 m, which is increasingly accepted as standard within the EURDEP community. In Germany or the Czech Republic for example, most dose rate

detectors are installed in this standard height, whereas in other countries, detectors are located sometimes at very different heights above ground, sometimes on roofs or close to buildings. Such detector siting further influences the measurements because of radiation shielding by the buildings and possibly by natural radioactivity contained in building materials (which also emanate ^{222}Rn).

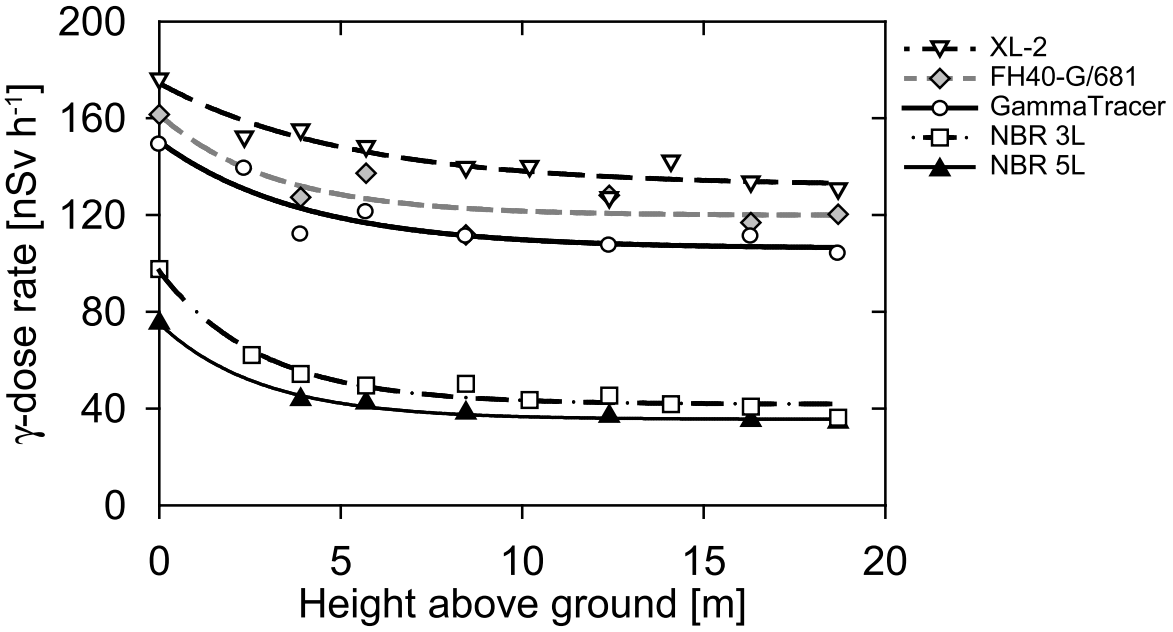

Figure 3.3: Measurements of the total γ-dose rate at different heights above ground on a look-out. The NBR devices are practically not sensitive to the secondary cosmic radiation.

As an example, Figure 3.3 shows the dependence on the altitude above ground of different γ-dose probe types: Geiger-Müller counter (GammaTracer, XL2; both instruments Genitron, Germany), two NBR detectors and another Geiger-Müller counter (FH40-G, all Thermo Fisher, Erlangen, Germany). The measurements took place at the Schauinsland look-out in the Black Forest (Germany). The tower has a height of about 18 m having 5 platforms. At every platform and additionally in-between the γ-dose rate was measured for 20 min. A second GammaTracer was placed at ground level as a reference to fluctuations due to meteorological conditions. Typically, all devices showed the highest decrease of the γ-dose rate within the first 5 m. From 5 to 18 m the γ-dose rate decreased relatively little. The results from the experiment were used to correct the measurements with detectors located more than 1 meter above ground. Of course, shielding on roof tops or by the building and natural radionuclides in the building material may have a greater influence on the measurement than expected. But considering that 78% of the field sites within EURDEP are reported to be 1 m above ground and 90% not higher than 5 m above ground, a correction was made for probes fixed at heights exceeding 1 meter above ground. Some uncertainty remains as for some countries information is vague, e.g. a common declaration of altitude above ground as 'between 2 and 317 m'. Where great heights were spotted (e.g. Towers, tall buildings), the detectors were excluded from further analyses.

3.5 Temporal variation

As our first objective is to generate a map of natural terrestrial γ-dose rate, we also explored the temporal fluctuations of our variable. As mentioned above, air pressure may cause fluctuations of ± 3 nSv h^{-1}. Still, there are other factors that might be even more relevant: Lebedyte *et al.* (2002) observed that the diurnal, periodical variation of ^{222}Rn concentration in outdoor air reflects in the γ-dose rate. A spatial and temporal correlation between ^{222}Rn flux and the terrestrial γ-dose rate has also been shown in Szegvary *et al.* (2007b). Other factors to be considered are the snow-cover, which is typically more important in the northern parts of Europe. Water present in soil absorbs γ-photons and, therefore, shields the detector against terrestrial radiation. This shielding effect can have a magnitude of ± 10 to 15%. The most important source of variability of the natural dose rate is probably washout of Rn progenies during the beginning of rainfall. Increases of the dose rate of 50 nSv h^{-1} and more due to this phenomenon have been observed. The relation between Rn washout and dose rate has been described in Greenfield *et al.* (2003b). Depending on the frequency and size of such "Rn episodes", our "cleaned" data of terrestrial γ-dose rate also contain a corresponding contribution.

3.6 Mapping the γ-dose rates

3.6.1 Data preparation

The last step of the data preparation before starting the mapping process is the harmonisation of the data. In Figure 3.4 the contribution of the above described partial dose rates are shown, the values represent the average values for EURDEP in 2006 with their standard deviations. We have, as far as possible, reduced the measurements of the total γ-dose rate to their natural, terrestrial component according to equation 3.1, based on the information available in AIRDOS:

$$\gamma_T = \gamma_0 - [\gamma_c + \gamma_a + \Delta_{SE}] \tag{3.1}$$

Thus the natural, terrestrial γ-dose rate γ_T is derived from the total, measured value γ_0 by subtracting γ_c, which represents the cosmic, and γ_a, the artificial parts, and the inherent background Δ_{SE}. γ_c needs a further correction for each detector type:

$$\gamma_c = \gamma_{c_0} \cdot \Delta_D \tag{3.2}$$

γ_{c_0} is the calculated cosmic ray dose rate and Δ_D the correction factor for the detector type, considering the sensitivity of the detector to cosmic radiation. Equation 3.1 therefore becomes

$$\gamma_T = \gamma_0 - [\gamma_{c_0} \cdot \Delta_D + \gamma_a + \Delta_{SE}] \tag{3.3}$$

From the EURDEP database, around $2.6 \cdot 10^7$ measurements made in 2006 were extracted. Weekly, monthly and seasonal (summer / winter) average values were calculated for every detector which provided data during that time; in this paper we discuss only the seasonal means. The number of stations providing data varied between 3326 and 3554. Because the platform is still growing and improving, the number of stations increased during the year though there was also a temporal loss of data due to modernisation processes of national networks. In addition to maps of the mean γ-dose rates in summer and winter,

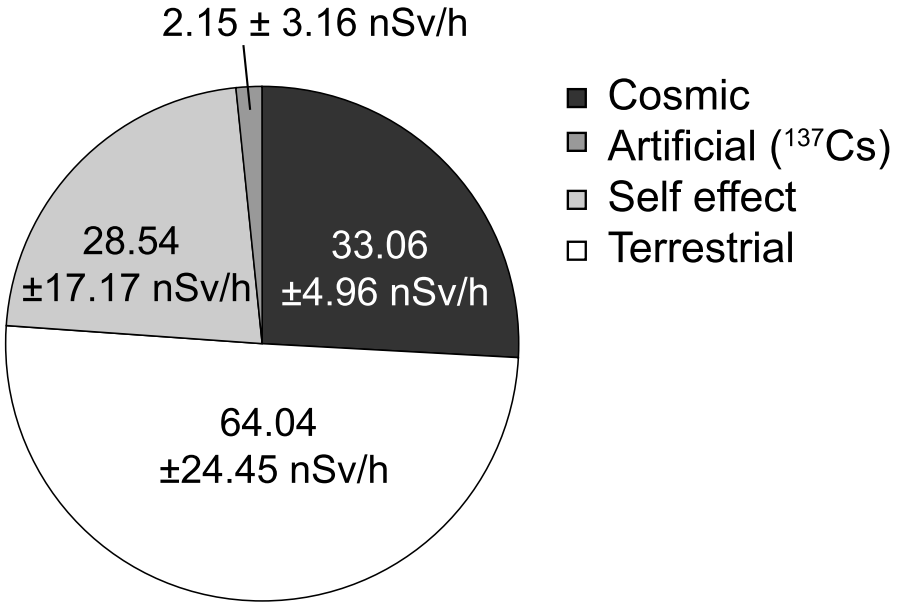

Figure 3.4: Contribution of different dose rates to the total measured dose rate of a gamma probe, average values and standard deviation for all stations within EURDEP for 2006.

we generate one of the difference summer - winter. For interpretation of the resulting map, consider the physical meaning of the difference: Let $S(x)$ and $W(x)$ denote the mean total dose rates (only cosmic part and zero effect subtracted) in summer and winter at location x. If we further denote with $n(x)$ and $a(x) = \gamma_a$ the natural and artificial parts, the "cleaned" dose rates according to the procedure described above are,

$$\gamma_T(Summer) = S(x) - a(x) = n(x) \tag{3.4}$$

$$\gamma_T(Winter) = W(x) - a(x) \tag{3.5}$$

The difference therefore is

$$D(x) = (S(x) - a(x)) - (W(x) - a(x)) = S(x) - W(x) \tag{3.6}$$

Now, define

$$\alpha(x) = \frac{W(x)}{S(x)} \tag{3.7}$$

a factor which describes the reduction or increase of the dose rate in winter, as compared to summer. Then, $D(x)$ will become

$$D(x) = S(x) \cdot (1 - \alpha(x)) = S(x) \cdot \beta(x) = [n(x) + a(x)] \cdot \beta(x) \tag{3.8}$$

Thus the spatial variability of the difference $D(x)$ reflects the spatial variability of the "source term", $n(x) + a(x)$, and the spatial variability of environmental factors which affect the dose rate, $\beta(x)$. Note that in $\gamma_T(Winter)$, the "full" value γ_a is subtracted, and not a modified one, $\alpha \cdot \gamma_a$, as would be more accurate. Therefore,

$$\gamma_T(Winter) = W - \gamma_a \tag{3.9}$$

is only approximately the natural terrestrial component in winter. Finally we attempt to establish a rough uncertainty budget of γ_T, as defined in Eq. 3.3. The uncertainty of the estimated γ_{c_0} is probably far below 5%, the one of Δ_D is unknown; we adopt an assumption of $u(\gamma_c) = 10\%$ (1 standard deviation), which is probably over-conservative. The estimation of γ_a from the ^{137}Cs inventory values from the Cs atlas and the use of a rough dose rate conversion factor, involves considerable uncertainty: $u(\gamma_a) = 50\%$ is probably realistic. For the self effect, we set $u(\Delta_{SE}) = 10\%$, probably slightly conservative. The uncertainty of γ_T from these sources, i.e. excluding the one from γ, that is, the uncertainty introduced by the correction procedure, is therefore

$$u(\gamma_T)^2 = (\gamma_{c_0} \cdot \Delta_D \cdot 0.1)^2 + (\gamma_a \cdot 0.5)^2 + (\Delta_{SE} \cdot 0.1)^2 \tag{3.10}$$

If we take the mean contributions of the components according to Figure 3.4, $(\gamma_{c_0} \cdot \Delta_D)/\gamma_T = 0.52$, $\gamma_a/\gamma_T = 0.03$, $\Delta_{SE}/\gamma_T = 0.45$ we find $u(\gamma_T)/\gamma_T = 0.07$ in average. Of course, $u(\gamma_T)$ can be higher in areas where γ_a, with its relatively high uncertainty, contributes more. For the total uncertainty of γ_T, the one of γ_0 must be added, which we roughly estimate 10%. With $\gamma_a/\ \gamma_T = 2$ (Figure 3.4), this yields a total $u(\gamma_T) \approx 21\%$. In the difference D, the correction terms cancel out, thus its uncertainty is only due to the ones of $\gamma_0(Summer)$ and $\gamma_0(Winter)$, which depend on their temporal fluctuations. Using the same rough 10%, we find an uncertainty of D in the order of magnitude $10\% \cdot \sqrt{2} \approx 15\%$.

3.6.2 Geostatistical analysis

Ordinary Kriging (OK) is the core estimator used in geostatistics for estimating values at unsampled locations and, as such, is probably nowadays one of the most frequently used functions for mapping environmental data. Geostatistics is based on Matheron's regionalized variables theory which is considering natural processes as a combination of a smooth global structure with random local variations. In contrast to deterministic interpolators, geostatistics takes explicitly into account the spatial correlation of the investigated phenomenon when calculating the weights attributed to neighbouring points during the interpolation process. We refer the readers to the standard literature in the field; see e.g. Chiles and Delfiner (1999) for more information about the method. For this work seasonal data for winter and summer 2006 in Europe were prepared after the necessary declustering process which was needed to reduce the influence of measurements made within densely designed networks. A moving window approach was thus used to randomly select a single measurement within each cell of size 50 x 50 km. The analysis of the spatial correlation of the variable (the computation of the experimental semivariogram) also showed the presence of a clear linear trend in the data which had to be removed since the range, i.e. the distance at which the semivariance becomes stable (and the measurements spatially independent), could be overestimated. It is believed that this trend is caused by the distribution of the geological units in Europe (Asch, 2005). As a result of this process, new semivariograms for the terrestrial γ-dose rate in winter and summer of 2006 were calculated (Figure 3.5).

The semivariograms shown in Figure 3.5 show clearly very similar patterns for both seasons: the spatial correlation is following some combined exponential and spherical models and the spatial correlation is getting insignificant at about 1200 km. Both also show some change of structure at around 500-600 km as indicated by an apparent nested structure.

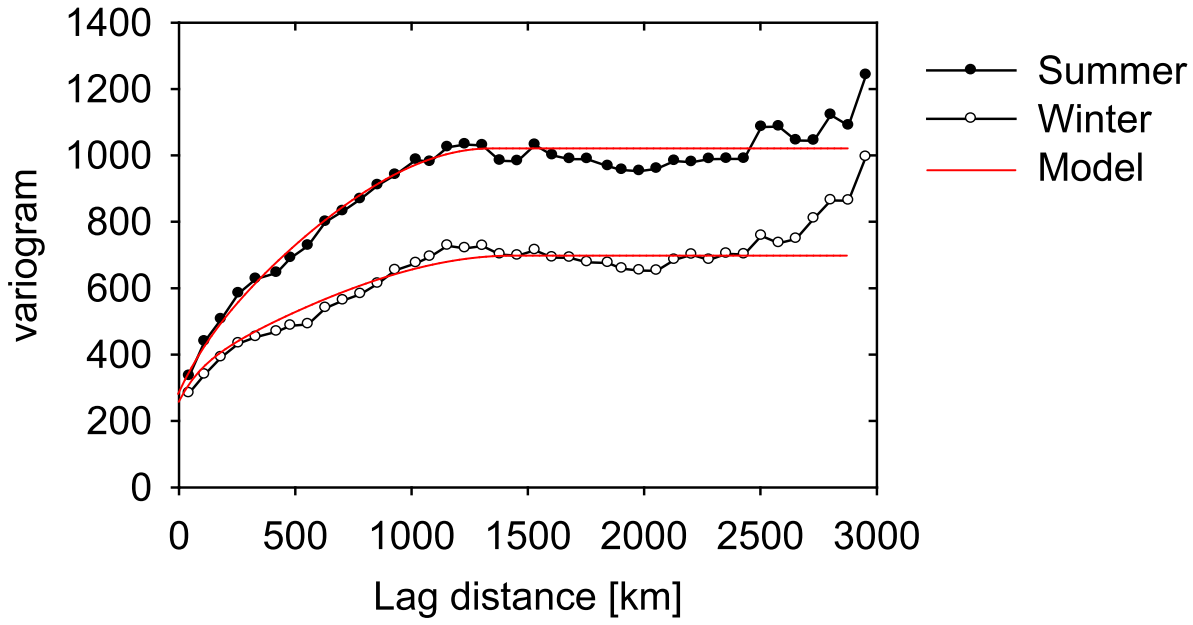

Figure 3.5: Omnidirectional variograms for seasonal average of terrestrial γ-dose rate for winter and summer 2006.

The dataset from summer 2006 shows, however, a much higher variance (almost two times larger) than the one of winter 2006. The latter may probably be explained with

- a "proportional effect": fluctuations are, as often the case for environmental data, proportional to their value, i.e. not the standard deviation itself, but the coefficient of variability (=standard deviation/mean) is approximately constant, which results in var(summer)/var(winter) = [mean(summer)/mean(winter)]^2
- different meteorological conditions between summer and winter which show larger fluctuations within the summer measurements.

A more in-depth analysis of the data using a higher temporal resolution is currently in progress to further explore these seasonal variations. Using the semivariograms shown in Figure 3.5, two seasonal maps of the average terrestrial γ-dose rates in winter and summer 2006 could be produced at the European scale as shown in Figure Figure 3.6. Uncertainties in these maps are proportional to the network density and are not further discussed. For what concerns the presence of extreme values, these are deriving from natural, terrestrial sources and can for instance be found in volcanic regions like north of Rome, Italy, where ^{222}Rn fluxes reach a high value and may increase the γ-dose rate significantly (Sciocchetti, 2002). For potential higher temporal resolutions (daily, weekly) other possible outliers may be caused from the washout of radon progeny by rain or calibration of the probes (with a radioactive source).
As we are interested in the terrestrial part, these "hot spots" must also be taken into account. On the other hand, if such an "anomalous" yet real phenomenon is monitored by an isolated detector of a network of low density (i.e. low spatial resolution), the interpolation process will most likely exaggerate the actual size of the phenomenon because of the low smoothing effect of our spatial interpolator. The presence of a strong spatial correlation at short scales allows us to have indeed a relatively low "nugget effect", which

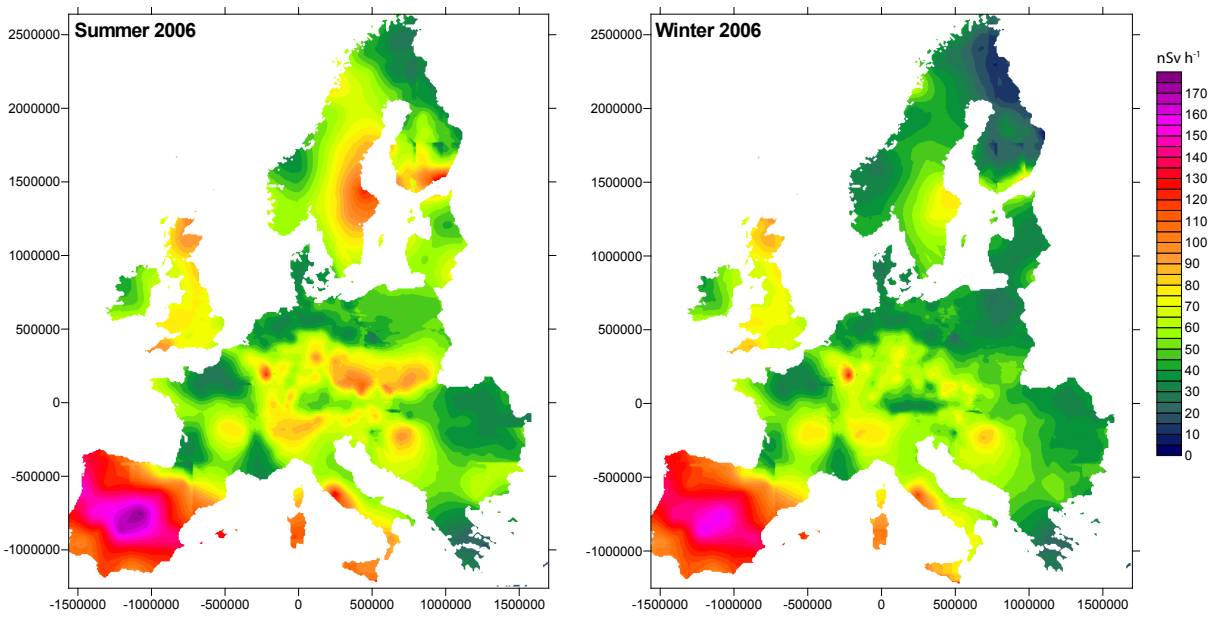

Figure 3.6: Average terrestrial γ-dose rates for summer (left) and winter (right) 2006 for Europe. Coordinates are in GISCO Lambert (units are in m).

is an expression of the low local variability of the measurements. More precisely, this nugget effect is defined as the semivariance value for separation distances tending towards 0, expressing thus errors within the measurements and the results of microscale variations. As a result of the low nugget effect, the interpolator is close to be exact at the locations of the measurement. The nugget effect is ca. 250-300 (nSv h^{-1})^2, as can be seen in Figure 3.5. Recalling $u(\gamma_T) \approx 21\%$, and if we assume $\gamma_T = 64$ nSv h^{-1} on average (Figure 3.4), a mean $u(\gamma_T) \approx 13.4$ nSv h^{-1} is found, or $var(\gamma_T) \approx 180$ (nSv h^{-1})^2. Whereas the sill variance is produced by the spatial variability of γ_T, its uncertainty adds to the nugget effect. The remaining ca. 100 (nSv h^{-1})^2 of the nugget variance must therefore be attributed to the micro-scale variability, that is, the spatial variability of γ_T below 60 km (the resolution of the variogram) equals ca. $\sqrt{100} = 10$ nSv h^{-1}. - To our knowledge, no literature data exist for comparison.

3.7 Discussion of the results

The terrestrial γ-dose rates for Europe in winter and summer 2006 in Figure 3.6 show levels ranging between 0 and 180 nSv h^{-1}. Particularly high values can be found in Spain, Southern Finland, in the Alps as well as in the Massif Central in France. Lower levels are in coastal regions, Scandinavia, Greece and Eastern Europe. The shown patterns largely correspond to the locations of magmatic rocks as shown in (Asch, 2005). Soils also have an effect that needs to be investigated, as it is known to affect outdoor Rn concentrations. As shown in Figure 3.7, substantial changes in terrestrial γ-dose rates across seasons (i.e. winter - summer) are mostly seen in the northern regions and central parts of Europe as well as in the Alps.
The explanation can be found in Eq. 3.4. Regions with high natural dose rate (granitic areas in Scandinavia, Bohemian mass or Spain, for the latter see Quindos Poncela *et al.* (2004)) or high Chernobyl fallout (parts of Scandinavia, Austria) show higher D, a con-

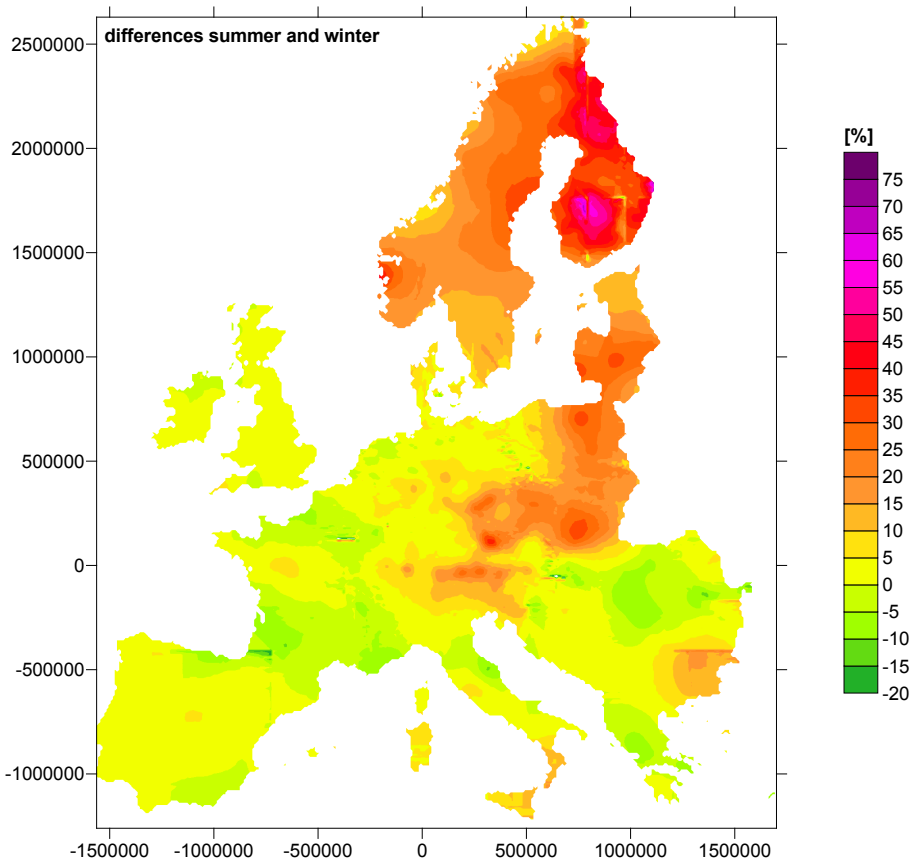

Figure 3.7: Differences resp. seasonal variation in terrestrial γ-dose rate between summer and winter 2006.

sequence of the "source term" factor in Eq. 3.4; Regions with, in average, longer lasting and thicker snow cover, or higher β, equally lead to higher D. Such conditions can be expected also in Scandinavia, in the Alps and in Central-Eastern Europe. An additional factor, also adding to β, may be higher soil moisture during winter time (Hatakka *et al.*, 1998; Baciu, 2006), since higher water content in soil leads to stronger shielding of -ray photons and also to reduced ^{222}Rn flux.

3.8 Concluding remarks

Access to a continuous measurement network for total γ-dose rate, covering a large area like the European continent, is a great opportunity for mapping and monitoring other parameters related to γ-dose rate. Some corrections of the measurements allowed us to go beyond the original objectives of the EURDEP network and provided us with useful information on spatial and temporal changes in the natural terrestrial part of γ-dose rate, which correlates with ^{222}Rn flux and soil moisture. Giving an estimation of goodness is difficult, as some uncertainties still remain after this study: For example our knowledge of the variety of the detector self-effects between the national networks is partly vague, so we had to make a number of assumptions regarding the detector types and producers. Over-response due to different calibration materials could not be considered. Especially Geiger-Müller probes can show an over-response in natural radiation environments of up

to 25% when calibrated with ^{137}Cs. However, almost 80% of the devices contributing to the EURDEP network are Geiger-Müller probes. Therefore, a comparable over-response can be expected for most parts of the network. For what concerns the secondary cosmic radiation we did not know the exact response behaviours of different -probes and therefore had to approximate the correction. Other uncertainties are related to the information provided by the national authorities who supplied information for the preparation of the AIRDOS report. In particular, information about the siting conditions of the detectors was not always available. Main gaps were found to be the specified height above ground of the detector, information about the accuracy of the coordinates (wrong coordinates were found) and information on the manufacturer of probes as well as the data handling. There is thus still space for improvements. Nevertheless, we gained a lot of information about the distribution of dose rate levels in Europe. The spatial patterns of the terrestrial γ-dose rate could be shown as well as their variations over time. Since different time intervals are used within different networks, the temporal resolution is limited to this factor. The highest temporal resolution is about one day, though some networks would allow a much higher resolution. But this is more than enough for using the data as a proxy for ^{222}Rn flux where a monthly resolution or even a seasonal average might suffice. As an outlook to future work on the subject, we think on (1) investigating the variability of γ_T and $\gamma_T + \gamma_a$ with higher temporal resolution (weekly, monthly), (2) studying the contribution of γ_a based on more accurate estimates of the ^{137}Cs inventory (at least regionally, where data are available) and the dose rate conversion factor, and (3) investigating if, or to what extent, different seasonal modification factors α should be used for natural radionuclides and ^{137}Cs.

3.9 Acknowledgements

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Chapter 4

European ^{222}Rn inventory for applied atmospheric studies

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Szegvary, T., Conen, F., Villani, M.-G., Bergamaschi, P., Schmidt, M., Kazan, V.: European ^{222}Rn inventory for applied atmospheric studies

4.1 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. Generally, a constant and homogenous ^{222}Rn source term of 1 atom cm^{-2} s^{-1} is assumed, sometimes reduced in northern latitudes. The imprecise nature of such assumptions is well known but a robust determination of the ^{222}Rn inventory 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 plus Norway, former Yugoslavia and Switzerland). It is based on the correlation between ^{222}Rn flux and the terrestrial γ-dose rate. Total γ-dose rate is monitored in this area by nearly 3'600 stations continuously and the terrestrial component can be extracted from these measurements. On the resulting 0.5° x 0.5° map, mean annual values of the ^{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}. Generally, 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. Using the atmospheric chemistry and transport model TM5, we present an example on how our spatially resolved ^{222}Rn inventory can improve simulations of atmospheric ^{222}Rn concentrations.

4.2 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; Bergamaschi *et al.*, 2005; Bousquet *et al.*, 2006; Hirsch *et al.*, 2006). Also, the accumulation of trace gases in the boundary layer has been extensively utilised to estimate the emissions of such gases by mass balance approach using ^{222}Rn as a tracer (Dörr *et al.*, 1983; Gaudry *et al.*, 1992; Moriizumi *et al.*, 1996; Schmidt *et al.*, 1996, 2001, 2003; Kuhlmann *et al.*, 1998; 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 accuracy of the ^{222}Rn inventory. Mostly, tuning and validation of atmospheric chemistry and transport models has 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. Errors in model validations and in flux estimates based on the ^{222}Rn mass balance approach are largely proportional to errors in the ^{222}Rn inventory. 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 than currently available is desirable (WMO, 2004). Direct measurements 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 3'600 stations in national emergency monitoring networks in Europe. These data are available in quasi real-time at the Joint Research Centre of the European Commission in Ispra, Italy. 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 modeling of the terrestrial 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). For this area we calculated weekly averages of ^{222}Rn flux for the year 2006. The primary objective of this paper is to present and describe this ^{222}Rn inventory. A secondary objective is to illustrate the impact of the new ^{222}Rn inventory on model simulations of atmospheric ^{222}Rn concentrations.

4.3 Constraining the European ^{222}Rn inventory

4.3.1 Approach

As described in Szegvary *et al.* (2007b), terrestrial γ-dose rate is a good proxy for ^{222}Rn flux. The terrestrial γ-dose rate is mainly determined by contributions from the decay of ^{40}K, ^{238}U and ^{232}Th including their daughter products plus, in some areas, ^{137}Cs derived from the nuclear power plant accident in Chernobyl 1986. The proportional contribution of ^{238}U and its daughter products to the terrestrial γ-dose rate is more or less constant

in Europe. It is on average 29% in North-West Italy (Chiozzi *et al.*, 2002), 27% in Spain in Quindos Poncela *et al.* (2004) and 30% in Cyprus (Tzortzis *et al.*, 2003). Continuous measurements of the total γ-dose rates in Europe have been intensified following the Chernobyl powerplant accident, when most European countries started to develop measurement networks to observe the environmental radioactivity. Such networks are used as emergency monitoring tools for early warning in case of a radioactivity incident. The data of 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 measurements are the total γ-dose rate, which consists of components from different sources. These include the terrestrial component (mainly from ^{40}K, ^{238}U and ^{232}Th and their progeny), the cosmic component (mainly muons), the anthropogenic component (mainly from ^{137}Cs) and the inherent background of the measurement devices. To derive the terrestrial γ-dose rate, all other components have to be subtracted. This was done for all data from 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 a 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 and Finland, described in Szegvary *et al.* (2007b). The regression equation reads:

$$^{222}Rn\ flux\ [atom\ cm^{-2}\ s^{-1}] = 11.75(\pm 1.27) \cdot \gamma - dose\ rate\ [\mu Sv\ h^{-1}] - 0.15(\pm 0.11) \quad (4.1)$$

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, 2004), because during field measurements in Finland only these soil types were found to be significantly overestimated by Eq. 4.1. Blank rocks were assigned a zero ^{222}Rn flux because the emanating surface is very small, leading to negligible exhalation rates. Continuous measurements by our group on a sandy loam, a typical soil texture in large parts of Europe, revealed a similar change in ^{222}Rn flux with changes in γ-dose rate as a result of variations in soil moisture (i.e. a reduction in ^{222}Rn flux of 0.013 atom cm^{-2} s^{-1} with a reduction of 1 nSv h^{-1}). The precision of the estimated ^{222}Rn flux maps depends largely on that of the terrestrial γ-dose rate, and therefore also on the density of the γ-dose rate monitoring stations in Europe. While a high density of stations (e.g. mean distance of 15 km in Germany) is important from a radiation protection point of view, a lower resolution would suffice for most atmospheric tracer applications. In the following, we discuss ^{222}Rn flux maps with a resolution of 0.5° x 0.5°.

4.3.2 Inventory description

First, we will describe the mean annual ^{222}Rn flux, focusing on the spatial heterogeneity. In a second step, we will look at temporal variations with a weekly resolution. Thirdly, 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 is shown in Figure 4.1. The picture demonstrates the considerable heterogeneity, significantly different from the commonly assumed uniformity. A major difference to previous assumptions are on average smaller flux values. Our map shows a mean ^{222}Rn flux of 0.62 atom cm^{-2} s^{-1} south of 60°N, where commonly 1 atom cm^{-2} s^{-1} is assumed, and 0.42 atom cm^{-2} s^{-1} further

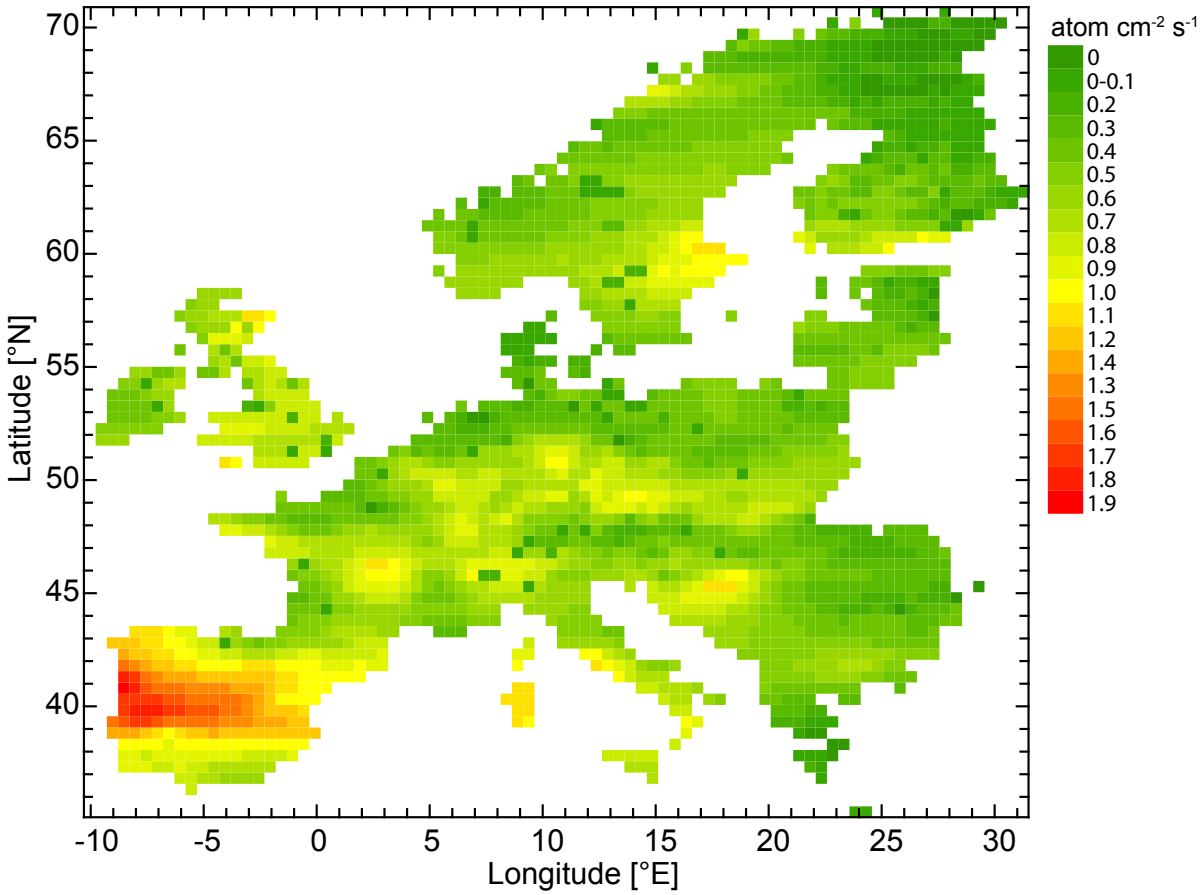

Figure 4.1: Estimated mean annual ^{222}Rn flux in Europe for the year 2006 (data available at http://www.radon.unibas.ch)

north, where 0.5 atom cm^{-2} s^{-1} are supposed (Rasch *et al.*, 2000, e.g.). Small fluxes in the northern parts of Europe may be caused by high soil moisture contents, shielding parts of the terrestrial γ-dose as well as constricting the ^{222}Rn exhalation at the soil surface. A further cause is certainly the abundance of organic soils with small mineral components and therefore low γ-dose and ^{222}Rn flux rates. Some coastal regions towards the Atlantic and the North Sea may show low levels of ^{222}Rn flux due to abundant rainfall and therefore also high soil moisture. More importantly, quarternary sediments, abundant in some of these areas, may tend to contain less radionuclides and therefore have a lower ^{222}Rn potential (Kemski *et al.*, 2001). Large ^{222}Rn fluxes can be found on the Iberian Peninsula, where dry soil conditions and the geological situation (Quindos Poncela *et al.*, 2004), i.e. crystalline rocks and soils developed from them, generate high γ-dose and ^{222}Rn exhalation rates. Even in the northern parts of Europe, in Sweden and Southern Finland values exceeding 1 atom cm^{-2} s^{-1} can be found. These large values are well correlated with the prevalence of crystalline rocks (e.g. granite) in those regions (Asch, 2005). The 'hot spot' region near Rome, Italy, may be explained by strong volcanic influence as described in Voltaggio *et al.* (2006). Figure 4.2 shows the frequency distribution of all ^{222}Rn flux values from the 0.5° x 0.5° map. Half of the values 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}). In Table 4.1, an overview of the mean ^{222}Rn

flux values is given for countries covered by the map. These values are extracted from the 0.5° x 0.5° raster map and represent the mean of all values within the borders of a country. The largest values were found for Portugal (1.35 atom cm^{-2} s^{-1}) and Spain (1.03 atom cm^{-2} s^{-1}), the smallest for Denmark (0.20 atom cm^{-2} s^{-1}), Greece (0.23 atom cm^{-2} s^{-1}) and The Netherlands (0.29 atom cm^{-2} s^{-1}). The standard deviation in Table 4.1 gives an estimate on the variation between 0.5° x 0.5° units in the corresponding countries, i.e. an estimation of the heterogeneity of ^{222}Rn fluxes. The largest relative standard deviation (standard deviation / flux average) was found in Finland (83%), almost twice as large as in the similar sized Germany (44%).

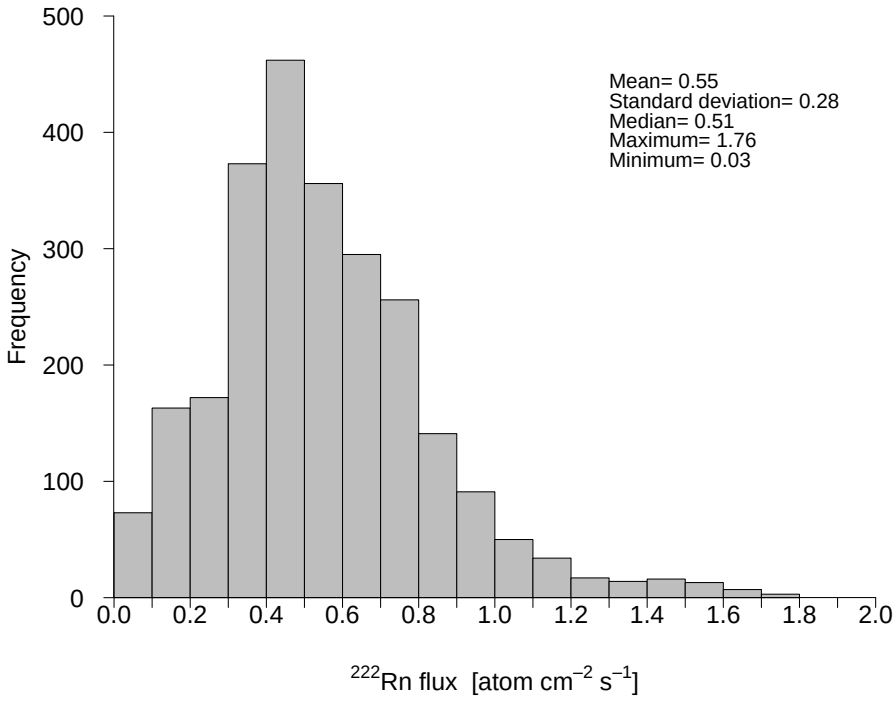

Figure 4.2: Distribution of estimated ^{222}Rn flux densities in Europe, extracted from ^{222}Rn flux map in Fig. 4.1

4.3.3 Temporal variation

Continuous γ-dose rates measurements allowed us to estimate the temporal variation in ^{222}Rn flux in Europe. In Figure 4.3, time series of estimated weekly averages in ^{222}Rn flux values are presented as composites bands of 5° in latitude (a) and longitude (b). The graphs indicate not only the 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, where snow and soil moisture are dominating in winter time and significantly drier conditions prevail in summer. 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. Based on measurements made over several years, Schüssler (cited in Levin *et al.* (1999)) estimated a seasonal variation of ±25% for the Heidelberg area (around 49.4°N, 8.7°E), which is somewhat larger than our

Country	^{222}Rn flux average atom cm^{-2} s^{-1}	spatial standard deviation atom cm^{-2} s^{-1}
Albania	0.45	0.10
Andorra	0.81	0.00
Austria	0.41	0.11
Belgium	0.65	0.14
Bosnia and Herzegovina	0.69	0.12
Bulgaria	0.49	0.11
Croatia	0.73	0.21
Czech Republic	0.72	0.18
Denmark	0.20	0.05
Estonia	0.31	0.11
Finland	0.30	0.25
France	0.60	0.18
Germany	0.52	0.23
Greece	0.23	0.15
Hungary	0.47	0.14
Irish Republic	0.42	0.21
Italy	0.70	0.19
Latvia	0.39	0.11
Liechtenstein	0.45	0.00
Lithuania	0.42	0.08
Luxembourg	0.91	0.07
Macedonia	0.53	0.04
Netherlands	0.29	0.10
Norway	0.38	0.20
Poland	0.45	0.12
Portugal	1.35	0.31
Romania	0.33	0.07
Serbia and Montenegro	0.53	0.11
Slovakia	0.67	0.11
Slovenia	0.63	0.07
Spain	1.03	0.26
Sweden	0.52	0.20
Switzerland	0.57	0.33
United Kingdom	0.68	0.21

Table 4.1: Estimated Average ^{222}Rn flux values for European countries within the area of the ^{222}Rn flux map (Fig. 4.1)

estimate of ±8% for a radius of 50 km around Heidelberg. This points to a difficulty of interannual variability of the seasonal amplitude. It may be larger when a particularly dry summer is followed by a snow-rich winter. In 2006, drier winter and wetter summer months than usual may have resulted in a smaller than usual seasonal amplitude in large parts of Europe. Therefore, we would consider our estimates of the seasonal variation as being at the lower end of what might be expected. The seasonality averaged in 5° longitudinal bands shows a generally decreasing ^{222}Rn flux to the East, which is partly explained by the fact that northern countries, exhibiting low fluxes being part of the East. A noteworthy seasonality appears only east of 5°E, where the climate also becomes more continental.

4.3.4 Comparison to previously used inventories

Conen and Robertson (2002) proposed a linear decrease in ^{222}Rn flux rate from 1 atom cm^{-2} s^{-1} at 30°N to 0.2 atom cm^{-2} s^{-1} at 70°N independent on longitude, based on re-

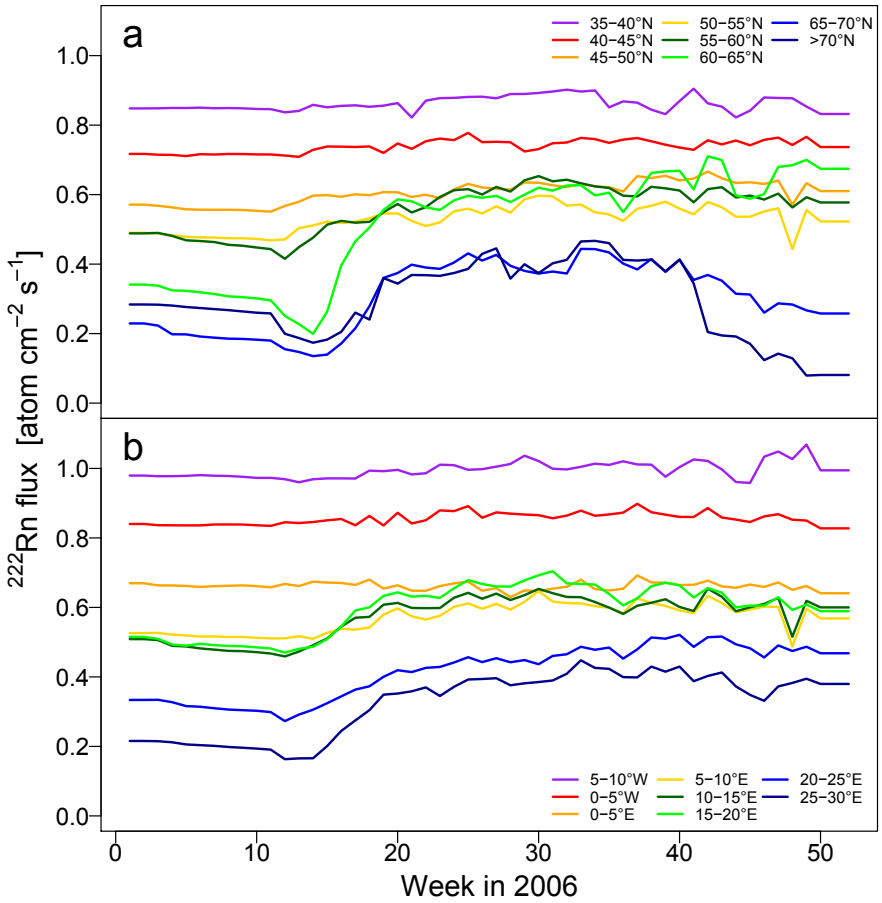

Figure 4.3: Seasonal variation of estimated average ^{222}Rn flux values for (a) 5° latitudinal bands and (b) 5° longitudinal bands

ported indirect measurements at different latitudes mainly in the USA and in the former USSR. Summary statistics of average annual ^{222}Rn flux in 5° latitudinal bands produced with our approach (Figure 4.1) reveal a very similar picture (Figure 4.4). This is surprising as both studies are based on completely different and independent approaches. The commonly used inventory (e.g. Rasch *et al.*, 2000) is close to our estimates only around 35° and 60°N, while generally over-predicting at other latitudes.

Jennings *et al.* (2006) directly measured ^{222}Rn flux rates with accumulation chambers in the Irish Republic during two campaigns, in October 2000 and in July 2001, and related them to soil texture classes as defined in the FAO soil map of the world (FAO, 1978). A total of 95 locations were assessed. They calculated an average ^{222}Rn flux for entire Ireland based on the relative abundance of soil texture classes in the order of 0.51 $\pm$ 0.1 atom cm^{-2} s^{-1}. This value is close to our annual prediction of 0.49 atom cm^{-2} s^{-1} for 2006. The value for Ireland is slightly larger than the one for the Republic of Ireland (0.42 atom cm^{-2} s^{-1}, Table 4.1) because of relatively larger emissions in Northern Ireland (Figure 4.1).

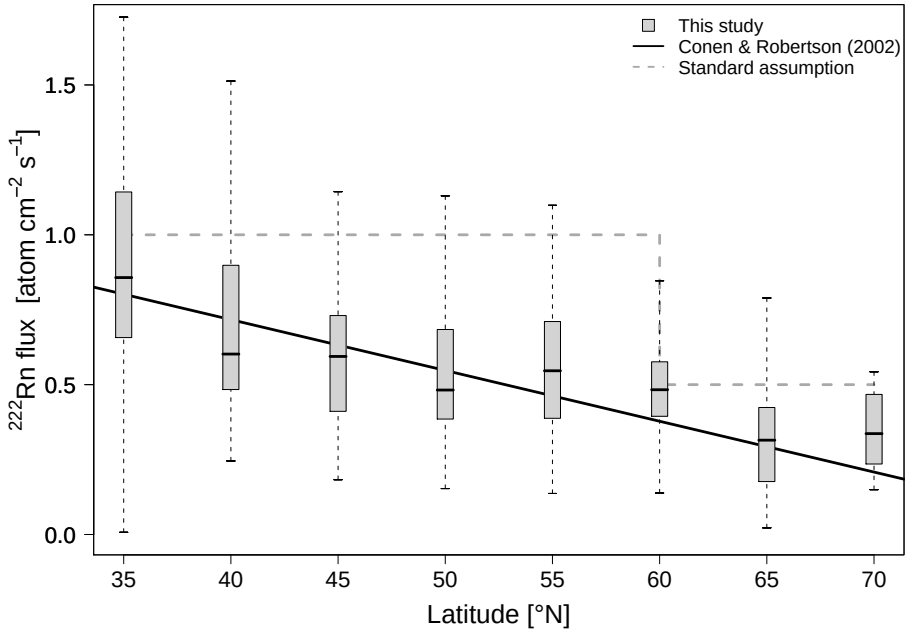

Figure 4.4: Flux rates of ^{222}Rn for 5° latitudinal bands from 35°N to 70°N, derived from the ^{222}Rn map in Fig. 4.1, compared to the linearly decreasing flux rate proposed by Conen and Robertson (2002)) and the standard assumption (Rasch *et al.*, 2000)

4.4 Specific effects on simulation of atmospheric ^{222}Rn concentrations

An important feature distinguishing our inventory from earlier inventories is its spatial heterogeneity. To illustrate the effect of this heterogeneity on the simulation of atmospheric ^{222}Rn concentrations, we provide an example, which is concerned with the prediction of the amplitude of ^{222}Rn accumulation in the atmospheric boundary layer depending on the origin of air masses. To this end, we compare simulations made with the atmospheric transport model TM5 (Krol *et al.*, 2005) based on a commonly used inventory (abbreviated: STD; 1 atom cm^{-2} s^{-1} south of 60°N and 0.5 atom cm^{-2} s^{-1} between 60°N and 70°N) and on the inventory described in this study with a weekly resolution (abbreviated: EU; Figure 4.1). In the simulation with the EU inventory we assumed outside the area presented in Figure 4.1 same emissions as in STD. The TM5 model has been widely used to simulate ^{222}Rn (Krol *et al.*, 2005; Bergamaschi *et al.*, 2006), CO (Gloudemans *et al.*, 2006), NO_2 (van Noije *et al.*, 2006), SF_6 (Peters *et al.*, 2004), CH_4 (Bergamaschi *et al.*, 2005, 2007) and SO_2 (de Meij *et al.*, 2006). It is a 3D atmospheric chemistry-transport zoom model, which allows the definition of arbitrary zoom regions (at horizontal resolution of 1° x 1°), which are 2-way nested into the global model (Berkvens *et al.*, 1999), run at a resolution of 6° x 4°.

Within the atmospheric boundary layer atmospheric trace gases are usually relatively well mixed. Hence mixing ratios largely depend on emissions and height of the boundary layer. Under stable conditions, the boundary layer typically exhibits are clear diurnal pattern, with the development of a convective boundary layer (1-2 km) during daytime and a much shallower nocturnal boundary layer during nighttime. A large increase in near surface ^{222}Rn concentrations can be observed during the formation and persistence

of a stable nocturnal boundary layer (NBL). The amplitude of the concentration increase depends on the stability and depth of the NBL and the source strength of the emitted gas along the path of the air masses arriving at the point of observation (Denmead *et al.*, 1996). Thus, air masses having passed over a strong source will result in larger amplitudes than those having passed over a weak source. An atmospheric ^{222}Rn measurement station influenced by contrasting source strengths is Gif-sur-Yvette. It is situated close to Paris (2.13°E, 48.70°N at 160 m asl), about 175 km from the coast of the Atlantic Ocean. The ^{222}Rn measurements were taken at the Laboratoire des sciences du climat et l'environnement (LSCE, France).

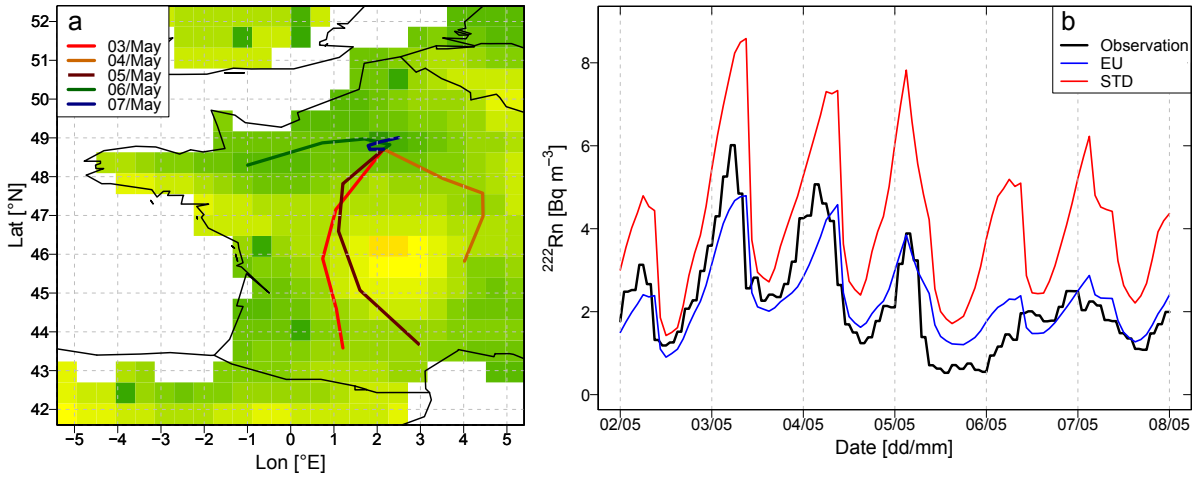

Figure 4.5: a: One-day back-trajectories of air masses having arrived at Gif-sur-Yvette around the end of the days (midnight) indicated in the legend. b: Observed and predicted ^{222}Rn concentrations (with TM5 atmospheric transport model) for the same period based on a standard ^{222}Rn inventory (STD) and the inventory presented in this study (EU)

Its location in the Paris Basin, and the Normandie to the West and North-West of it, are characterised by a ^{222}Rn source only about half that of the Massiv Central area to its South (Fig. 4.1). During relatively stable conditions at the beginning of May 2006, the origin of air masses arriving at Gif-sur-Yvette around midnight shifted from strong to weak ^{222}Rn source areas (Fig. 4.5a). Mean ^{222}Rn flux on 50 km wide corridors along one-day back-trajectories were 0.61 atom cm^{-2} s^{-1} for 3., 4. and 5. May and 0.30 atom cm^{-2} s^{-1} for 6. and 7. May. Correspondingly, the amplitude of the observed nocturnal increase in ^{222}Rn concentrations decreased from an average of 3.4 Bq (3., 4., 5. May) to about 1.1 Bq (6., 7. May) (Fig. 4.5b). The simulation based on the EU inventory followed this trend closely (average during 3., 4., 5. May: 2.9 Bq ; during 6., 7. May: 1.3 Bq). Assumption of a spatially uniform ^{222}Rn source (STD) generally over-predicted the amplitudes on average by 70% for the first three days and by 260% for the following two days. Accordingly, a mere reduction of the source strength in the STD scenario would not enable a match with observations which underlines the significance of the spatial heterogeneity represented in the EU inventory as a prerequisite for accurate prediction in such a case.

4.5 Conclusion

We have presented a new ^{222}Rn inventory for Europe, based on the correlation between ^{222}Rn flux and terrestrial γ-dose rate, together with information on total γ-dose rate from existing emergency monitoring networks in Europe. The derived ^{222}Rn emissions exhibit a significant spatial heterogeneity. The spatial coefficient of variation on a 0.5° x 0.5° grid was 51%. Temporal variability was small in southern and western parts and large in northern and eastern parts of Europe (because of larger soil moisture contents). The new ^{222}Rn inventory corresponds well to a previously proposed decline in source strength with increasing latitude and also with recent intensive measurements in Ireland. Comparison of predicted and observed ^{222}Rn concentrations in the surface layer showed a marked improvement when the new inventory was used compared to the previous assumption of homogeneous source strength, for a 1-week period at Gif-sur-Yvette with changing airmasses characterized by exposure to significantly different ^{222}Rn source strength. The three-dimensionality (N-S and E-W, as well as time dimension) of the inventory provides for a new quality which enables an improvement of simulations impossible with previous (one-dimensional, N-S) inventories.

4.6 Acknowledgements

This project was funded by the Swiss National Science Foundation (project no. 200021-105123). The authors greatly acknowledge data support by the British Atmospheric Data Centre for providing access to calculated trajectories using data from the European Centre for Medium Range Weather Forecasts.

Chapter 5

Overall conclusions

The commonly used ^{222}Rn source term of 1 atom cm^{-2} s^{-1} used for atmospheric tracer calculations has been shown to generally overestimate the measurements. Therefore, we aimed to investigate a better ^{222}Rn source term over Europe than the commonly used uniform source term.

As a first step, the hypothesis of a relationship between ^{222}Rn flux and γ-dose rate was tested and found to be suitable for this purpose. For this, measurements over longer time periods (a few weeks to a year) and spot measurements were established in Switzerland. Both parameters showed a good relationship, which could be used to map ^{222}Rn fluxes using γ-dose rate maps. Beside the spatial distribution, also the temporal variations of the parameters showed to be well correlated. Hence, we decided to produce not only an average ^{222}Rn flux map for 2006, but to consider a temporal variation as well. But as the contributions of the main radionuclides (^{238}U, ^{232}Th and ^{40}K) to the total γ-dose rate are not always equal, especially the variability of ^{40}K is large, predictions for smaller areas are difficult. Soil types and geological information might be used to improve the prediction of ^{222}Rn on the small scale, but as for the purpose of the ^{222}Rn flux map (i.e. use for atmospheric tracer models) larger areas are of interest, where the model was suitable, such additional information could unnecessarily complicate the model.

To scale up the model on a European scale, the development of a European terrestrial γ-dose rate map was the focus of a second step, which would be the base map for the new ^{222}Rn flux inventory. Continuous measurements of the γ-dose rate in Europe, taken from the EURDEP database, allowed to produce temporal γ-dose rate maps for 2006 for Europe. The methods to extract the terrestrial component from the total measured γ-dose rates have shown to be useful not only for predicting ^{222}Rn flux but also for the harmonization of networks. This would allow authorities to compare measurements of the γ-dose rates at European boarders, in consideration of emergency monitoring. However, the extraction of the terrestrial part of the total γ-dose rate measurements has shown to be more complicated than expected. The subtraction of the contributions of cosmic rays and anthropogenic radiation (^{137}Cs) was the same for all field sites, and caused no seriuous problems. But as 30 countries are participating in EURDEP, almost the same number of measurement devices and data handling appeared, which had to be harmonized. Another difficulty was the self-effect (inherent background) of γ-probes, which could be different for any device of the networks. Therefore, some assumptions had to be made, to estimate these effects in order to finally extract the terrestrial γ-dose rate.

In a third step, the combination of the correlation between ^{222}Rn flux and terrestrial γ-dose rate and the terrestrial γ-dose rate maps resulted in annual, seasonal and weekly ^{222}Rn flux maps for Europe. These confirmed previously made studies, as for example the linearly decreasing ^{222}Rn flux rate northwards (most likely due to increasing soil moisture and snow cover), but also resulted in substantial improvements regarding spatial resolution. We found a mean ^{222}Rn flux value of 0.55 atom cm^{-2} s^{-1} for Europe, which is far beyond the standard assumption, with a coefficient of variation of 51%, resulting from values ranging from 0.03 to 1.76 atom cm^{-2} s^{-1}.
The application within the TM5 atmospheric tracer model showed that the spatial resolution improves not only the average model predictions over longer time periods (weeks, month) but also considers directions of air mass trajectories in a short time period (days) at one location.

The application of the methods used to produce ^{222}Rn flux maps over large areas can also be applied to other continents, such as the Russian and USA territory, where γ-dose rate maps already exist.
Now, as the uncertainty of the ^{222}Rn source term for atmopsheric transport models could be improved, the uncertainty of measurements of ^{222}Rn air concentrations is still remaining. Hence, for the comparison of different atmospheric transport models the calibration of ^{222}Rn air concentration device monitors is highly desirable.

Appendix A

Discussions and Reviews

A.1 Discussion on "Atmospheric Chemistry and Physics Online Discussion" for Chapter 2

A.1.1 Reviewer Comment (Anonymous)

Received and published: 15 February 2007
Interactive comment on Atmos. Chem. Phys. Discuss., 7, 1877, 2007.

General Comments:
I think this is a valuable paper worthy of publication. The ^{222}Rn flux from the earth's land surface is an important subject that is sorely in need of more information related to spatial and temporal variation and overall quantification. Especially needed are field measurements of ^{222}Rn flux for diverse regions and conditions both to be used directly,and for calibration of models. An important aspect of this paper is its presentation of valuable new field measurements of ^{222}Rn flux for central Europe. Although the subject of a correlation between ^{222}Rn flux and γ-dose rate is not new, this paper provides new data and appropriate evaluation for the selected European countries. The authors do a good job of discussing some of the sampling issues related to more localized ^{222}Rn flux measurements versus γ-dose rate measurements that is probably average over a larger region of soil. The paper could be strengthened by some revision. Radon flux measurements using the closed chamber method are a tricky business. Although the authors offer a fair amount of discussion and references, I'm still worried they may have missed some things. Careful calibration of their chamber technique may not be so important for conclusions related to the correlation with γ-dose rate, but for archival purposes and future use by other scientists, the subjects of calibration and accuracy are worthy of careful attention. Since the authors have gone to a lot of trouble making measurements at diverse locations, it would be a shame not to have as much information as available on accuracy and calibration. Similarly, it would be helpful to have a little more background information on the location (regional map?) and conditions of flux measurements (soil properties? meteorology? climate?) and a more quantified breakdown of results by location and/or measurement conditions. In other words, if a new researcher wishes to apply the same methodology at some new location or time, provide him/her with as much information as possible for projection of expected results for the new situation.

Specific Comments:

Let me start with some comments on the closed can technique. Since the authors have gone to a lot a trouble to make measurements at diverse locations, it would be nice if we could have as much confidence as possible in the accuracy of the results for use by others in perhaps other contexts. One of the best ways to establish accuracy in this field is to do intercomparison measurements with other research groups. If something like this has been done, it would be helpful if the authors specifically referenced it and gave a one sentence summary. Unfortunately, ^{222}Rn flux standards usually do not exist in this field, so lacking intercomparison measurements, the authors are then left to try to estimate errors by analysis. In section 2.5 the authors mention a ± 15 % error. Where did this come from? I can name two potential sources of error that especially worry me and I could not find addressed (they might be buried in a listed reference I do not have). One is the subject of 'back diffusion': it does not take much build up of radon in a chamber for that radon to reduce the gradient from soil in a very tiny surface soil layer and reduce the apparent flux density of radon. Have the authors considered this effect? This subject has been around for a long time. Some of the more recent references related to it are Mayya (2004). Another more general reference is Hutchinson *et al.* (2000). Another source of error that can be important is a difference in pressure between the inside and outside of the chamber. A pressure difference as small as a fraction of one Pascal can be important. Now pressure differences much larger than this are possible due to the pumps and lines sampling the radon from a chamber, depending on how they are connected. Sometimes two pumps are required, one at the inlet and one at the exhaust to avoid impedance-caused pressure drops.
Sometimes a Bernoulli effect from wind can cause a pressure difference. The only way to be certain is to check the pressure difference under field conditions. Have the authors considered this issue? The authors discuss some other factors affecting ^{222}Rn flux such as local variability in soil properties and rain (soil moisture). There are other possible factors. Did they see any correlation with other factors such as temperature or atmospheric pressure? For example, there seem to be some systematic patterns in Figure 2.3 top that are not explained by rain. Do they have ideas about the explanation? Any chance of an effect due to diurnal atmospheric pressure variation? On the subject of more complete, and quantitative, reporting of results here are some thoughts. I have in mind trying to have a fuller understanding of the background conditions for the results reported by Table 2.2, Figure 2.1c, Figure 2.2, and Figure 2.3(top), but only to the extent that such information is easily available. Would a map help for better referencing the location of measurements? Can the authors supply any other supporting information for measurements conditions? If not, say, averages for detail like radium content of soil and soil moisture, maybe more generic information such as soil type, climate classification, etc. If grass or small plants were present at a sampling site, were chamber measurements made over them or were measurements restricted to bare soil? Maybe indicate whether numbers such as given in Table 2.2 are averaged over all times of days or seasons, or some specific time of day or season. Would it possibly help to provide both median and means for certain tabulated results? The authors should be able to see the gist of what I have in mind. In other words, if I wanted to go back to your sites and replicate your results, what would be helpful to know. The authors do not need to go overboard here, only give what could be helpful and easily provided in a short paper. A major conclusion of the paper is establishing the correlation between ^{222}Rn flux density and γ-dose rate. The authors make a very convincing case. It might be worth giving more quantitative detail such as the actual factor to multiply γ-dose rate at some height to get average ^{222}Rn flux density.

Possibly include an estimated error in these factors.
One of the authors' main conclusions was that the relation between ^{222}Rn flux and γ-dose rate was fairly robust and constant from one region to the next. The authors commented about a correction for anthropogenic cesium. How about the following issue. Generally, outdoor γ-dose rate comes from the soil uranium series, the soil thorium series, soil potassium 40, and cosmic rays. There are geochemical reasons why uranium and thorium are often correlated in soils. However, γ-dose rate varies with altitude, and potassium geochemically is somewhat independent of uranium and thorium. I have seen radiation survey maps for the entire United States, and noted there are regions where the soil potassium is high without a proportional increase in the uranium/thorium series. Keeping in mind issues such as potassium variation and a cosmic ray altitude effect, do the authors care to speculate on how robust their conclusions will remain for regions beyond the area of Europe they studied?

A.1.2 Interactive Comment to Anonymous Reviewer

Received and published: 8 March 2007
Interactive comment on Atmos. Chem. Phys. Discuss., 7, 1877, 2007.

Thank you very much for your comments and your valuable suggestions for improving our manuscript.
Inter-comparison of instruments to measure ^{222}Rn flux are an important issue and large differences between instruments have been reported (Hutter and Knutson, Health Physics 74: 108-114, 1998). The instrument we used in our study was compared to the one described in Iida *et al.* (1996) in Robertson *et al.* (2005). The mean flux determined at six locations was with our instrument (52 Bq m^{-2} h^{-1}, standard error 9 Bq m^{-2} h^{-1}) and compared well with the mean flux measured with the instrument described in Iida *et al.* (1996) (49 Bq m^{-2} h^{-1}, standard error 8 Bq m^{-2} h^{-1}). As for other possible correlations between environmental parameters and ^{222}Rn flux, we have tested for correlations with air temperature, atmospheric pressure, soil temperature and difference between air and soil temperature. If one of these parameters was correlated with ^{222}Rn flux, it was very weak. We do not think these parameters have a strong direct effect on ^{222}Rn flux but rather coincide with precipitation events or dry spells. In principle, diurnal pressure variations may cause mass flow through periodic expansion and contraction of the soil gas volume and influence the otherwise mainly diffusion-driven exchange of radon between soil pore space and atmosphere. We would expect this to be a major factor in deeply weathered dry soils with large air volumes. In the commonly humid regions in Europe we studied, it might not be a major issue.
Also, the scale of the problem with back diffusion in chamber measurements is particularly large for soils with large air-filled porosity and with shallow chamber heights. Theoretically, it is possible to correct for back diffusion, if air-filled porosity is known (Conen and Smith, 2000). As we did not measure this parameter, we can not make reliable corrections. However, we estimate that not accounting for back diffusion might have lead to an average under-estimation of ^{222}Rn flux by up to 5%.
You mention pressure differences between inside and outside of the chamber as possible source of error. This is another issue that has received much attention in the past and still does (Xu *et al.*, 2006). Small pressure differences can cause major errors in flow-through (dynamic) chambers because they induce a sustained and continuous mass flow into or

out of the chamber. In static chambers like ours, pressure differences cause smaller errors because they are equilibrated quickly by a much smaller mass flow (i.e. a pressure deficit of 1 Pa in a 10 litre closed chamber is equilibrated by an inflow of 0.1 ml of soil air).
A table with field site coordinates, measurements, air pressure, soil texture properties etc. (i.e. all information a reader would need to go back to our sites and replicate our measurements) will be included in an improved version of our manuscript and made available for down-load from our website (http://www.radon.unibas.ch), which is under construction at the moment. All the in-situ measurements were carried out during daytime (between 10 a.m. and 4 p.m. local time) during summer and autumn 2005. The website will also provide downloadable data for all kind of ^{222}Rn flux maps we develop based on the approach discussed in the present manuscript.
The correlation between ^{222}Rn flux y (in Bq m^{-2} h^{-1}) and terrestrial gamma dose rate x (in nSv h^{-1}) we found was: y = 0.995 ($\pm$0.10) x - 14.97 ($\pm$8.11). Proportion of U decay series to total gamma dose rate is relatively constant in Europe (see reply to Heinz Surbeck). Outside Europe, we will have to investigate this in a further study. For further publications we are investigating possibilities to compare our model to data and models for other countries (e.g. China, North-America).

A.1.3 Reviewer Comment (H. Surbeck)

Received and published: 20 February 2007
Interactive comment on Atmos. Chem. Phys. Discuss., 7, 1877, 2007.

The authors have done a tremendous and very useful work by measuring ^{222}Rn fluxes at many sites in Europe. It would be worth publishing these data as a map or as a table, together with details about how the measurements have been done. But I'm not convinced that the data shown in the paper submitted are sufficient evidence for the claims made.

Specific Comments
Figure 2.2 shows that terrestrial dose rate and ^{222}Rn flux are not well correlated. There are several good reasons for not using terrestrial dose rate as a proxy for the amount of radon escaping to the atmosphere :

1) Decay products from the ^{228}Ra series and ^{40}K contribute the same order of magnitude to the dose rate as ^{226}Ra decay products do. Neither the ^{228}Ra/^{226}Ra nor the ^{40}K/^{226}Ra ratios are constant and thus terrestrial dose rate will not be a good proxy for the ^{226}Ra concentration unless these ratios are known.

2) Depending on where the ^{226}Ra is located, emanation coefficients can vary by a factor of 10. Only some% of the radon produced inside a grain will escape to the pore space, whereas up to more than 50% of the radon produced from radium adsorbed on thin layers covering the grains (Mn- or Fe-oxyhydroxides) will be available for transport through the soil. The soils mentioned in the Von Gunten *et al.* (1996) paper are an extreme case but large variations for the emanation coefficient are known from other studies too. So even if the ^{226}Ra concentration in the soil is known, the radon concentration in the pore space cannot be calculated unless the emanation factor is known. Figure 2.1c clearly shows that ^{226}Ra and ^{222}Rn flux are not well correlated.

3) Dose rate is a measure for the radon (and thoron) that could not escape to the atmosphere. A high emanation from the soil thus would result in a decrease in the dose rate and not in an increase.

4) Dose rate is not very sensitive to layered soil structures (and if, it goes the wrong direction), but radon emanation clearly is. Frequently soils are less permeable close to the surface due to fine grained weathering products. This reduces radon emantion, but will not reduce dose rate, rather increase it because of radon decay products building up below this less permeable layer.

A.1.4 Interactive Comment to H. Surbeck

Received and published: 8 March 2007
Interactive comment on Atmos. Chem. Phys. Discuss., 7, 1877, 2007.

Thank you for your comments reflecting a more cautious view on our approach. In principle, we agree that each of the points you made is valid. The factors you mention can be of major importance at small scales, such as relevant for predicting the radon potential at a building site. Yet, the scale we are interested in, is much larger. We would like to be able to predict average ^{222}Rn flux for a country or at best for a particular region, such as Central France or Northern Germany. At these scales, the effects of the factors you mention largely even out.
As a first example, we may take the contribution of the ^{238}U decay series to the total gamma dose rate, relating to your point (1). We are aware that ^{238}U and ^{226} may not always be in secular equilibrium but data on ^{238}U is more widely reported and allowing us to discuss your concern. Proportions of the contribution of the ^{238}U series to total gamma dose rate are reported in our manuscript for Switzerland, for North- West Italy in Chiozzi *et al.* (2002), for Spain in Quindos Poncela *et al.* (2004) and for Cyprus in Tzortzis *et al.* (2003). Contributions of the ^{238}U series for individual types of rocks reported in these four studies range from 12% to 90%. However, the average for each country or region ranges from 27% (Spain), 29% (North-West Italy) to 30% (Cyprus, Switzerland). Thus, in the context of our objective to predict larger scale averages for ^{222}Rn flux, it seems justified to assume a constant contribution of the ^{238}U series to the total gamma dose rate.
A second example relates to your point (2). Indeed, the emanation coefficient for radon can vary by a factor of 10. Again the magnitude of this variation is a question of scale. Greeman and Rose (1996) determined emanation coefficients for each horizon in 12 contrasting soil profiles in the North-East of the United States. Emanation coefficients ranged from 5.5% to 33% for individual horizons. However, average emanation coefficients for entire soil profiles only ranged from 13% to 29% and two-thirds of the soil profiles were in the narrow range between 18% and 22%.
Regarding your point (3), there might be a misunderstanding. You write that dose rate is a measure for the radon (and thoron) that could not escape to the atmosphere and a high emanation from the soil thus results in a decrease in the dose rate and not in an increase. This would be correct, if we related ^{222}Rn flux to the dose rate component of radon and its daughters only. This is not the case. We relate ^{222}Rn flux to the total terrestrial gamma dose rate, including the ^{238}U series, ^{232}Th series and ^{40}K. Grasty (1997) once made a theoretical study of the effect you may have related to. He predicted an increase in the

gamma dose rate originating from the ^{238}U series of about 6% when moisture saturation increases from 0.70 to 0.95 (assuming an emanation coefficient of 25%). However, the increase in the contribution of the ^{238}U series is more than compensated by a concurrent decrease in the contributions of the ^{232}Th series an ^{40}K. Thus, the overall result of increasing soil moisture is a decrease in total gamma dose rate. This decrease coincides with a decrease in ^{222}Rn flux resulting from increased tortuosity and diffusion resistance of the moister soil and is in line with our findings. Extremely low rates of emanation are also possible in very dry soils with water contents $< 5\%$ (Bossew, 2003). In these soils the probability is high that radon atoms are captured by neighbouring grains due to their high recoil energy not being partly absorbed in a water film covering the grain of origin. Thus, these atoms can not escape respectively are no longer available for exhalation. But as these observations were made in the laboratory and water contents smaller than 5% are very unusual in nature, so we consider this effect to be negligible. In point (4) you make the case that soils are frequently less permeable close to the surface due to fine grained weathering products, reducing ^{222}Rn flux but not dose rate. We consider exchange of radon between natural soils and the atmosphere to be largely driven by diffusion, not by mass flow. Thus soil diffusivity rather than permeability will determine ^{222}Rn flux. In general, soil bulk density increases with depth and so does soil moisture, resulting in reduced air filled porosity and diffusivity with increasing depth. The importance of this effect is probably minor because the contribution to the surface flux of radon decreases rapidly with the depth of any particular soil horizon.
We were inspired by your comments to look again at our approach and underlying assumptions. Our conclusion is that our assumptions may not always be valid on a small scale but that they hold at the larger scale at which we try to predict ^{222}Rn flux. This conclusion is supported also by the close correspondence between predicted and measured regional ^{222}Rn flux shown in Table 2.2 of our manuscript.

A.2 Reviewer comments at "Radiation Measurements" for Chapter 3

Francesco d'Errico
Editor-in-Chief, Radiation Measurements
17 August 2007

Congratulations for an excellent work linking many aspects of environmental radiation monitoring: detector basis, national networks, data exchange, geostatistics and challenging proposals for mapping from data analysis.

I fully agree with the method and comments on the influence of inherent background. However, I have some comments on the cosmic substraction. First, please state that 'The cosmic radiation AT GROUND LEVEL mostly consists of muons' (at higher altitudes there are many other ionising particles, including neutrons). Authors mention that the latitude influence is negligible, but I'm not so sure about this sentence especially considering that EURDEP data covers roughly from parallel 35°N (Spain) to 80°N (Norway). I guess that this influence is at least as the reported variance due to changes in air pressure (1–3 nSv h^{-1}). An adequate reference or some data should be given to support that latitude effect is negligible.

Simplication on artificial sources is needed and it seems to work. However, there are many papers reporting different data but most of them are based on very local studies that are clearly out of the scope of this paper.

Perhaps the only weak point of the paper is that the influence of the detector photon energy response and calibrations are factors that affect the results even more than the precedent. For instance, many national networks use GM-based radiation detectors that show important over-response (up to 30%) when comparing ^{60}Co exposures to ^{137}Cs exposures. This means that if the GM is calibrated with ^{137}Cs and then it is exposed in regular natural radiation environments where the mean photon energy is closer to 1 MeV due to ^{40}K and U&Th daughters, the instrument will overestimate the dose rate by at least 20%. This is probably why the data from Spain and Finland are regularly higher than from neigbouring countries. However, this point does not affect to the aim of the paper to compare winter and summer seasons to get information on radon.
I specially appreciated the inclusion of Geostatitics which is an essential tool for this field. The way on which the tool is explained is clear, the references are good and the conclusions are relevant. Just to say, well done!
Please correct Table 3.1 and order the data according the Country name in English. (I think at least Germany, Switzerland Croatia and Spain are positioned according to the name in their own languages). It would be easier to read for any international reader!
As the authors state, it is not easy to get information about Rn from γ-dose rate measurements even when most of the factors are controlled. So, it is really remarkable to note the good results of the paper, specially considering the multiple difficulties and inhomogeneities in the analysed data. That means that the employed methods in the analysis and the corrections implemented are on the right way.

Appendix B

Predicting terrestrial ^{222}Rn flux using γ-dose rate as a proxy

This Appendix contains additional, unpublished data from Chapter 2. Beside an itemised correlation of all measured ^{222}Rn flux and γ-dose rates during the measurement campaign the long-term measurements in Switzerland are shown here, as well as the the complete continuous measurements of ^{222}Rn flux, γ-dose rate, soil moisture and precipitation in Basel (Switzerland).

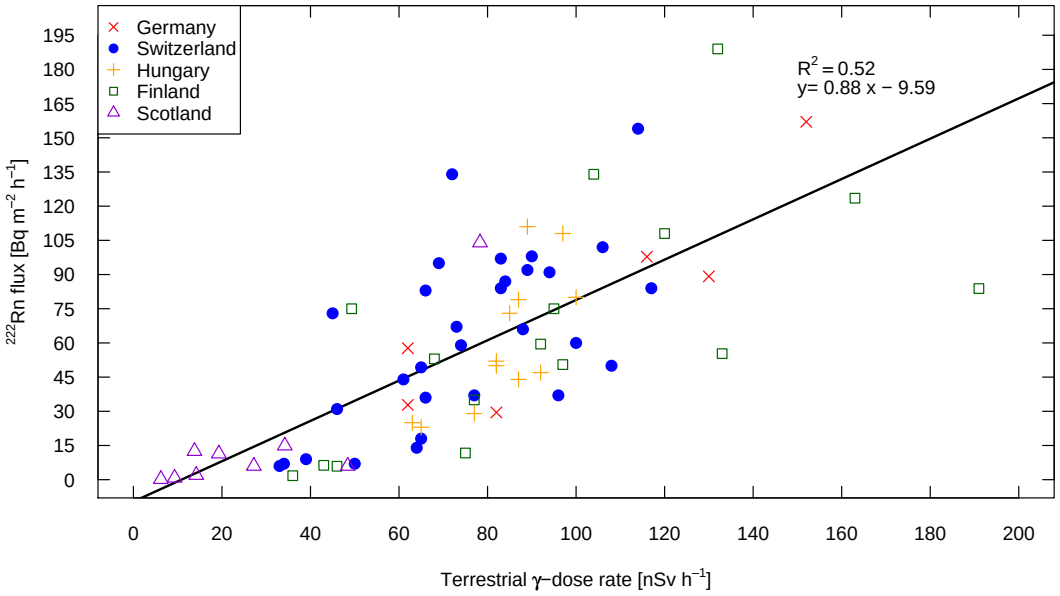

Figure B.1: Complete date for correlation of ^{222}Rn flux and terrestrial γ-dose rate measured at field sites in Switzerland, Germany, Finland and Hungary. Additionally data from Robertson (2005) was taken for scottish ^{222}Rn flux measurements.

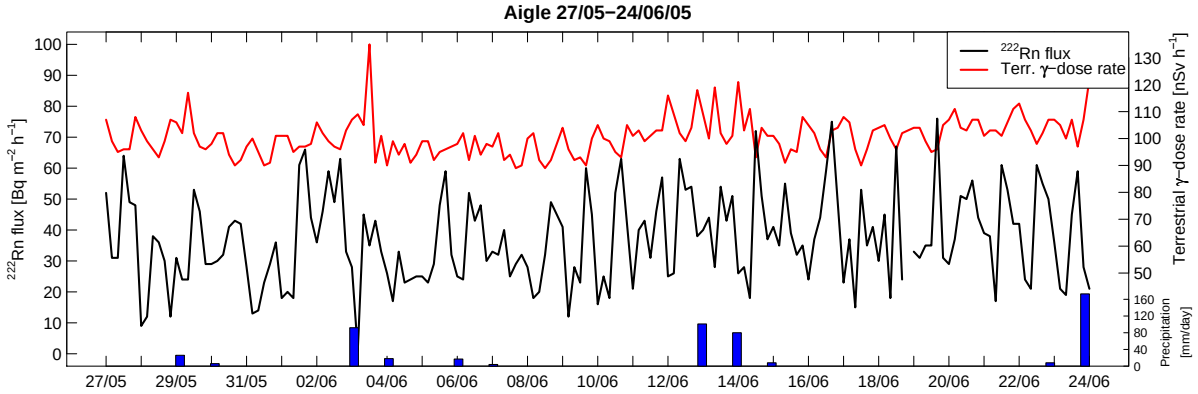

Figure B.2: Time series of ^{222}Rn flux and terrestrial γ-dose rate measurements in Aigle from 27/05/05 - 24/06/05

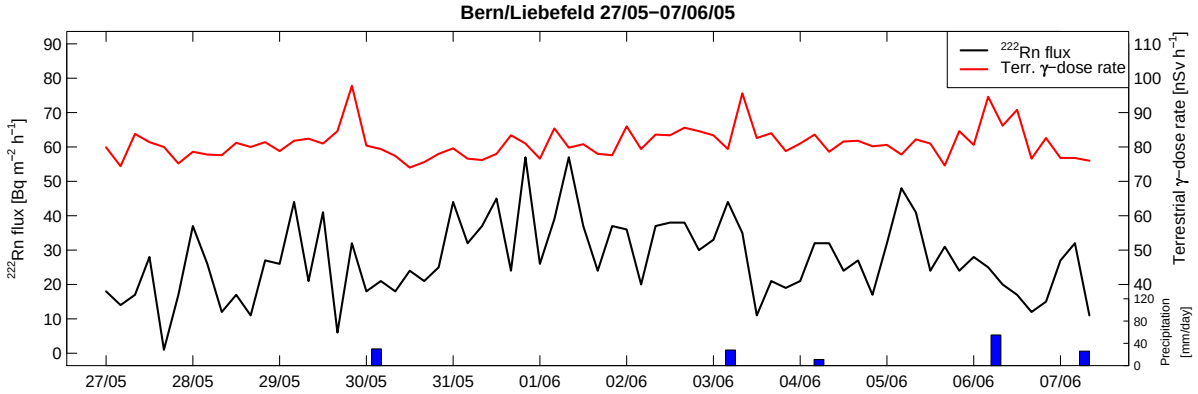

Figure B.3: Time series of ^{222}Rn flux and terrestrial γ-dose rate measurements in Bern/Liebefeld from 27/05/05 - 07/06/05

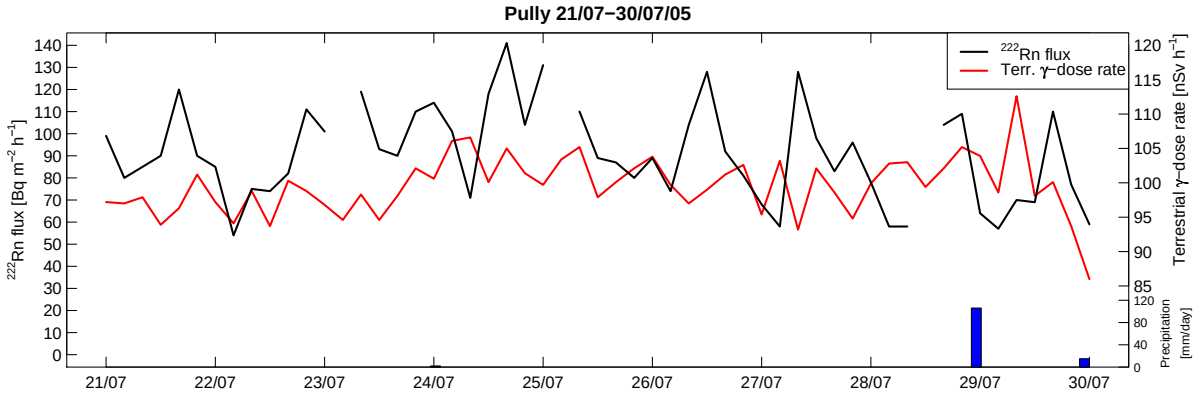

Figure B.4: Time series of ^{222}Rn and terrestrial γ-dose rate measurements in Pully from 21/07/05 - 30/07/05

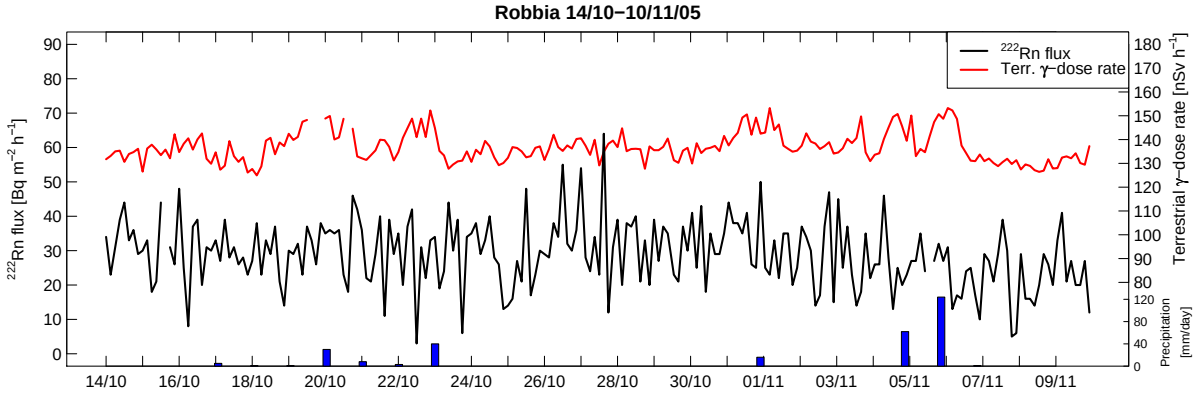

Figure B.5: Time series of ^{222}Rn flux and terrestrial γ-dose rate measurements in Robbia from 14/10/05 - 10/11/05

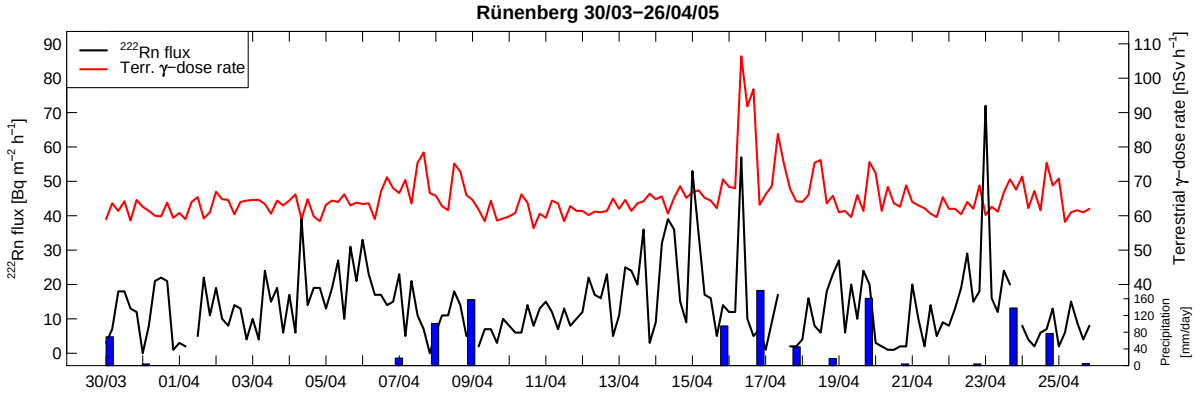

Figure B.6: Time series of ^{222}Rn flux and terrestrial γ-dose rate measurements in Rünenberg from 30/03/05 - 26/04/05

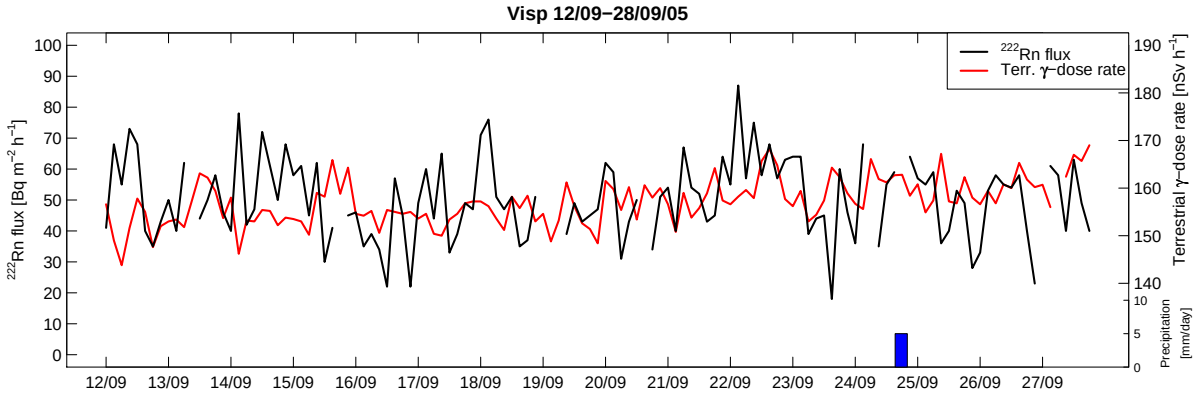

Figure B.7: Time series of ^{222}Rn and terrestrial γ-dose rate measurements in Visp from 12/09/05 - 28/09/05

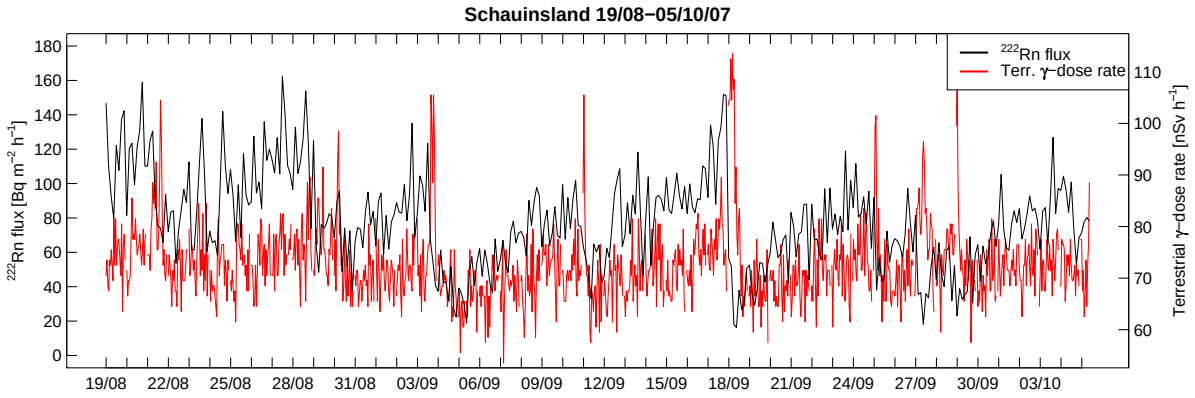

Figure B.8: Time series of ^{222}Rn flux and terrestrial γ-dose rate measurements at Schauinsland (Germany) from 19/08/07 - 05/10/07 (no precipitation data available so far)

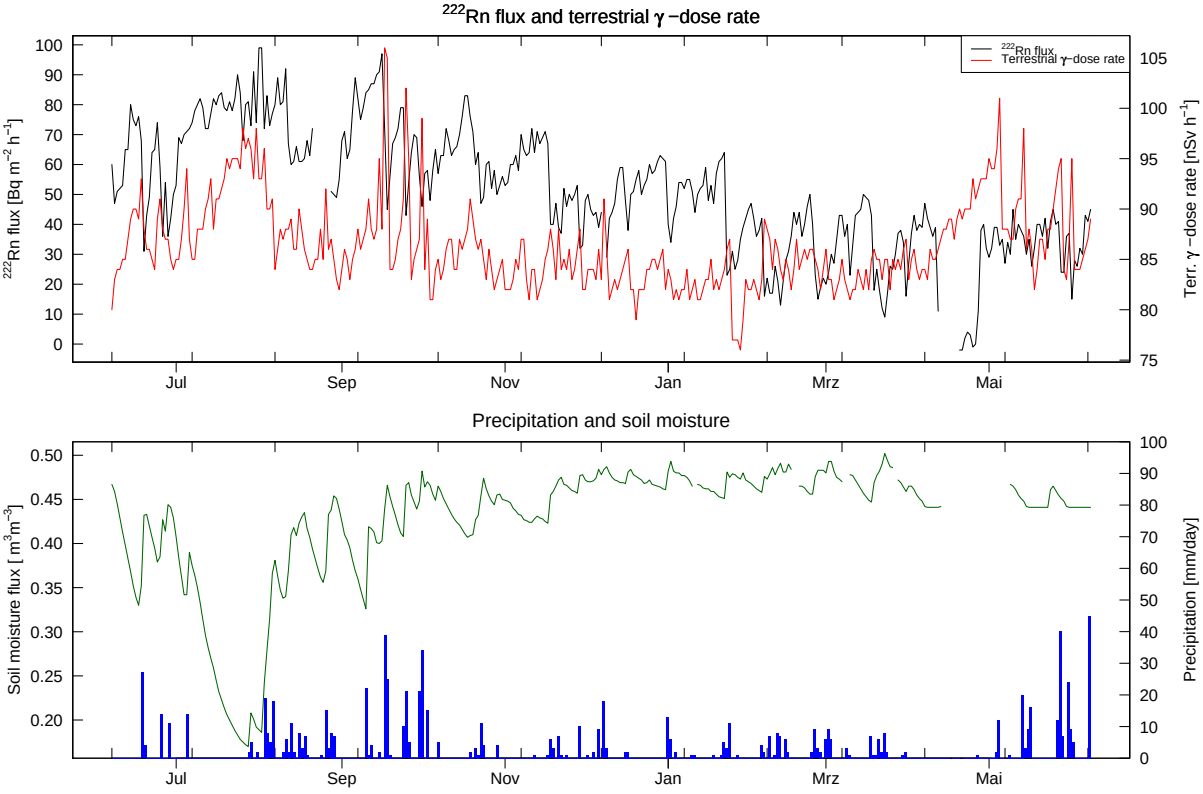

Figure B.9: Time series for one year of ^{222}Rn flux and terrestrial γ-dose rate measurements in Basel/Binningen, and additional measurements of soil moisture and precipitation

Appendix C

European ^{222}Rn source description for applied atmospheric studies

C.1 Results from the atmospheric tracer model TM5

This Appendix contains data from the atmospheric tracer model TM5, for which Mariella-Gabriella Villani at the JRC in Ispra made a run with the standard assumption for the ^{222}Rn flux of 1 atom cm^{-2} s^{-1} and the new ^{222}Rn source term presented within this PhD thesis. Figure C.1 presents monthly mean values for the modelled data as well as the observed ^{222}Rn flux. The following diagrams (Figure C.2 to C.7) show time series for 6 stations in Europe with the corresponding description of these field sites (Table C.1).

Station	Code	Longitude	Latitude	Altitude a.s.l.
Rostock	RST	12.08°E	54.18°N	0 m
Mace Head	MHD	9.90°W	53.33°N	5 m
Freiburg	FRB	7.85°E	48.00°N	180 m
Schauinsland	SIL	7.91°E	47.92°N	1'205 m
Gif-sur-Yvette	GSY	2.13°E	48.70°N	160 m
Hohenpeissenberg	HOH	11.02°E	47.80°N	780 m

Table C.1: Description of the field sites, where observations of ^{222}Rn concentrations are available

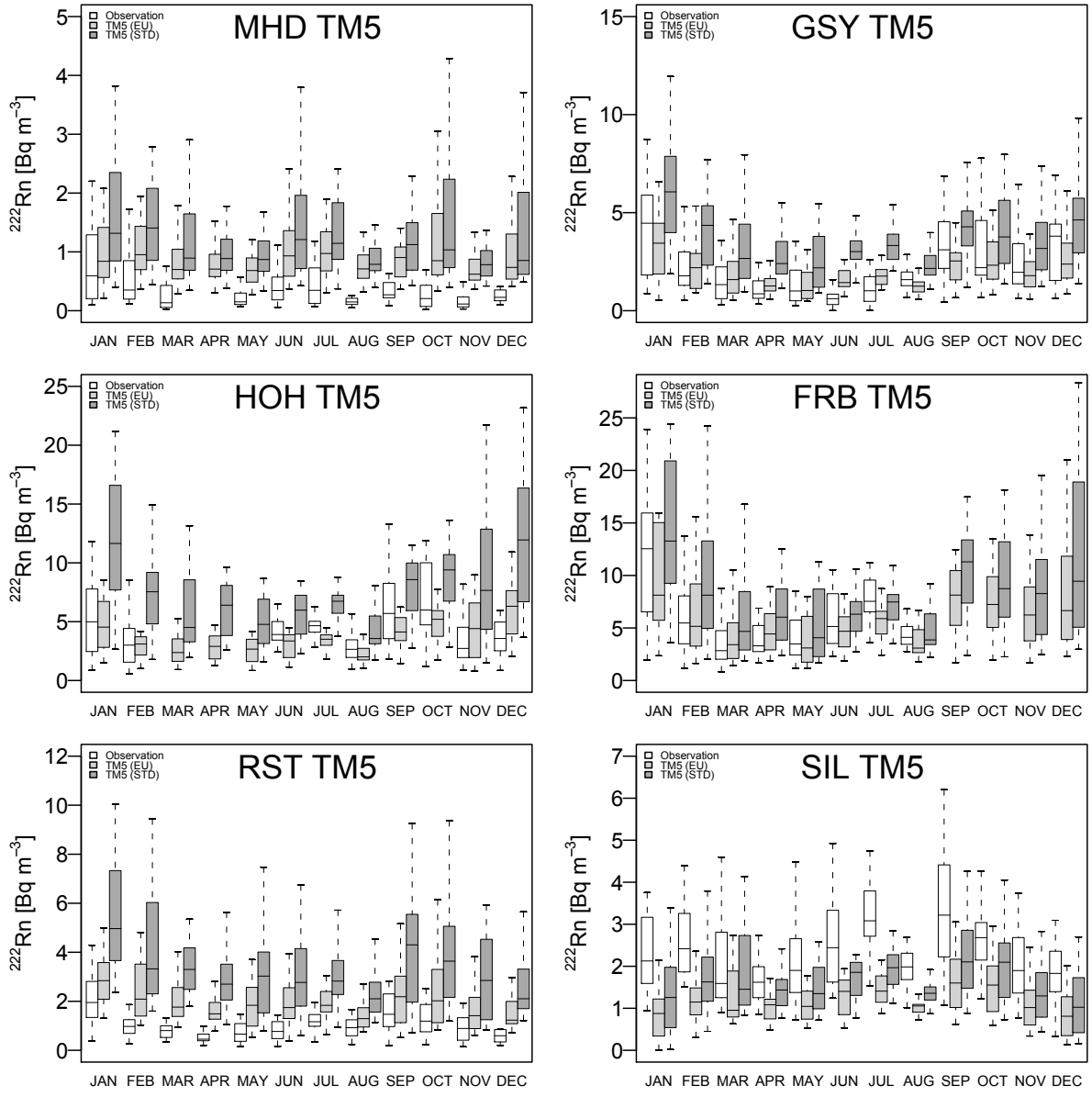

Figure C.1: Boxplots for monthly average ^{222}Rn concentrations for Mace Head (MHD), Gif-sur-Yvette (GSY), Hohenpeissenberg (HOH), Freiburg (FRB), Rostock (RST) and Schauinsland (SIL) for 2006. White boxes indicate observed data, lightgrey boxes are modeled data with the new ^{222}Rn inventory (EU) and dark grey boxes are standard modeled data (STD)

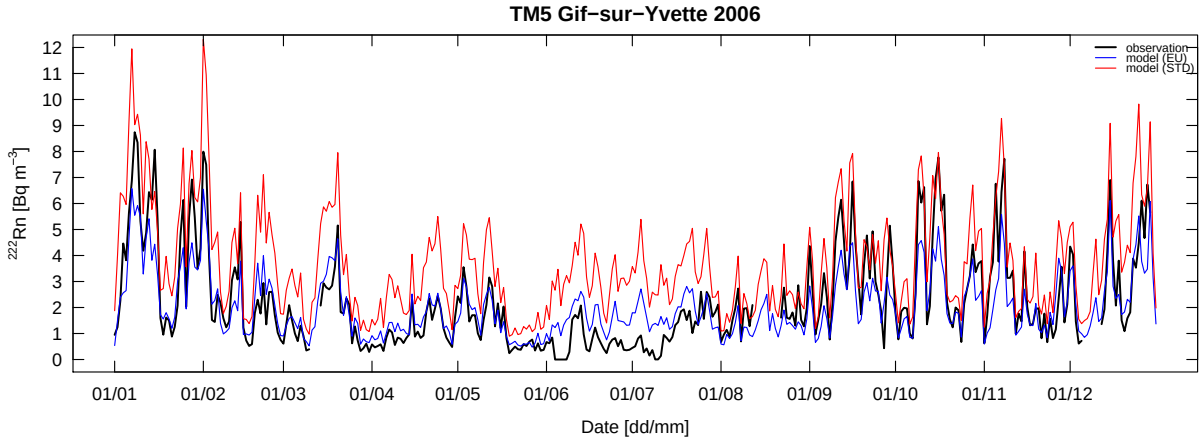

Figure C.2: Time series of modelled and observed ^{222}Rn concentrations in air with TM5 for Gif-sur-Yvette for 2006

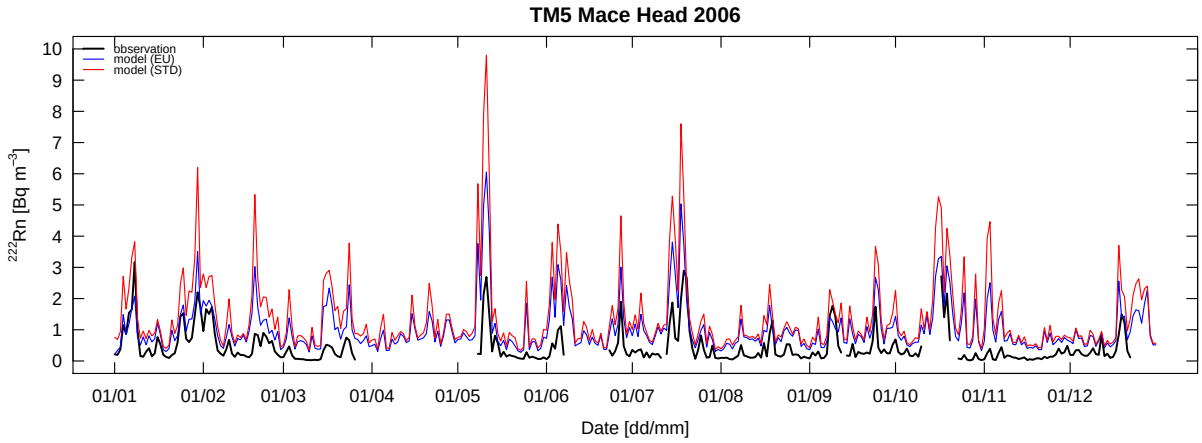

Figure C.3: Time series of modelled and observed ^{222}Rn concentrations in air with TM5 for Mace Head for 2006

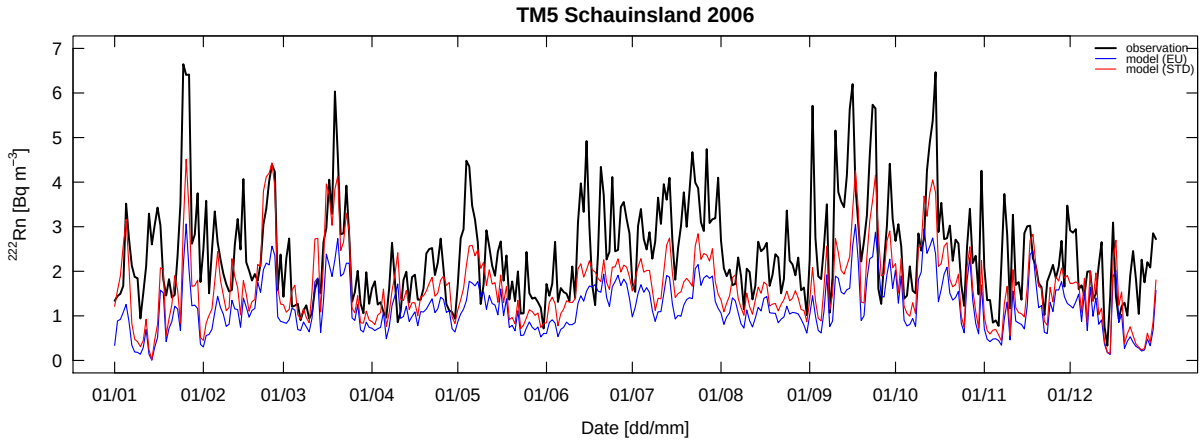

Figure C.4: Time series of modelled and observed ^{222}Rn concentrations in air with TM5 for Schauinsland for 2006

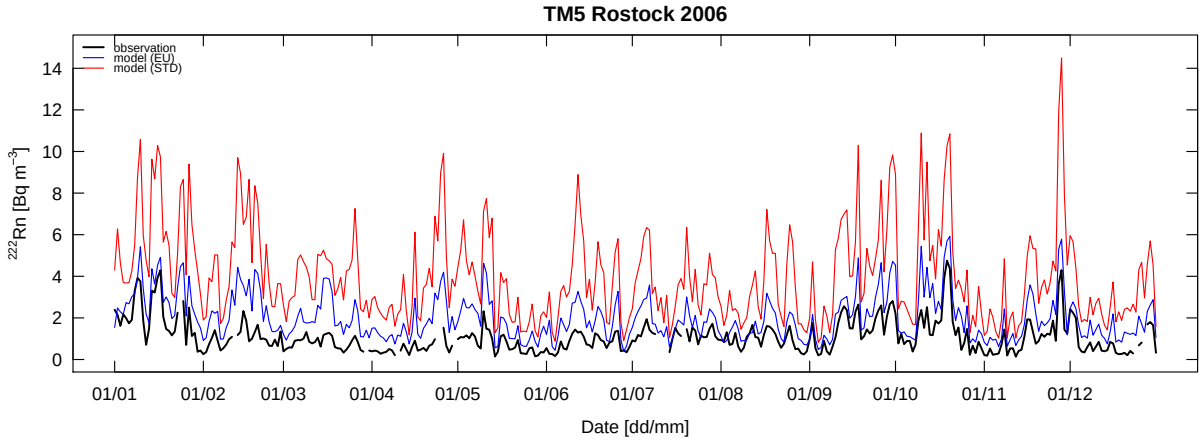

Figure C.5: Time series of modelled and observed ^{222}Rn concentrations in air with TM5 for Rostock for 2006

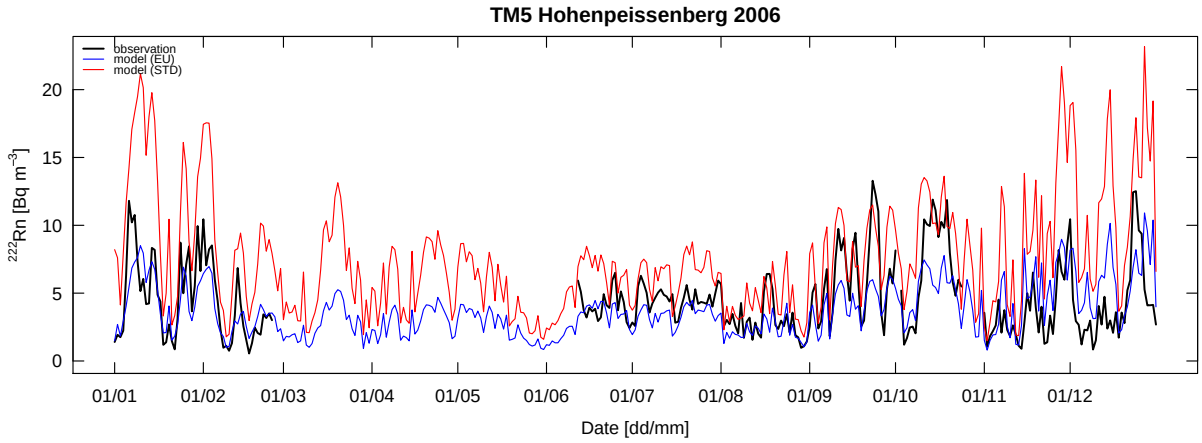

Figure C.6: Time series of modelled and observed ^{222}Rn concentrations in air with TM5 for Hohenpeissenberg for 2006

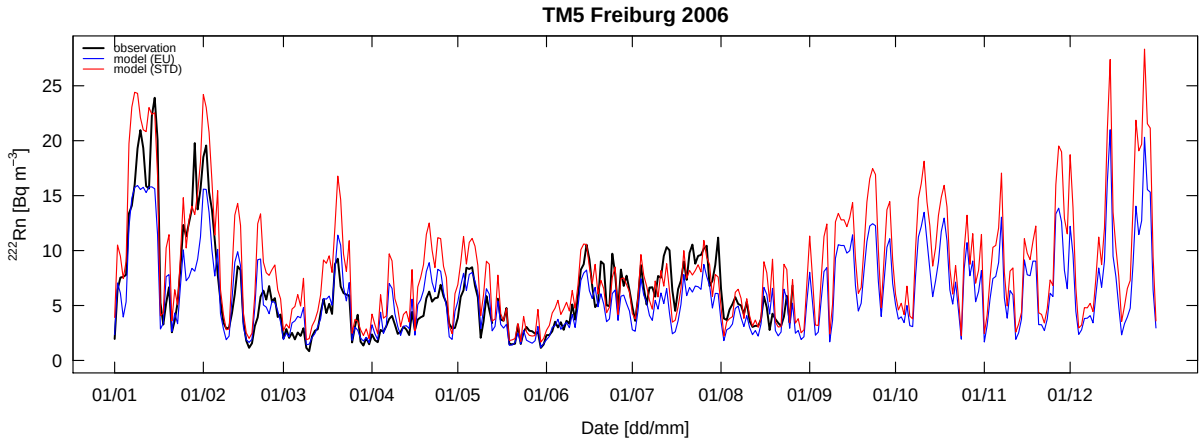

Figure C.7: Time series of modelled and observed ^{222}Rn concentrations in air with TM5 for Freiburg for 2006

C.2 ^{222}Rn flux maps for the United States of America and the Russian territory

The following maps are representing ^{222}Rn flux estimations for the Russian Federation territory and the United States of America territory (conterminous USA). As the map for Europe is based on a terrestrial γ-radiation map for Europe, which is based on continuous measurements of γ-dose rate in Europe (emergency monitoring networks), the maps presented here are provided by a a sligthly other base map, because for Russia and the USA such networks were not available. For Russia, a γ-dose rate map (Vysokoostrovskaya *et al.*, 1995), derived from aeroradiometric measurements, was used as the base map to be converted into the ^{222}Rn flux map. Using the same function as in equation 4.1, a large step change in the magnitude of ^{222}Rn fluxes at the boarders to the European ^{222}Rn flux map appeared. The reason is possibly because during the field measurements for the correlation between ^{222}Rn flux and γ-dose rate in chapter 2, the measurements were referenced to γ-probes (Geiger-Müller tubes) at 1 m above ground. Aeroradiometric nuclide (respectively γ-dose rate) measurements are completely different (higher altitudes, different measurement devices) and therefore our approach might not have the same reference. So we corrected the Russian ^{222}Rn flux map minimising the step change at European boarders by deviding the Russian values by a factor of 2.5.
The base map for the USA ^{222}Rn flux map is a map for ^{238}U, ^{232}Th and ^{40}K activities from the US Geological Survey (Duvala *et al.*, 2005). These maps were produced by aeroradiometric surveys, possibly presenting similar problems as for the Russian map discussed above. An additional bias may be included by converting nuclide activities (in Bq kg^{-1}) into γ-dose rates (in nSv h^{-1}) using the conversion factors by Grasty *et al.* (1984).
Using equation 4.1 to convert the map into a ^{222}Rn flux map, the results showed significantly low (and negative) values. We assume the same problems appeared as for the Russian map.
We had no occasion to compare the results as for the Russian map, which is bordering the European ^{222}Rn flux map, and therefore further assumptions had to be made. These assumptions were (a) no significant small or negative ^{222}Rn fluxes, (b) a mean flux of 0.87 atom cm^{-2} s^{-1} as described in Piliposian and Appleby (2003) and (c) a similar coefficient of variation in fluxes as in Europe.
A best estimate was found for the correlation function

$$^{222}Rn flux\ [atom\ cm^{-2}\ s^{-1}] = 21.92 \cdot \gamma\text{-}dose\ rate\ [\mu Sv\ h^{-1}] - 0.08 \qquad \text{(C.1)}$$

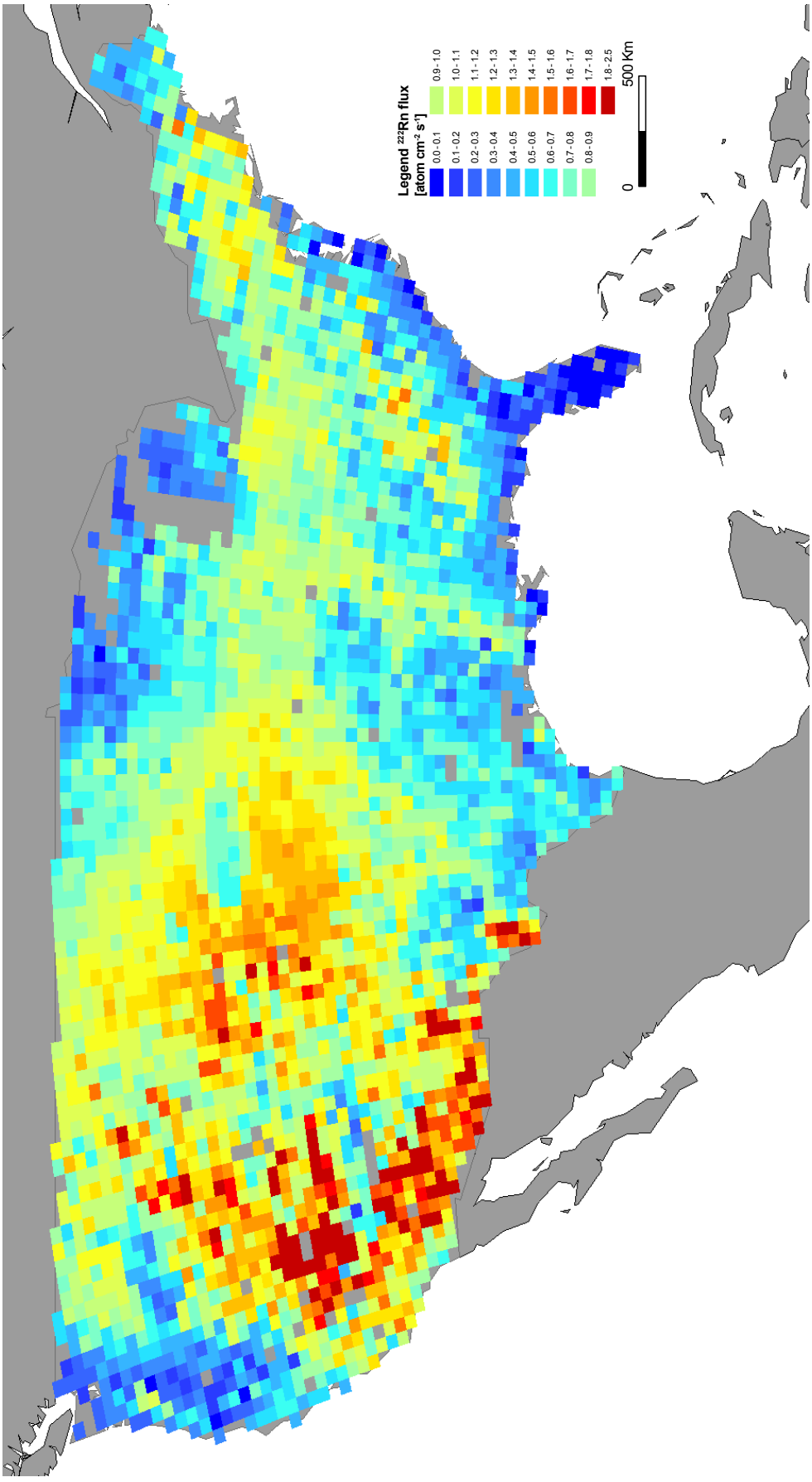

Figure C.8: ^{222}Rn map for the United States territory, based on aeroradiometric γ-ray measurements by the US geological survey (Duvala *et al.*, 2005), with a resolution of 50 x 50 km

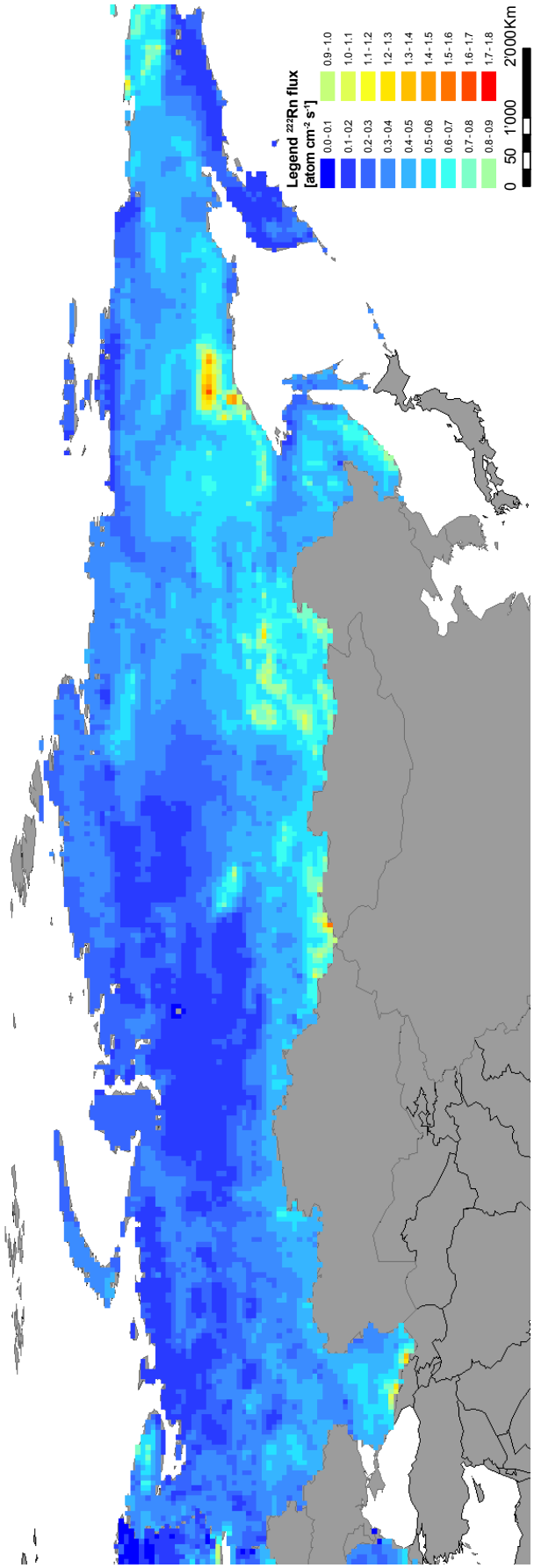

Figure C.9: ^{222}Rn map for the russian territory, based on aeroradiometric γ-ray measurements (Vysokoostrovskaya *et al.*, 1995), with a resolution of 0.5° x 0.5°

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