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Analysis of radon origin by backward atmospheric transport modelling

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ABSTRACT

This work shows how ambient radon concentrations measured at Cabauw station in central Netherlands are influenced by transport from different regions under typical transport conditions occurring during April and November, 2007 by means of atmospheric Lagrangian particle dispersion modelling in a receptor-oriented approach. Four specific regions have been isolated to assess their contribution to the modelled radon ambient concentrations at Cabauw, and two different radon flux assumptions.. Westerly flows coming from the ocean are poor in radon and do not increase radon air concentrations unless there is some fetch over the British Isles. Continental transport, mainly from eastern and southern Europe, significantly increases radon background concentrations, reaching increments of 3 Bq m^{-3}. A constant 0.66 atoms cm^{-2} s^{-1} radon flux over land and zero over water bodies is a good approximation for the source term in order to study regional contributions and modulation of the radon background.

1. Introduction

Back trajectories and trajectory statistics based on residence time analysis have been used extensively to determine the possible origin of air masses arriving at a particular site and consequently the probable source regions of pollutants observed at that site (Stohl et al., 1998; Sakashita et al., 2004; Chambers et al., in press). Recently, and thanks to improvements in computing performance, a switch from simple trajectory models to Lagrangian particle dispersion models (LPDM) has taken place. These models fully account for dispersive phenomena occurring in the atmosphere (Stohl, 1998), in contrast to the trajectory models. Nowadays, LPDMs are widely used in air quality studies as long as no second-order chemical reactions are involved. Should this be the case, Eulerian models are the method of choice because of the easier implementation of nonlinear chemical reactions. However, Eulerian models suffer from numerical diffusion and instantaneous mixing of point-source emissions in the grid cell. For receptor-oriented studies, where the receptor acts as a point source of an adjoint tracer, LPDMs are therefore the method of choice.

The physical and chemical characteristics of radon (Porstendörfer, 1994), as well as its well-defined origin from the soil, make it an excellent tracer for atmospheric transport studies. It is an inert radioactive gas, mainly exhaled from land bodies (Conen, 2003; Schery and Huang, 2004) and is unaffected by removal processes apart from its radioactive decay. Moreover, its half-life of 3.82 days makes it an invaluable tool to identify air masses which have passed over land within the previous few days or, on the other hand, those having passed over the sea and are hence poor in radon (Whittlestone et al., 1992).

^{222}Rn has been widely used in atmospheric transport studies ranging from very local scales, in order to analyse vertical mixing within the atmospheric boundary layer (Lee and Larsen, 1997; Lupu and Cuculeanu, 2001), up to regional and hemispheric scales, in order to validate atmospheric transport models (Feichter and Crutzen, 1990; Zahorowski et al., 2004; Conen, 2003).

Radon or radon progeny concentrations in ambient air are measured at various stations, both at inland and coastal sites, all over the world on a continuous basis, typically with hourly resolution. In Europe, one of these stations is the Cabauw tower in the centre of the Netherlands. This station is particularly interesting because it provides half-hourly measurements at two vertical levels (20 and 200 m a.g.l.), and because it is not far from the coast, allowing to study radon in maritime as well as in continental air masses. The aim of this work is to analyse how radon concentrations at this particular site are related to different transport patterns. The state-of-the-art LPDM FLEXPART has been used to model the origin and transport pathways of radon arriving at Cabauw.

2. Station location

The Cabauw station (latitude 51.97°N, longitude 4.93°E, altitude −0.7 m a.s.l.) is located in the Netherlands about 50 km east from the

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North Sea. Its location is within a rather flat area with mainly agricultural land use and moist soils (Van Ulden and Wieringa, 1996). The soil type in the southerly region is mainly river-clay. North of the tower, the soil type is peat or peat on clay. In this moderately maritime region there is a predominance of westerly flows (HELCOM, 2001). Nevertheless, eastern and southern continental air masses often reach the area, hence a radon increase can be expected in such situations. This makes the Cabauw station perfect for this sort of study. It has the possible influences both of continental and oceanic flows and it is situated in flat terrain. This means that the only mesoscale phenomena that may influence the site are land-sea breezes, unlike other continental stations, which may be affected, for instance, by valley and slope wind circulations. However, the influence at the station of the land-sea circulation is only important when there is a very weak background flow, which may allow sea breeze to travel up to 100 km inland (Tijm et al., 1999).

3. Instrumentation

The Cabauw station includes a 213-m-high tower which provides vertical profiles of the meteorological data as well as tracer gas data including ^{222}Rn. ^{222}Rn is continuously measured at two different heights, 20 m and 200 m above ground level, with an ANSTO radon monitoring device (Whittlestone and Zahorowski, 1998; Zahorowski et al., 2004) based on the two-filter technique. Air is pumped in through a first filter with an inlet flow rate of 80 L min^{-1} to remove all radon progeny and aerosols, but not the radon gas itself. In a 1500-L delay chamber, the filtered air is given enough time to allow radon decay and therefore to obtain newborn radon progeny, which will be subsequently retained on a second filter. Afterwards, the activity of the progeny nuclides on this filter is measured by gross alpha counting and the radon air concentration is inferred from the progeny concentrations.

4. Methods

This section provides an overview of the atmospheric transport model and its set-up as well as the methodology to construct the time series of the radon air concentration from the model output and radon flux maps.

4.1. Atmospheric transport and dispersion modelling

Transport simulations were done with the Lagrangian particle dispersion model FLEXPART version 6.2 (Stohl et al., 1998, 2005, see also http://transport.nilu.no/flexpart). This model has been extensively validated using data from various long-range tracer studies (Stohl et al., 1998; Stohl and Trickl, 2001) and has been used in many long-range transport applications (e.g., Forster et al., 2001, 2004; Stohl et al., 2003). The model computes the trajectories of a large number of released fictitious particles which are transported and dispersed by the mean wind plus a turbulent contribution (Stohl and Thomson, 1999). Within the atmospheric boundary layer, turbulent velocities are obtained from the Langevin equation assuming Gaussian turbulence, a common approximation in long-range transport. In addition, a moist convection scheme has been introduced (Forster et al., 2007). For this study the model was driven with operational analyses from the European Centre for Medium-Range Weather Forecasts (ECMWF, 2002) with a horizontal resolution of 0.5° and 60 vertical levels. The fields used were the 6-h analyses (00:00, 06:00, 12:00 and 18:00 UT) together with extra 3-h forecasts (03:00, 09:00, 15:00 and 21 UT).

Since the number of sources is large while there is only a single receptor point, the Cabauw station, a receptor-oriented approach was adopted in order to make the whole procedure as efficient as possible. That is, for every measurement time at the receptor point ficticious particles of a tracer are released and followed backwards in time and each release is tracked separately.

FLEXPART was set up to generate 3-hourly tracer releases from the Cabauw station during the months of April and November 2007. Each release consisted of 10.000 particles from 20 m a.g.l.. Its set-up fixed the minimum mixing height allowable to 25 m a.g.l and used a time step of 600 s. A preliminary analysis showed that particles coming from North America could reach the Cabauw station within 15 days. Therefore, in order not to lose important contributions from far-away sources while constraining computational resources needed to keep all particle information, a maximum particle age of 15 days was allowed in the simulations. Being 15 days in the order of four times the radon half-life, the additional error introduced in the modelled concentrations through this time limitation is not significant. The output source-receptor relationships were produced for 10 m high 0.5° grid boxes with 3-h temporal resolution and for an outgrid domain comprising Europe (−10E/54E/30N/60N). The source-receptor relationships describe the sensitivity of each of the receptor concentration values to each of the possible source elements and are thus also called source-receptor sensitivity (SRS, Wotawa et al., 2003). In the present case without sinks, they correspond directly to residence times of the computational particles. For a detailed derivation and explanation of this concept and its implementation in a Lagrangian particle model run in backward mode, see Seibert and Frank (2004).

4.2. Calculation of modelled radon concentrations

The time series of radon concentration at the receptor point are calculated from the gridded source-receptor relationships obtained by FLEXPART and gridded radon flux data.

In the absence of radioactive decay, the concentration at a specific time would be obtained by multiplying the SRS values for all grid elements with the respective flux value and summing over all these contributions. Decay could have been calculated directly in FLEXPART, as a loss of mass of the fictitious particle according to the species half-life and the time it is being transported. However, and since decay acts just as a time-dependent exponential multiplicative factor to the SRS, it is computationally more efficient to introduce it as a correcting factor depending on the respective transport times, which are saved during the calculations, and the species half-life in a post-processing step. This was done here with the decay constant for ^{222}Rn. Thus, one single run could be used for different species without having to run the model again.

The simplest assumption for the radon source, which is widely used in atmospheric transport model validation studies (e.g. Conen, 2003), is to assume a constant radon flux over land, normally of 1 atom cm^{-2} s^{-1}, and 0 over water bodies or, at most, a variable source term depending only on the latitude. On the other hand, the most complicated source term would vary both in time and space according to meteorological conditions and soil properties. Since the latter is beyond the scope of this study, two basic source terms were considered in this work. Conen and Roberston (2002) used an average exhalation rate for the middle northern hemisphere ranging from 0.5 to 0.8 atoms cm^{-2} s^{-1} depending on latitude. In this work, the simple source term adopts an exhalation rate of 0.66 atoms cm^{-2} s^{-1} (50 Bq m^{-2} h^{-1}) radon flux above land and 0 over water bodies and was chosen after the work of Levin et al. (2002) who gave an average of 50–60 Bq m^{-2} h^{-1} for Heidelberg and the whole of western Europe. The second, more complex source term was based on the European radon flux map of Szegvary et al. (2007, 2008). More information on this map can be found at http://www.radon.unibas.ch/. It gives a spatially variable radon exhalation rate for Europe. Over water bodies the flux is zero and