

Radon measurements for atmospheric tracing

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1 Introduction

Radon is a useful tracer of atmospheric dynamics because of its simple source and sink. Radon is a noble gas; as such it does not react chemically with other atmospheric species; its sink is predominantly by radioactive decay, with a half-life of 3.81 days. The half life of radon is comparable with the chemical lifetimes of short-lived air pollutants such as NO_x , SO_2 , CO , O_3 . It is also comparable with the resident times of such important atmospheric constituents like water and aerosols. Many important dynamic atmospheric features occur on a time scale in the order of days.

Radon comes from land. The flux depends on the mineralogy and varies depending on changes in atmospheric pressure and soil moisture. The oceanic radon flux is on average at least 100 times smaller than that from land. This makes radon present in air samples at characteristic levels indicative of contact with land within the previous few weeks.

Radon flux from the soil to the lower atmosphere is always positive or zero as in water saturated soils or in presence of surface barriers like ice sheets. The flux from the soil to the atmosphere is maintained by a strong concentration gradient with the concentration above the ground about 1000 times lower than a few cm below the ground.

Once injected into the atmosphere radon can be used as a tracer on local, regional and global scales. Sub-grid mixing processes as well as long-range transport cause significant changes in radon concentrations which can be analysed in the context of local mixing schemes or regional/global circulation models.

Changes in the radon concentration are characteristic of the mixing and transport processes in the atmosphere. These changes can be measured continuously with adequate precision in ground stations using automated detectors. Vertical profiles up to 13 km have also been measured using grab samples collected from aircraft platforms; detectors have been developed for in situ airborne measurements within the boundary layer.

Thoron (radon-220) and its decay product lead-212 complement very well radon tracing capabilities on smaller time and spatial scales. Thoron (half-life 55.6s) is a gas and like radon, emanates from land. It is used as an effective tracer within a few meters above the surface. Airborne lead-212 (half-life 10.6h) is indicative of contact with local land.

This paper will describe recent developments in the measurement and application of atmospheric radon for baseline air pollution stations, the characterisation of local and regional transport processes, and the development and verification of global transport models.

Instrumentation

Radon concentration in marine air can be as low as a few radon atoms per litre of air. This makes the task of measuring radon levels with a time resolution matching prevailing weather conditions a demanding task. Radon measuring techniques and commercially available instrumentation developed for radiation protection are inadequate for the atmospheric research.

The volume of air which needs to be analysed for radon has to be large since the expected radon concentrations can be very low. For instance, in the air at Cape Grim that has not been in contact with a

land mass has only 10 to 100 mBq.m⁻³ radon, which corresponds to between 5 and 50 radon atoms per litre of air. In a hypothetical detector of with a volume of 1 m³, 100% counting efficiency for radon decays and no background, radon concentration at 10 mBq.m⁻³ level would result in 18 counts per half hour. In practice, counting efficiency is significantly lower than 50% and instrumental background is a serious problem for concentrations below 20 mBq.m⁻³.

ANSTO detectors for measurement of radon times series

In the last decade, ANSTO has developed and commissioned two types of radon detectors with the sensitivity matching baseline concentrations (Whittlestone and Zahorowski, 1995; Whittlestone and Zahorowski, 1998). Both designs are based on the two-filter method which relies on the sampled air being drawn continuously through one filter which removes all radon decay products, then through a delay chamber in which some new progeny are produced. Finally air passes through a second filter which collects the progeny at a rate proportional to the radon concentration. The larger the delay chamber volume, the more sensitive the detector will be since more progeny are produced in the chamber.

A major challenge in the design of high sensitive two filter detectors is to prevent the progeny from being plated out on the walls of the delay chamber. Most two-filter detectors use a high flow rate to ensure that the air passes from the inlet to the outlet filters in a time short compared to the mean plate-out time. In a detector with a volume of one or two cubic metres, the plate-out time is a few minutes. This implies that flow rates of about a cubic metre per minute would be necessary. As a result, pumping power and the second filter had to be unacceptably large.

One can solve this problem by injecting and maintaining a constant concentration of sub-micron particles in the delay chamber. This has been done in the first of the two designs (Whittlestone and Zahorowski, 1996). With particles in the chamber, the progeny become attached to the particles, which have a mean plate-out time of many hours in delay chambers of a volume greater than about 2 m³.

This solution delivers excellent results as far as lower limit of detection is concerned but it cannot work unattended at remote sites and on mobile platforms.

The second design called dual flow loop wire screen radon detector addresses the plate out problem by introducing a division of the air flow into the low rate external and high rate internal components and the use of the wire screens as the second filter (Whittlestone and Zahorowski, 1998). Hence, the function of supplying filtered air to the detector has been separated from the function of delivering air to the collecting wire screen used for collection of the radon progeny. The high diffusivity of radon progeny makes it possible to use a wire screen to remove the progeny with high efficiency and very low flow impedance.

The principle of operation is shown in Figure 1. The external flow takes the sampled air through the thoron delay volume that removes thoron and an inlet filter where existing radon progeny are filtered from the sample. The internal flow rate is high to maximise the likelihood of the progeny plating out on the wire mesh filter rather than on the walls of the chamber. The collected progeny decay by alpha decay which is detected in a scintillator/photomultiplier assembly.

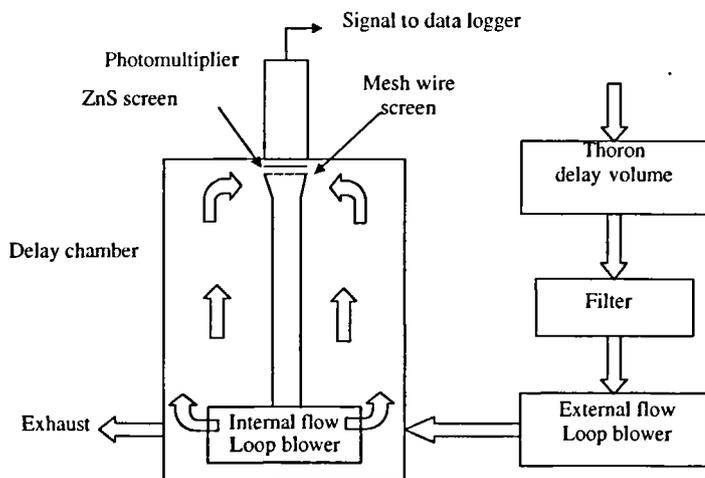


Figure 1
A schematic of the dual flow loop wire screen detector.

Dual flow loop wire screen radon detectors can measure radon concentration in air down to 5 mBq.m^{-3} with a time response of about 45 minutes to 50% of the maximum after a step increase in radon concentration. Implementations of the design have varied delay volumes (from 750 l to about 5,000 l) to match different sensitivity requirements. A “middle of the range” 1,500 l detector has a lower limit of detection equal to 20 mBq.m^{-3} with the internal flow rate at about 800 l.min^{-1} and the external sampling flow rate at 80 l.min^{-1} .

Deployment of ANSTO radon detectors

Figure 2 shows locations where ANSTO radon detectors have been deployed. Three detectors (at Cape Grim, Mauna Loa and Cape Point) take part in Global Atmospheric Watch, a World Meteorological Organisation network of about 20 world-wide research stations for measuring atmospheric composition and long-lived pollutants. The stations’ output underpins research into global climate change and stratosphere ozone depletion. Other locations point to installed detectors which serve ongoing air mass character-



Figure 2
Deployment of ANSTO's baseline detectors.
Locations in italics indicate a network of detectors
under construction.

isation and recording of long term radon time series for model development and verification. A new set of detectors has been proposed and is now being established in the international Aerosol Characterisation Experiment (ACE-Asia) in East Asia and the Northern Pacific.

Local and regional land surface/ atmosphere characteristics derived from radon

Both radon and thoron have been used as a tracer of near-surface mixing processes.

At the soil-air interface the typical radon profile changes dramatically: above the surface at 1m height the radon concentration is 10 Bq.m⁻³ and at 1000 m is half of that; below the surface at a depth of 5cm it is between 500-1000 Bq.m⁻³ and the equilibrium concentration of between 70,000-90,000 Bq.m⁻³ is reached at a depth of 2 m. For thoron, the concentration gradient is much steeper, owing to its much shorter half-life, with the depth and height at which half of the surface level are reached being 1.5 cm and 14 cm, respectively.

Local vertical mixing

Vertical distribution of a radioactive tracer concentration $C(z)$ can be estimated using a simplified vertical diffusion equation:

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial z} \left(k_z \frac{\partial C}{\partial z} \right) - \lambda \cdot C$$

where k_z is the vertical eddy diffusivity and λ is the decay constant of the tracer.

The vertical eddy diffusivities can be estimated from radon observations at different vertical scales within the boundary layer. Alternatively, modeled vertical eddy diffusivities can be validated by comparing calculated with observed profiles.

Lee and Larsen (1997) sampled radon profiles in the boundary layer as a function of altitude (up to the top of the layer) and time of day using an instrument designed for *in situ* aircraft measurements. The measured profiles were used to examine distributions of the profiles of the vertical eddy diffusivities $k_z(z,t)$. In particular, it was possible to validate modeled distributions of vertical eddy diffusivities by comparing measured radon profiles with simulated ones obtained by solving the above vertical diffusion model with the modeled profiles of the vertical eddy diffusivities.

In another study, radon was shown to provide an independent estimate of the trace gas exchange at the biosphere-atmosphere interface (Ussler *et al.*, 1993). Radon time series were recorded at different heights up to 18m of a micrometeorological tower located in a dry, open-canopy forest. Radon flux was estimated independently of the profile measurements. From the experimental data a time evolution of the forest canopy trace gas exchange coefficient was derived as well as the average total forest canopy resistance. The results compared well with those obtained by eddy correlation techniques. The study demonstrated that automated radon measurements of concentrations in air can be successfully employed in remote locations where daily maintenance required to standard micrometeorological equipment (such as a sonic anemometers) is not possible.

Butterweck *et al.* (1994) characterised vertical diffusion in the near-surface atmosphere (0-5m) from continuous measurement of radon and thoron concentrations within and above a wheat field over the complete vegetation cycle of the crop. They used the data to calculate vertical eddy diffusivities for two horizontal layers: one within the crop (0.14-1.5 m) and the other above the crop (1.5-5 m). Mean vertical diffusivities for three weather conditions and two vertical temperature gradient intervals were then calculated. When compared with results obtained from meteorological data for the above crop layer (1.4-5 m) good agreement was obtained. It was claimed that the tracer method was superior especially inside the crop where

meteorological methods are not effective. Thoron proved to be complementary to radon in the lower interval within the crop where the vertical change in radon concentration was too small to derive the gradient with a satisfactory precision.

Regional trace gas emissions

Trace gas emissions originating from large land areas can be estimated using radon as a marker for emission from soil. This has been demonstrated by Wilson *et al.* (1997). They used a 9 year database of nitrous oxide and radon recorded at Cape Grim to assess the origin of N₂O by examining the cross covariance between N₂O and radon signals originated from land surface. A clear maximum in the cross covariance very close to the zero time delay indicated the same source regions of the two gases. With the common origin of the investigated radon and N₂O signals established, they calculated the average flux of N₂O by assuming that the ratio of fluxes is directly proportional to the average ratio of the individual measurements of radon and N₂O from land:

$$\overline{f_{N_2O}} = \overline{f_{Rn}} \cdot \left(\frac{\overline{N_2O}}{\overline{Rn}} \right) \cdot c$$

where c is a unit-dependent conversion factor. The precision of such estimates benefits from precision and accuracy of the radon concentration measurements. Another advantage is that such estimates are averaged over hundred of square km of land. Seasonal and inter-annual variations were also retrieved from the data with the authors showing that the nitrous oxide flux was higher for rainfall periods. Clear peaks in angular radon and N₂O signals made it possible to obtain independent flux estimates from two main land masses.

The uncertainties of the above method can be made smaller with a better knowledge of radon fluxes in regional areas. The study demonstrated that the method can be used as an independent method of evaluation of regional trace gases emitted from soil.

Radon time series at baseline stations

Baseline selection criteria

A world-wide network of baseline air pollution stations has been established to monitor long term trends in concentrations of climate-sensitive trace gases in the atmosphere. Above all, there is a need to characterise the origin of the air sample, or, more specifically, to develop baseline selection criteria. Air samples showing no recent contact with land (and hence with anthropogenic pollution) are termed “baseline”. No generally accepted operational definition of baseline air has been formulated. Instead, site- and species-dependent baseline selection criteria have been proposed. For instance, Zahorowski *et al.* (1996) showed that in the case of ozone at Cape Grim a baseline criterion using exclusively radon concentrations rather than wind speed and direction selected a more consistent ozone subset for the baseline sector. In another study of baseline criteria, Gras and Whittlestone (1992) compared condensation nuclei (CN) and the radon concentration time series measured at two baseline stations (Cape Grim, Tasmania and Mauna Loa Observatory, Hawaii). They concluded that addition of CN and radon concentration to meteorological criteria significantly improved the objectivity of baseline selection. It was demonstrated that a combination of wind direction and CN concentration will indicate pollution from sources within a few km. At longer distances, the radon is a better indicator of fetch over land. Radon will be indicative of contact with land at distances greater than a thousand km, a situation when wind direction and CN are poor indicators.

Radon time series measured over a 10 day period in 1999 at Cape Grim is shown in Figure 3. Wind speed and sector indicators are shown at the top. The sector indicators are designated by the letters T (Tasmania), M (Mainland) and B (Baseline). All data points are hourly averages. As the source of radon changes, concentrations change from a few tens of $\text{mBq}\cdot\text{m}^{-3}$ to more than $1 \text{ Bq}\cdot\text{m}^{-3}$. These changes can occur within a few hours. This is well illustrated by two

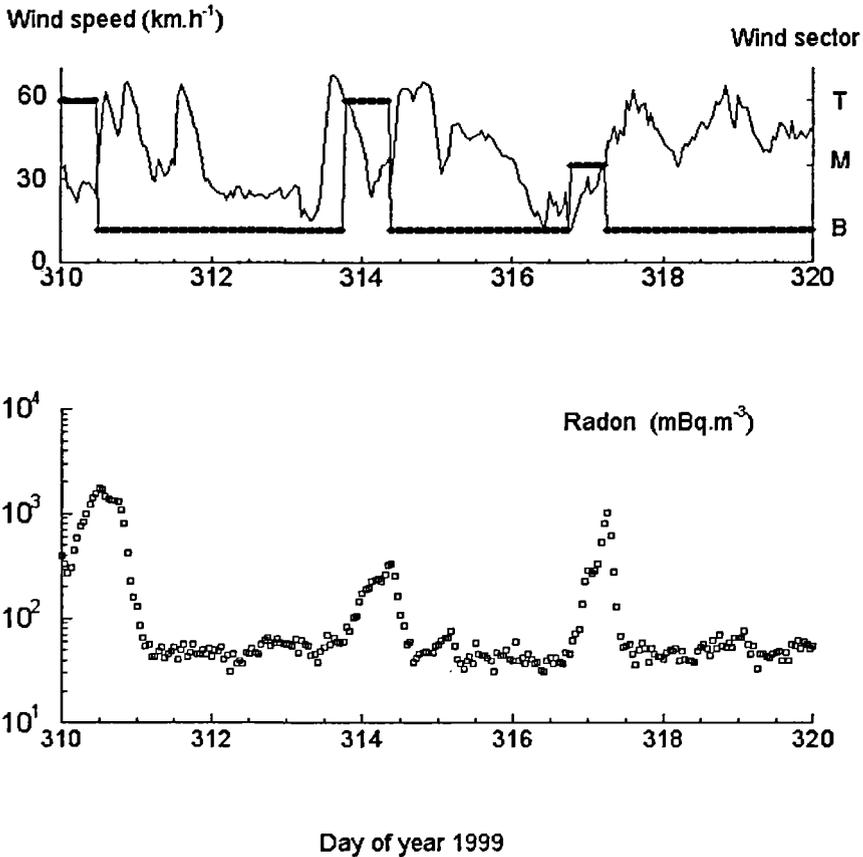


Figure 3
 An example of hourly radon observations recorded at Cape Grim (see text for explanations).

events: in a relatively brief period, the change in wind direction from Baseline to Tasmanian (day 313/314) and Baseline to Mainland (day 316/317) resulted in increased radon concentrations of up to one order of magnitude. Both the radon range and rate of change impose demanding conditions on detection systems.

Local influence

An important issue in baseline observations is the problem of local influence on the chemical/elemental composition of air samples. The local influence is site-specific, depending on a combination of the location of the station, synoptic meteorology and availability and spatial distribution of local sources of radon and thoron. The problem of tracking and separation of the local signal is difficult to solve for continental sites. At coastal and island sites, lead-212 has been used in combination with radon to separate local from distant events (Polian *et al.*, 1986; Whittlestone *et al.*, 1996a). A local knowledge of radon and thoron fluxes in the vicinity of a baseline station can be important in this context. For instance, high sensitivity thoron flux measurements were required for an evaluation of the thoron source term from barren lava around the Mauna Loa Observatory in Hawaii (Whittlestone *et al.*, 1996b).

The problem of local influence also affects modelers who view it as a sub-grid source problem beyond their control. Mahowald *et al.* (1997) and Dentener *et al.* (1999) reported that in order to make meaningful model-observation comparisons they had to filter radon time series to remove the local influence. This needed to be done for Kerguelen and Crozet Is sites where about 13% of the radon data were rejected using simultaneous measurement of lead-212. In case of Mauna Loa Observatory data where the supporting in lead-212 data were not available for the investigated period only night data (12 pm till 7 am) were used to exclude daytime up slope conditions bringing local radon from the island (Dentener *et al.*, 1999). At some sites (Bermuda and Cape Grim) applying a wind sector criterion is sufficient to remove the locally influence from the data (Mahowald *et al.*, 1997 and Dentener *et al.*, 1999). However, concurrent measurements of lead-212 is always desirable: first, for an independent experimental verification of any data rejection scheme, second, for preservation of as much of the data as possible because the wind selection criterion is not precise and hence its effective application leads to rejection of some unaffected data.

Simulation of radon with global circulation models

The last decade has witnessed the first serious attempts to model radon concentrations on the global and regional scale and to compare the modeled results with observations. The purpose of radon simulations varied; some aimed at the development or validation of sub-grid mixing schemes and indication of regions associated with largest uncertainties (e.g. Jacob and Prather, 1990; Stockwell *et al.*, 1998), others at the comparison of models (e.g. Genthon and Armengaud, 1995; Jacob *et al.*, 1997) or the comparison of different meteorological input data sets (e.g. Mahowald *et al.*, 1997). More generally, a better understanding of all key atmospheric features which control the transport, mixing and distribution of radon has been sought by detailed comparisons of the modeled radon time series and vertical profiles with best available radon data sets (e.g. Mahowald *et al.*, 1997; Dentener *et al.*, 1999; Stockwell *et al.*, 1998).

The most comprehensive comparison between modeled and observed vertical radon profiles in the boundary layer and the troposphere covering the range 0-12 km was published by Stockwell *et al.* (1998). They parameterised radon emissions in a global off-line three-dimensional chemical transport model forced using meteorological analyses. Sensitivity analysis was performed using two horizontal resolutions ($2.8^\circ \times 2.8^\circ$ and $7.5^\circ \times 7.5^\circ$). The effect of implementing vertical diffusion and moist convection was also tested. The inclusion of both vertical diffusion and moist convection as well as the higher resolution was necessary for a realistic simulation of radon. An analysis of model-observation correlations revealed that the modeled radon concentrations higher than those observed very close to the surface and generally much lower than those observed in the planetary boundary layer/lower troposphere. This was attributed to insufficient vertical mixing. An inclusion of a non-local vertical diffusion scheme was postulated, which was expected to give a better mixed planetary boundary layer.

Most recent extensive comparison between the simulated and observed radon time series recorded at a number of ground stations was published by Dentener *et al.* (1999). They utilised the data from 8 ground stations: 2 continental, 2 coastal under continental influence and 4 remote stations. Some vertical radon profiles were also compared with the model results. Similarly to Stockwell *et al.* (1998) and Mahowald *et al.* (1997) they constrained the two models used in the comparison with meteorological. The quantitative comparison was made using monthly means and correlation coefficients. To eliminate variations on time and spacial scales which cannot be reproduced by the models the monthly correlation coefficients were defined from daily averaged measurements and model results. Overall agreement was good for continental stations and coastal stations where correlation coefficients 0.6–0.8 were obtained. The highest monthly correlation coefficients of around 0.8 were obtained for Cape Grim, which was attributed to the fact that the model was constrained by observations performed on and near the Australian continent. Higher uncertainties in the meteorological fields constraining the models were indicated as a reason for lower correlation coefficient (0.5–0.6) for remote stations (especially for Crozet, Kerguelen, and Amsterdam Islands in the Indian Ocean). A similar problem with the same remote sites (and also with the Macquarie Is site) was noted by Mahowald *et al.* (1997) who simulated successfully observed pollution events at Cape Grim but had difficulty to model events farther from continental source regions.

Radon source strength

Several processes may influence radon emissions to the atmosphere (e.g. Nazaroff, 1992; Holford *et al.*, 1993). These include the abundance of the parent radium-226 and soil properties with diffusion coefficient being the most important factor. The diffusion coefficient strongly depends on soil moisture, with the coefficient values ranging from 3×10^{-6} to $10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$ for a typical dry soil and fully saturated soils, respectively. Although the predominant transport

mechanism responsible for delivering radon to the surface is by molecular diffusion, the flux is also sensitive to changes in atmospheric pressure. Soil freezing also affect the radon flux (Dörr and Münnich, 1990). The flux to the atmosphere is inhibited by snow and ice covers.

Even with this variability, there can be large areas where there is relative uniformity of radon emanation rate. However, a task of constructing radon emission maps is far from simple.

Spot measurement of radon flux

A radon flux survey aimed at covering large, frequently remote areas needs specific instrumentation. The main requirement, besides the lower limit of detection matching expected flux levels, is an adequate sampling frequency. ANSTO has developed a fast, general purpose emanometer for measuring radon and thoron fluxes (Zahorowski and Whittlestone, 1996). Figure 4 shows the essential features of the emanometer. The principle of operation of the instrument is as follows. At the start of the measurement the accumulation chamber (A) is placed over the ground. Air is drawn from the cham-

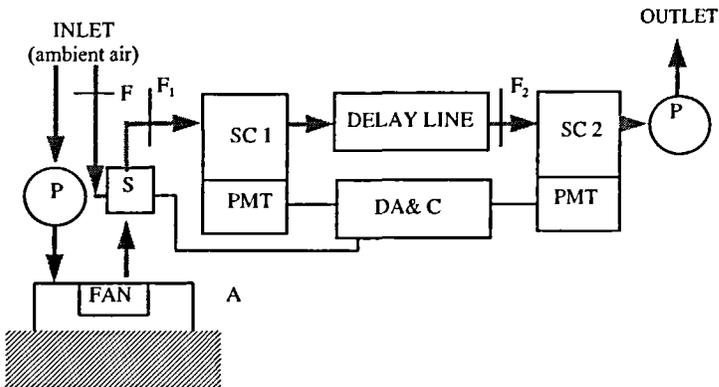


Figure 4

A simplified diagram of the fast emanometer.

A, accumulator chamber; DA&C, data acquisition and control;

F, filter; PMT, photomultiplier tubes; SC, scintillation cells.

ber into the first scintillation cell (SC1) which records counts from both radon and thoron. From here, the air passes via a 6 minute delay tube in which thoron decays, into the second cell (SC2) which records radon counts. For a one hour measurement the lower limit of detection is $1 \text{ mBq}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ for radon and $20 \text{ mBq}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ for thoron. Lower limits can be achieved by counting for longer. One can set up the instrument for a semi-continuous flux monitoring by fitting an automatically operated ventilation lid to ensure a quick ventilation of the accumulator. Such a system is capable of unattended recording of hourly radon and flux measurements for many days.

It has been demonstrated (Whittlestone *et al.*, 1996b) that a high sensitivity thoron emanometer can be assembled in situations requiring thoron flux estimates down to $1 \text{ mBq}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$. Due to thoron short half-life such an instrument has to operate at a high flow rate and mesh filters similar to the ones used in dual flow loop radon detectors have to be fitted for lead-212 collection.

Regional radon fluxes

Spot flux measurements are too resource intensive for construction of regional or continental radon emission maps.

Some large area estimates have been done in the past. Apart from some early coarse estimates (Turekian *et al.*, 1977) and some limited area studies, no large regional or continental emission maps was published. Nevertheless, a commonly held assumption is that the radon flux is quite uniform and equal to $1 \text{ atom}\cdot\text{cm}^{-2}\cdot\text{s}^{-1}$.

A map for the Australian continent has been constructed based on radon emissions combined with data from airborne gamma survey (Zahorowski and Whittlestone, 1997). In a first step maps were selected covering areas of $100\times 120 \text{ km}$ for which both radon fluxes and airborne gamma data were available. An average gamma count for the area was found and converted to a map average radon flux. The maps were then matched as well as possible to the $5^{\circ}\times 4^{\circ}$ grid, and grid averages obtained.

The results of the flux evaluation procedure is shown in Figure 5. The top number in a cell is derived from spot radon measurements and the bottom from airborne gamma data. The two values, when

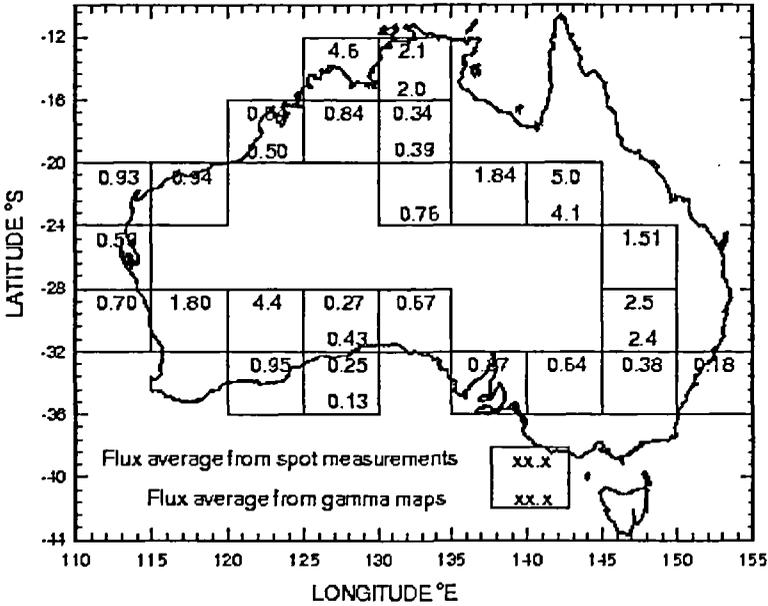


Figure 5
Average radon fluxes (in $\text{atom}\cdot\text{cm}^2\cdot\text{s}^{-1}$) from 50 x 40 grid boxes.
1 $\text{atom}\cdot\text{cm}^2\cdot\text{s}^{-1}$ corresponds to about 21 $\text{mBq}\cdot\text{m}^2\cdot\text{s}^{-1}$.

both are available for a cell, agree quite well. It is also clear that the 1 $\text{atom}\cdot\text{cm}^2\cdot\text{s}^{-1}$ assumption is very inaccurate even on the $5^\circ\times 4^\circ$ grid basis. From the southern part of Australia, the average radon emission is 1.1 $\text{atom}\cdot\text{cm}^2\cdot\text{s}^{-1}$, but for the northern part of Australia it is 1.8 $\text{atom}\cdot\text{cm}^2\cdot\text{s}^{-1}$. The whole data set gives 1.4 $\text{atom}\cdot\text{cm}^2\cdot\text{s}^{-1}$.

Radon flux in global transport models

As radon time series become more frequently used for development and validation of global transport models, area-averaged radon fluxes are expected to attract more attention. In a recognition of the problem, the first radon emission scenario was recommended by the World Climate Research Programme (WCRP) in 1993. The only physical process taken into account, apart from

obvious effects related to ice covers, was an observation that radon flux depends on soil freezing. Global annual radon emission to the atmosphere was constrained to 72 mol of radon. The scenario was subsequently used as a basis for a comparison of 20 global models (Jacob *et al.*, 1997).

The WCR scenario has been useful as it allowed for intercomparison between results of simulations of radon concentrations obtained by different models. Most recent studies which adopted similar emission scenario (e.g. Mahowald *et al.*, 1997; Stockwell *et al.*, 1998; Dentener *et al.*, 1999) raised the problem of some obvious inadequacies of the scenario, which does not even allow for different source strengths on continental basis.

Some researchers attempted a simple sensitivity analysis involving the radon emissions. For instance, Stockwell *et al.* (1998) identified origins of radon in different altitude bands by varying continental radon source strengths within the prescribed global annual maximum emission value. Because of a constant emission source in their model the authors did not expect perfect agreement between the observed and the calculated values even if all transport processes were modeled perfectly. Their results suggested that the WCRP scenario might seriously underestimate the Asian radon source. Dentener *et al.* (1999) arrived at a similar conclusion regarding the Asian source while discussing reasons for large discrepancies between modeled and observed radon concentrations recorded at Mauna Loa Observatory.

Dentener *et al.* (1999) were also the first to address the source problem in a systematic way by parameterising the radon emission in their global model using a classification based on soil types (Dörr and Münnich, 1990). Although the flux used in the scheme varied significantly, from 0.4 to 1.5 atoms.cm⁻².s⁻¹, the measure did not lead to an improvement in the agreement between the modeled and observed data. Similar lack of response was also noted when the sensitivity of the model was tested to a reduced (3 times) flux due to snow cover.

Some modelers varied radon emissions with atmospheric pressure (e.g. Jacob and Prather, 1990; Lin *et al.*, 1996) but no significant improvement to model-observations comparison was reported.

Conclusions

The accuracy and precision of radon instrumentation is now more than adequate to provide the radon time series required for comparing with the simulated ones obtained from regional and global circulation models. The measured high quality radon time series data from selected locations will continue to contribute to the development and validation of the models.

Only a few limited sensitivity analyses involving continental radon sources have been performed. There are no systematic sensitivity analysis that consider variation of grid-averaged fluxes within experimentally derived constrains is required. Furthermore, circulation model performance strongly depends on the quality of the constraining meteorological data. Model-observation correlation coefficients fro radon levels as low as 0.5 and as high as 0.8 have been calculated depending solely on the quality of the meteorological input. Such a strong dependence of model performance on meteorological data indicates that radon is a sensitive tracer for model evaluation.

The present World Climate Research radon emission scenario needs to be improved by incorporating parameterisations of important factors controlling radon emanation like soil moisture. Improved area-averaged radon fluxes can be constructed from the geographical abundance of radium-226 (through airborne or car-borne radiometrics or geological data) with spot measurements providing a means to verify area emissions.

More observation stations will help to constrain the radon emissions. More regional stations will provide additional opportunities for pseudo-Lagrangian experiments and in new possibilities for model development and data interpretation.

Radon is a valuable tool for investigating atmospheric transport and mixing on different horizontal and vertical scales. In the last decade significant advances have been made both in radon instrumentation and simulation of radon concentration in the atmosphere using global circulation models.

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