1. Introduction

Radon-222 (radon) is the most frequently used naturally occurring radioactive tracer of atmospheric dynamics on local, regional, and hemispheric scales [1]. This is due to a number of useful radon properties, which include: (a) a terrestrial flux that can be assumed to be horizontally homogenous on regional scales and approximated by a global average of $22 \, \text{mBq} \, \text{m}^{-2} \, \text{s}^{-1}$; (b) a very low flux from oceans (about three orders of magnitude lower than from land); (c) its non-reactive nature, with a simple, well defined sink by radioactive decay alone; and (d) a half-life (3.8 days) that is long enough compared to the turbulent mixing time in the Atmospheric Boundary Layer (ABL) but short enough to ensure a significant gradient across the ABL-troposphere interface and to minimize horizontal variations in concentration above the ABL.

Radon’s tracing capacity has been applied to (a) selecting the least perturbed marine air masses at baseline stations [2]; (b) defining footprints of ground-based sites [3]; (c) calibrating, on regional scales, terrestrial emissions of greenhouse gases [4]; (d) evaluating the performance of regional and global atmospheric transport models [1]; and (e) quantifying mixing within the ABL [5].

ANSTO has developed advanced instruments and methods for radon observations on the ground, at sea, and from airborne platforms. Such measurements are demanding as radon concentrations in air may vary from 10 to $>10,000 \, \text{mBq} \, \text{m}^{-3}$, with the lower limit corresponding to 5 radon atoms per litre of air, a very low concentration by any standard. The time resolution of such instruments, necessitated by the processes of interest, is also demanding. While it is sufficient for ground-based measurements to resolve diurnal and synoptic time scales for horizontal transport, sampling times shorter than 10-15 min are essential to conduct vertical profiles. Also, as a rule, ground- and sea-based instruments have to run unattended for prolonged periods, and airborne samplers are tightly constrained by weight and size, have to meet strict aviation standards and be simple enough to operate easily in flight.

This paper illustrates how radon has been applied as a tracer on regional scales in the East Asia/Northern Pacific region. It also reports on the most recent ANSTO developments in the measurement of vertical radon profiles for the characterisation of mixing within the ABL and exchange with the troposphere, from ground-based and airborne observations. Together with supporting meteorological measurements, these systems provide new information regarding mixing and exchange processes across the atmospheric surface layer, the nocturnal stable layer, and the convective mixed layer, and can be used in the evaluation of mixing schemes for numerical models.

2. Tracing atmospheric transport on regional scales

Examples presented here are derived from a five year experiment in which hourly measurements of atmospheric radon concentration were made at three ground-based coastal stations in East Asia (Hok Tsui, Hong Kong Island, China; Gosan, Jeju Island, Korea; Sado, Sado Island, Japan). Results are compared with data from Mauna Loa Observatory (MLO), Hawaii, gathered during the same period. The experiment was part of the most recent Aerosol Characterization Experiment in East Asia (ACE-Asia), and aimed to comprehensively characterise air masses associated with East Asian continental outflow events to the Pacific. Site locations were chosen to span the latitudinal band within which most of the low level Asian continental outflow events to the Pacific occur (20-40°N), and coincided with ACE-Asia network sites and locations where campaign-style observations were made. The data set is unique with respect to its spatial and temporal coverage, and well suited to air mass characterisation of a region that is a globally significant source of natural and anthropogenic pollution.
Dual-flow loop, two-filter detectors, developed at ANSTO [6], were used at each of the sites. The principle of operation of these detectors is illustrated in Figure 1a. The external flow loop draws sample air through the detector at a low flow rate. Between the inlet and the first filter the sample is delayed to reduce radon-220 (thoron) content to negligible levels. The first filter removes aerosols and ambient radon progeny. The air then enters the radon delay volume where it is circulated at a high flow rate by the internal flow loop, passing repeatedly through the second filter. A fraction of the new radon progeny generated in the radon delay volume is captured on the second filter. These short-lived, alpha emitting radon progeny are counted using an assembly consisting of a ZnS scintillator, photomultiplier tube and counting electronics. Each of the three detectors deployed in East Asia had a volume of 750L and sampled at a flow rate of ~40L min⁻¹; the volume and the flow rate of the MLO detector were 1,500L and 80L min⁻¹, respectively. The response time of the detectors, defined as the time required to reach 50% of the maximum count rate after a step change in radon, was about 45 minutes.

Atmospheric species of terrestrial origin, including aerosols and climatically sensitive gases, are usually monitored at ground stations where air masses are sampled and their composition determined. That composition will depend upon the stations’ footprint. A station’s footprint is the region most likely to contribute to the atmospheric composition of air masses arriving at the station. Back trajectories, derived from assimilated meteorological data collected around the world, approximate paths that air parcels follow on their way to a station (Figure 1b). However, back trajectories alone can not quantify the degree of interaction between an air parcel and the Earth’s surface, which largely determines constituent concentrations. Interaction with the Earth’s surface can only be estimated by concurrent measurements of a suitable tracer. Because of its unique properties, radon is the most suitable tracer in this context.
Since many factors can affect species concentrations, the best way to analyse the variability in a station’s footprint is by determining footprints corresponding to extremes of land contact. Previously this has been achieved by analyzing back trajectories corresponding to high and low hourly radon concentrations over a period of interest [3]. A separate consideration is the length of back trajectories used. Under conditions of a spatially homogenous radon flux and entrainment velocity, approximately 80% of the radon observed in an air mass is contributed over the last 10 days of its journey. The following examples of back trajectories were calculated using the NOAA Air Resources Laboratory (ARL) HYSPLIT 4 package [7].

The first example of radon-derived footprints from Hok Tsui in winter (Figure 2) shows that the technique can identify subtle changes in fetch. The left and right panels show trajectories corresponding to hourly radon concentrations higher (lower) than the seasonal 90th (10th) percentile values, respectively. Although the corresponding air masses approach Hok Tsui from a similar direction (dotted lines) their composition is very different. A high radon concentration indicates recent land contact whereas a low concentration most likely indicates recent marine fetch. In each case the last 3-5 days of the Hok Tsui footprint is well defined, indicating an overall coastal fetch in winter, but with distinct on-shore and off-shore components.

The second example illustrates the capability of the above method to obtain footprints characterised by strong air mass interaction with continental emissions. In Figure 3a schematic footprints are superimposed on the corresponding mean trajectories for winter (solid lines) and spring (dotted lines). The result clearly indicates that, although Hok Tsui and Gosan are located within the 20-40° latitudinal band, their recent fetch...
is limited coastal regions. Furthermore, their footprints extend to the interior of the continent above 50º north. As such, they offer limited information about inland continental emissions within the ACE-Asia study domain (20-40ºN). As indicated in blue in Figure 3a, however, such information can be derived from the MLO observations. Trajectories of high radon events in spring corresponding to the MLO footprints (Figure 4), demonstrate that MLO high radon events can be separated into those originating from the 20-30º and 30-40º latitudinal bands, thus enabling separate probing of these two important continental bands.

An interesting aspect of the East Asian network measurements is that their radon background varies strongly with both season and latitude (Figure 3b). Here the background is defined by the weekly 10th percentile concentration to reduce the influence of synoptic scale events. Only in summer (weeks 25-30), when regional flow is onshore, does the background approach concentrations associated with oceanic radon emissions. In other seasons, when offshore flow dominates, the background reflects seasonal and latitudinal dependence.

3. Tracing local air movement on diurnal time scales

Vertical radon profiles in the lower atmosphere can be used to determine the degree of mixing in the ABL and exchanges with the surface and the free atmosphere. The use of radon in ABL process and model evaluation studies therefore carries the potential to significantly reduce systematic errors in the reproduction of diurnal and seasonal cycles in weather and climate prediction models on a range of scales.

In the last two years ANSTO has developed and implemented a measurement system for obtaining accurate radon concentration profiles through the ABL and extending into the free troposphere. The adopted approach involves a combination of ground-based continuous hourly observations of radon concentration gradients and aircraft-based measurements of radon profiles.

The ground-based observations rely on taking air samples using inlet lines mounted at two heights, on two towers: one tower covering the first 50m above the ground; the other extending to 200m. Each of the towers is equipped with two radon detectors of the type discussed in the previous section. All four instruments have identical technical parameters (1,500 L delay volume; 80 L min⁻¹ inlet flow rate; 30-40 mBq m⁻³ lower limit of detection). At present, inlets are mounted and 2 and 50m, and 20 and 200m, on the 50m and 200m towers, respectively. The shorter tower is located at ANSTO and became operational in mid 2005. The taller tower is located at the Cabauw Experimental Site for Atmospheric Research (CESAR) in the Netherlands and became operational in March 2006. The two-tower system offers a unique opportunity for high precision measurements of diurnal and seasonal cycles of vertical mixing using naturally occurring radioactivity.

Radon time series from 2 and 50m on the ANSTO 50m tower at are compared in Figure 4 for the whole month of September 2005 (top panel) and for a 10-day subset (bottom panel). Close to the surface, radon concentrations build up at night with the formation of the nocturnal stable boundary layer (SBL). On some
nights, indicated by arrows, the radon data indicates that the local SBL height is below 50m and the surface is thermodynamically decoupled from the air aloft.

Figure 5  Hourly radon recorded at 2 and 50m on the 50m ANSTO tower for 4 days in September 2005 (top panel) as well as temperature, wind speed and solar radiation.

A radon sampler was developed at ANSTO after completion of successful tests of two prototypes. The new radon sampler consists of 6 charcoal traps (Figure 6a). Each trap has a volume of approximately 300 cm$^3$, holding 110g of charcoal. During a sampling event a known volume of air passes through the traps using a 24V dual membrane pump. Samples are collected for periods ranging from 5 to 15min at a flow rate of about 65 L min$^{-1}$ STP. Instrument dimensions, weight and power consumption (760 x 300 x 200 mm, 20kg, and 240W, respectively) make it possible to fit in the wing-pod of a motor-glider (Figure 6a). Radon collected in the traps is transferred to evacuated Lucas cells in a two step process in a laboratory-based radon rig. Estimated counting errors as a function of radon concentration, calculated for three counting periods, are shown in Figure 6b. The estimate points to the possibility of getting the lower limit of detection down to 10 mBq m$^{-3}$. The actual lower limit of detection depends critically on the concentration of radium-226 in charcoal: a typical commercial charcoal batch results in a peak-to-background ratio of 0.2 at the 10 mBq m$^{-3}$ radon concentration level – the same ratio for the counting system alone is about 6.

Together with the aforementioned two-tower system, the capability of the new system to record vertical radon concentration profiles covers the boundary layer and lower troposphere up to 3000m above the ground.
4. Conclusions

A comprehensive system of measurements, including unique instrumentation and methods, has been established at ANSTO to apply radon tracing capacities for the current problems in atmospheric research. On a regional scale, these measurements have provided accurate footprint description for important coastal ground stations in East Asia and the Northern Pacific. On a local scale, the newly developed airborne measurements offer potential for new insights into mixing and exchange processes across the Atmospheric Boundary Layer.

5. References


