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APPLICATION OF SENSITIVE AND SUPERSENSITIVE RADON DETECTORS FOR RADON FLUX DENSITY AND RADON CONCENTRATION IN ENVIRONMENTAL MONITORING

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Abstract

The paper presents a review of principles and operational parameters of the latest instrumental development in sensitive and high sensitive radon detectors at Australian Nuclear Science and Technology Organisation (ANSTO). The focus is on advances in measurement technology of radon concentration in air and radon flux density. Two areas in which ANSTO is actively involved are discussed. The first area concerns radon in air monitoring at Cape Grim Baseline Air Pollution Station. Results recorded at the Station with a supersensitive radon detector characterised by lower limit of detection down to few mBq m^{-3} with time resolution better than 90 minutes are presented to illustrate importance of the technique in global monitoring of airborne pollution. The second area concerns estimates of radon and thoron fluxes from large geographical areas. This is illustrated by results obtained during an Australia-wide survey of radon fluxes and from thoron flux measurements around the Mauna Loa Observatory in Hawaii. The radon flux estimates from Australia come from a coarse net of spot measurements combined with data from aerial gamma surveys. It is argued that as radon global flux and air concentration estimates improve, the data will provide progressively more stringent tests of global air transport models.

1. INTRODUCTION

It is now widely accepted that anthropogenic atmospheric pollution is strongly linked with global climate change. There are limits to the amount of pollution which can be injected into the atmosphere without causing climate change or serious deterioration of the human environment. It is therefore very important to know how much of what gases can be safely injected into the atmosphere. A precise answer to this question is of major economic and political significance. An essential component of the answer comes from air transport modelling. With modern computers one can take any available pressure, temperature, wind speed and direction, and other data to estimate air transport with a horizontal resolution of the order of a hundred kilometres, and vertical resolution better than a thousand meters.

Despite these achievements the problem of the models' accuracy and precision needs to be addressed at each new step in improvement. Inevitably, a question arises of an independent validation of the atmospheric transport models.

One way to monitor polluting gases on the global scale is to establish a set of baseline air monitoring stations. This is done to monitor pollution far away from its source. In such a way background changes in concentration of polluting gases are recorded and the data can be compared with predictions calculated by models. Transport and dispersion components of global models can be verified using suitable tracers. Radon is a naturally available tracer which can be successfully used for verification of transport models. Radon can also be used in establishing the baseline condition. Measurements of radon concentration in baseline air impose very demanding requirements on instrumentation. A description of the latest in the series in radon detectors for baseline research deployed by ANSTO will be given in Section 2.

Section 3 will elaborate on two related problems concerning the mapping of the distant and local radon source. The distant radon source term is a crucial parameter in global transport models.

In this context, it will be demonstrated how one can use radon flux measurements and radiometric data to evaluate average fluxes from large continental areas. A detailed mapping of the local radon and thoron source term is valuable as a quantitative measure in evaluation of both local and distant air analysed at a baseline air monitoring station.

2. RADON CONCENTRATION IN BASELINE AIR MONITORING STATIONS

Figure 1 shows locations where ANSTO radon detectors have been deployed. At the moment ANSTO records radon in three baseline stations. The first one is located at Cape Grim in the north-western Tasmania, the second station is at Macquarie Island, and the third radon detector is installed at the Mauna Loa Observatory (MLO) in Hawaii. The map shows other places where ANSTO supplied radon detectors and run radon projects together with other organizations.

2.1. Validation of global atmospheric transport models based on radon concentration measurement

Imagine a distant continent which is a source of pollution (and also a source of radon) and a small island away from major pollution sources where a baseline pollution station is located (Fig. 2). Once radon is injected to the atmosphere on the continent it might travel over the ocean and be detected on the island. While over the ocean, there will be no further radon injection since the flux from the ocean is about two orders of magnitude smaller than that from land. Because of radon half-life there is no radon build-up in the atmosphere. On the other hand, the half-life is long enough to allow radon to move over large distances. Also, being a noble gas, radon does not interact chemically with other species. These features make radon a very good tracer [1]. In fact, there is no other tracer as good as radon for global transport on a time scale of a week, except for gases released in occasional special events such a volcanic eruption.

Overall characteristics of radon concentrations in baseline air, such as the range and rate of change following the change in meteorological conditions do not differ much in baseline air from station to station. Results will be given of observations at Cape Grim baseline pollution station. The definitions of wind sectors at the station are given in Fig. 3: the baseline sector with westerly winds coming from the Southern Ocean, the Tasmanian Sector covering the island of Tasmania and the Australian Sector where most pollution comes from. Melbourne is about 500 km to the north.

A typical weekly data sheet is shown in Fig. 4. Important meteorological data are at the top. They include wind speed and sector. The sector indicators are given by letters T, A and B, for Tasmanian, Australian (continental) and Tasmanian wind sectors, respectively. Radon concentrations are shown at the bottom. All data points are hourly averages. Such weekly data sheets are used for quality control. Ozone and condensation nuclei (CN) concentrations are usually included since they provide further useful information. There are two observations to be made here. First, the radon range is from few tens of mBq m^{-3} to few Bq m^{-3}. Second, the time scale at which meteorological conditions can cause significant changes in radon concentration is an hour. This is well illustrated in an event at the beginning of day 106, when a relatively brief period of the change in wind direction from Baseline to Tasmanian resulted in about 5 times higher radon concentrations. Both the radon range and expected rate of change impose demanding conditions on radon detection.

2.2. Radon as a baseline criterion

Baseline atmospheric observations aim at monitoring long term trends in concentrations of trace constituents of the atmosphere. It is very important to distinguish between air masses which are

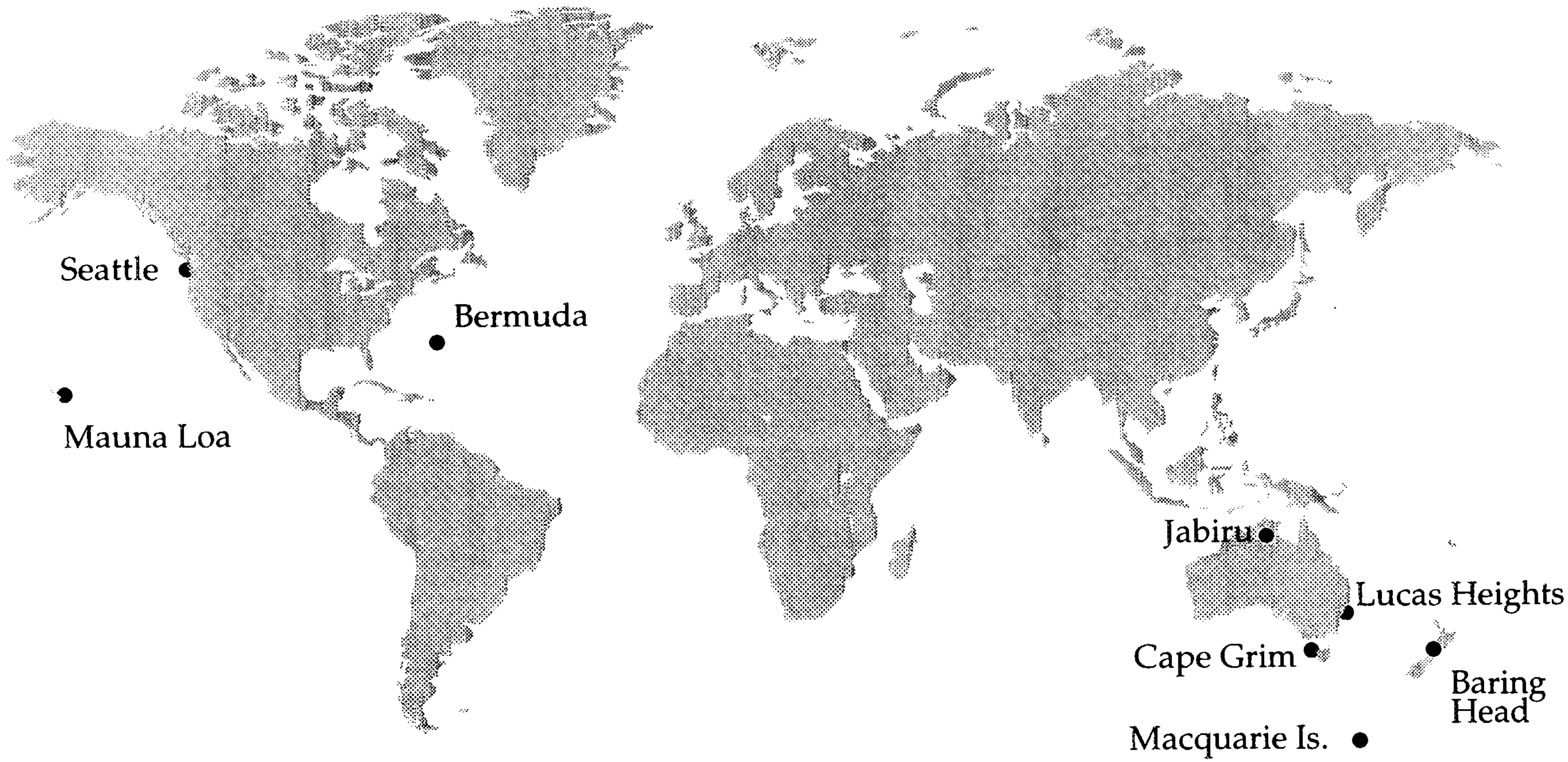

FIG. 1. Deployment of ANSTO's baseline radon detectors.

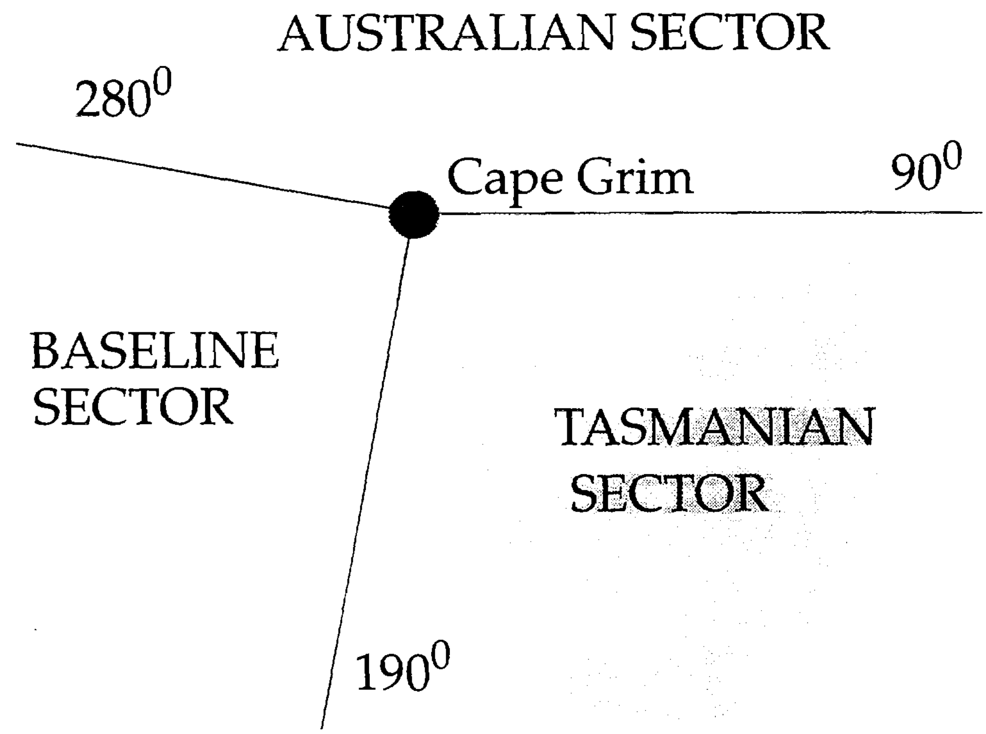

FIG. 2. Radon in boundary layer, sources and sinks.

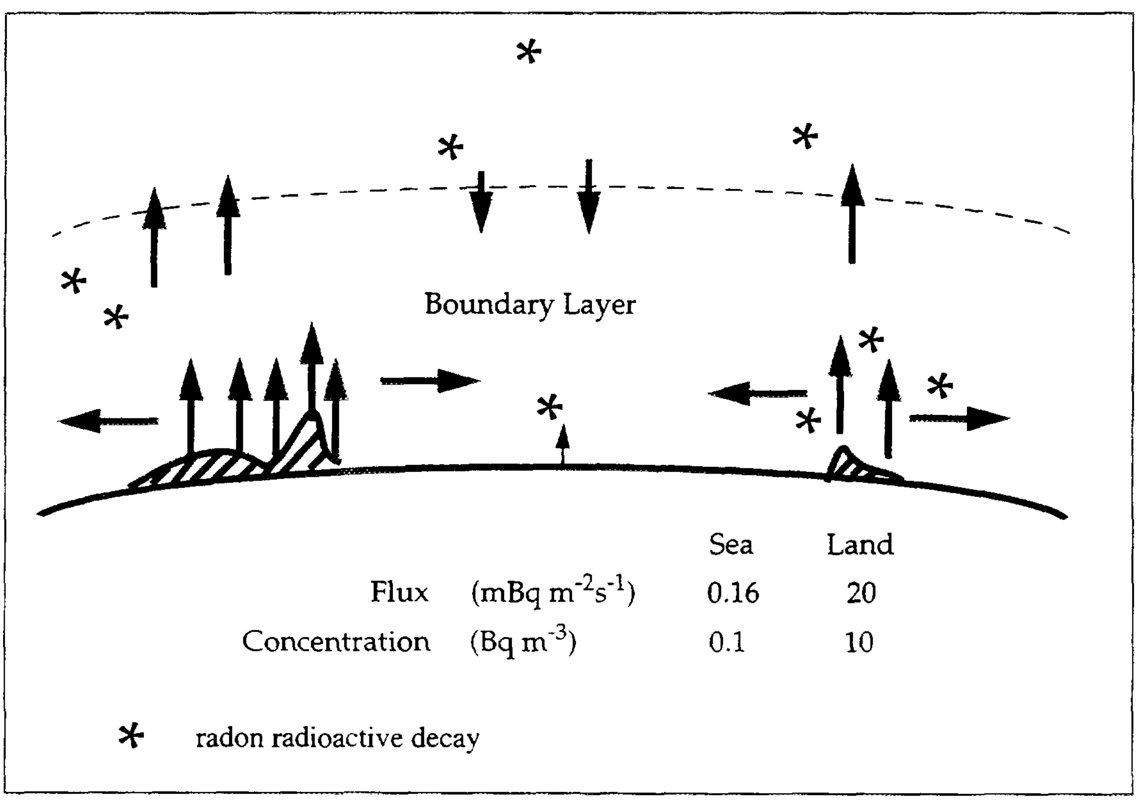

FIG. 3. Sector definition at Cape Grim.

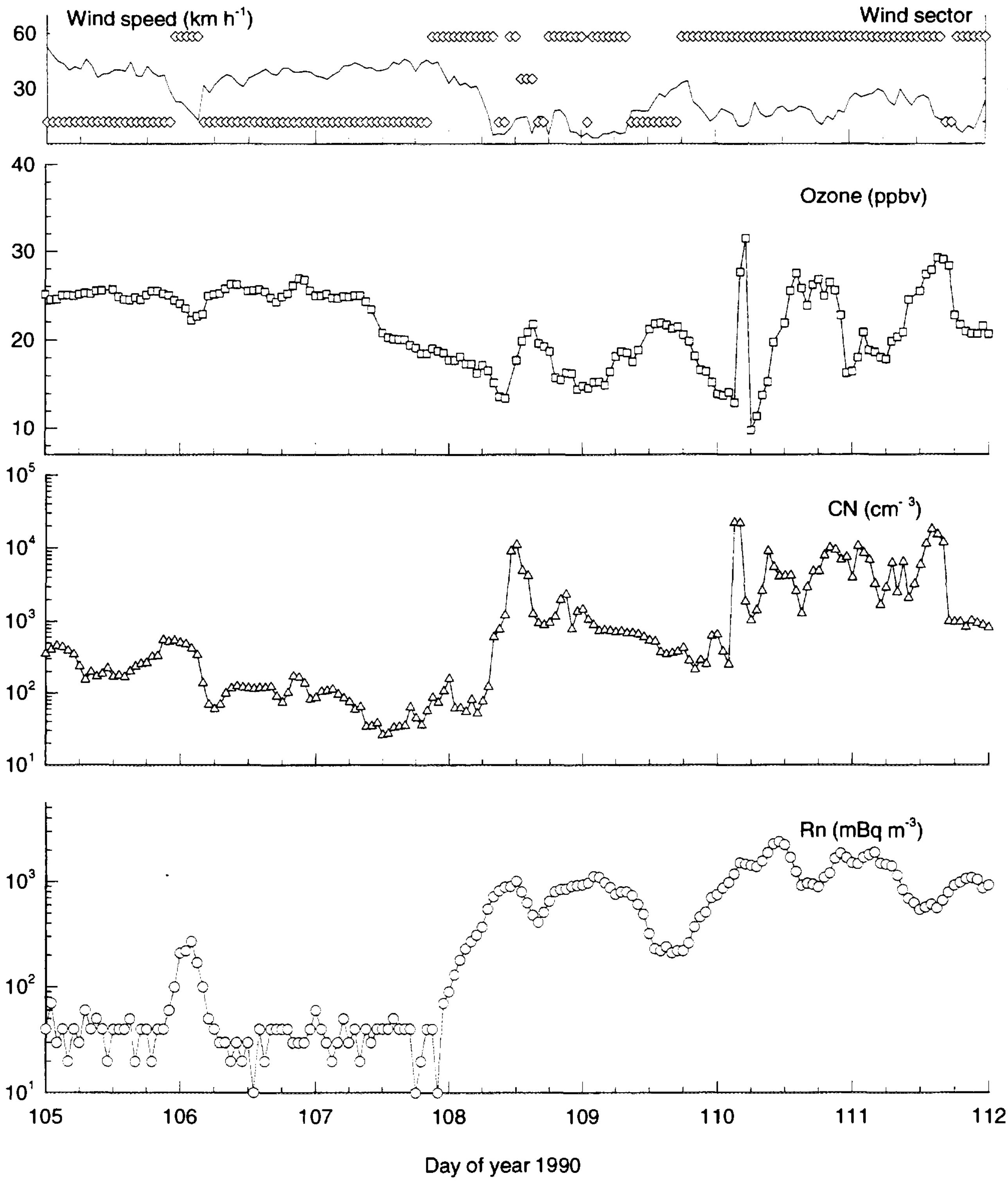

FIG. 4. Typical weekly data recorded at Cape Grim.

baseline from those which have been subject to recent pollution. This requirement is very important, especially for those analytical methods which rely on long sampling periods or when air is collected for archiving purposes.

Table I shows a list of measurable quantities which are the most generally applicable as baseline selection criteria because of the reliability and low cost of the instruments involved and the ease of interpretation of the data.

TABLE I. RANGE OF APPLICABILITY OF DIFFERENT BASELINE SELECTION CRITERIA

Criterion	Distance of pollution source from sampling point (km)			Timing Accuracy
	< 10	10–1000	> 1000	1 minute
Wind direction	yes	yes	no	1 minute
Condensation Nuclei (CN)	yes	yes	no	1 minute
Lead-212	yes	yes	no	1 hour
Radon-222	no	yes	yes	1 hour
Air Trajectories	yes	yes	yes	hours–days

Radon concentration is the only practicable baseline criterion for air parcels coming from distances over 1000 km. In fact, because of the negligible radon flux from the ocean and 3.8 days radon half-life, the low radon signal always indicates that the sampled air parcel has not been with a significant contact with land for two-to three weeks. This can correspond to distances far exceeding 1000 km distance which is the limit for other baseline criteria. It might be argued that for distances lower than 1000 km the wind direction can be a universal baseline criterion. Some case studies revealed, though, that it is radon rather than the wind direction which provides a better criterion for the baseline condition at such distances [2]. The radon baseline threshold can be easily established by examining a sufficient amount of hourly observations. In case of Cape Grim it is equal to 100 mBq m^{-3}.

2.3. Principles and practice of radon detection in baseline air

The principle of operation of ANSTO's high-sensitivity radon detector is based on two filter detector: air is drawn continuously through one filter which removes all radon and thoron decay products (daughters), then through a delay chamber in which some daughters are produced [3]. Finally air passes through a second filter which collects the daughters at a rate proportional to the radon and thoron concentration. The larger the delay chamber volume, the more sensitive the detector will be since more daughters are formed in the chamber. Unfortunately, there are some difficulties when one attempts to scale-up the instrument. A major challenge in the design of high sensitive two filter detectors is to prevent the daughters 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 means that flow rates of about a cubic metre per minute are necessary. As a result, pumping power and the second filter are have to be unacceptably large.

One can solve this problem by injecting and maintaining a constant concentration of sub-micron particles in the delay chamber [4]. With particles in the chamber, the daughters 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 m^3. This solution delivers excellent results as far as lower limit of detection is concerned but it requires a certain level of maintenance and too high a standard of housing to work unattended at remote sites.

A new ANSTO detector design addresses the above 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 [5]. The first measure separates the function of supplying filtered air to the detector from that of preventing plate-out of daughters. The former needs only a low flow rate. The latter requires very high flow rate.

The second measure utilizes the high diffusivity of radon or thoron daughters. Inside the delay chamber there are no aerosols, so the daughters remain as either atoms or small clusters of atoms no more than a few manometers in diameter. Therefore a wire screen is able to remove the daughters with high efficiency and very low flow impedance. Since the major power consumption in these detectors is by the air pump, use of screens makes it possible to dramatically reduce system power requirements.

Figure 5 illustrates schematically the principle of operation of this new dual flow loop two filter radon detector. The low flow loop takes ambient air through a delay volume to remove thoron and a first filter into the delay chamber. The high flow loop circulates air within the chamber forcing the air to go through a wire screen which acts as the second filter. The screen is mounted as close as possible to a scintillator which emits flashes of light when hit by alpha particles from the decay of the trapped daughters. Also indicated in the figure is a diffuser fitted across the chamber at the outlet of the internal blower. The diffuser makes airflow in the chamber close to laminar thus preventing excessive plate-out of daughters. The ratio of the internal to external flow rates is about 6 to 1.

The lower limit of detection of a 750 L detector is about 30 mBq m^{-3}. The detector can be a cost-effective basis for a global network of high sensitivity radon detectors.

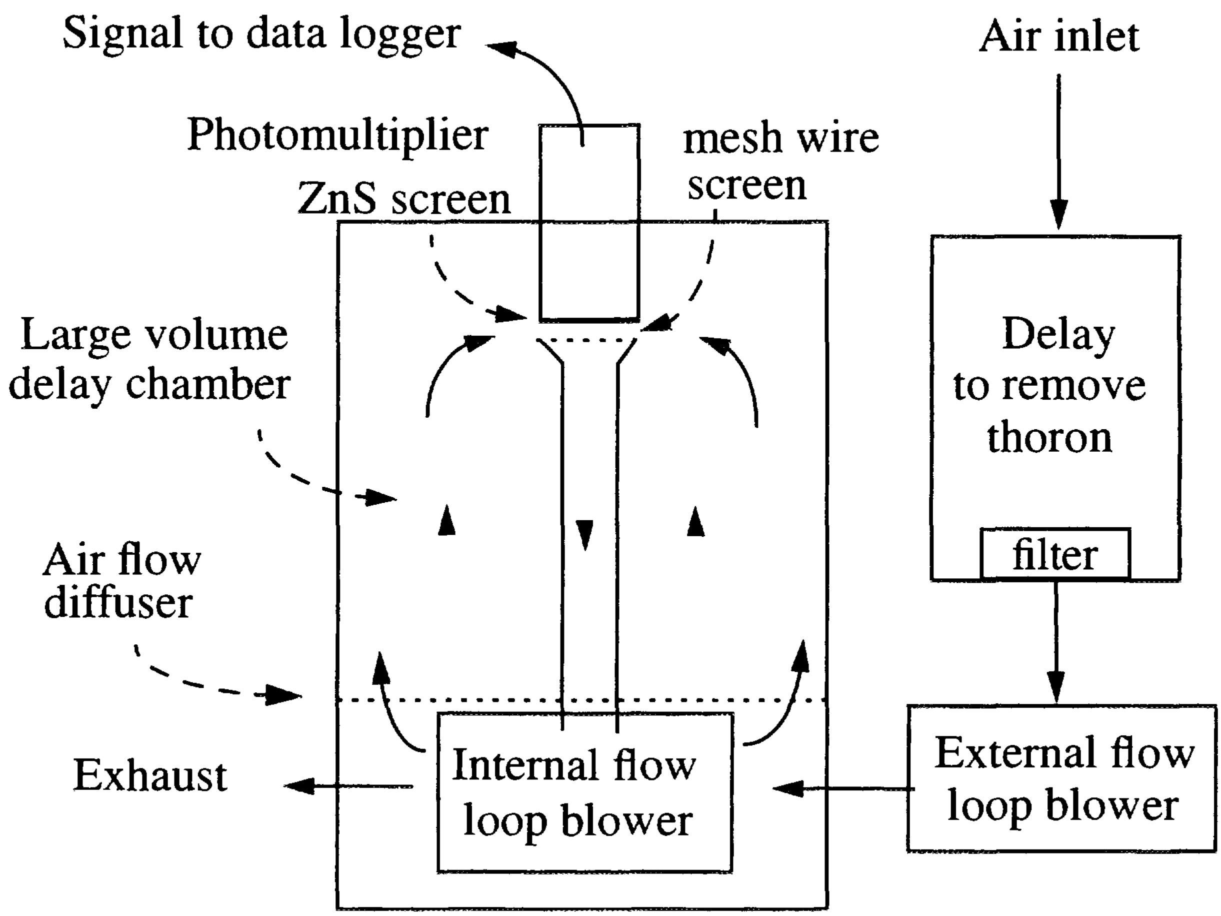

FIG. 5. Diagram of dual loop radon detector.

3. MAPPING OF RADON AND THORON SOURCE

3.1. Radon global source

Figure 6 shows an example of a simulation of radon concentration in air using a global atmospheric transport model. The radon data were in 1990 at Macquarie Island (solid line) using an ANSTO radon detector. The simulation (dashed line), based on a global air transport model, have been performed by Michel Ramonet of CNRS, France.

Although there is a measure of agreement between the experiment and calculations, there is a number of discrepancies, too. One reason for the discrepancies may be uncertainties in the radon source term. In the calculations shown, it has been assumed that all land emanates radon at the rate of 1 atom cm^{-2} s^{-1} (approximately 21 mBq m^{-2} s^{-1}). The assumption is an obvious simplification. An improved estimate of radon flux from every 5 × 40° grid has been done for Australia.

Australia has a good regional radon flux survey [6] and an extensive aerial gamma survey. Figure 7 shows a summary of the flux survey. In the figure some arrows are reduced to dots. This is a consequence of the radon scale (0 to 50 mBq m^{-2} s^{-1}). Results obtained around uranium mines have not been included in the figure. Rectangular grids indicate areas where airborne gamma data and/or radon flux measurements are available. A simple procedure was adopted to find the average radon flux from spot flux measurements and aerial gamma surveys (Fig. 8). The first step was to select maps covering an area of 100 × 120 km for which both radon fluxes and airborne gamma data were available. A relationship was found between the two by selecting six points from each gamma map according to a strict rule, so the selections were unbiased. The six values were averaged to give an estimate of the average gamma count which was converted to a map average radon flux. The maps were then matched as well as possible to the 5 × 4° grid, and grid averages obtained.

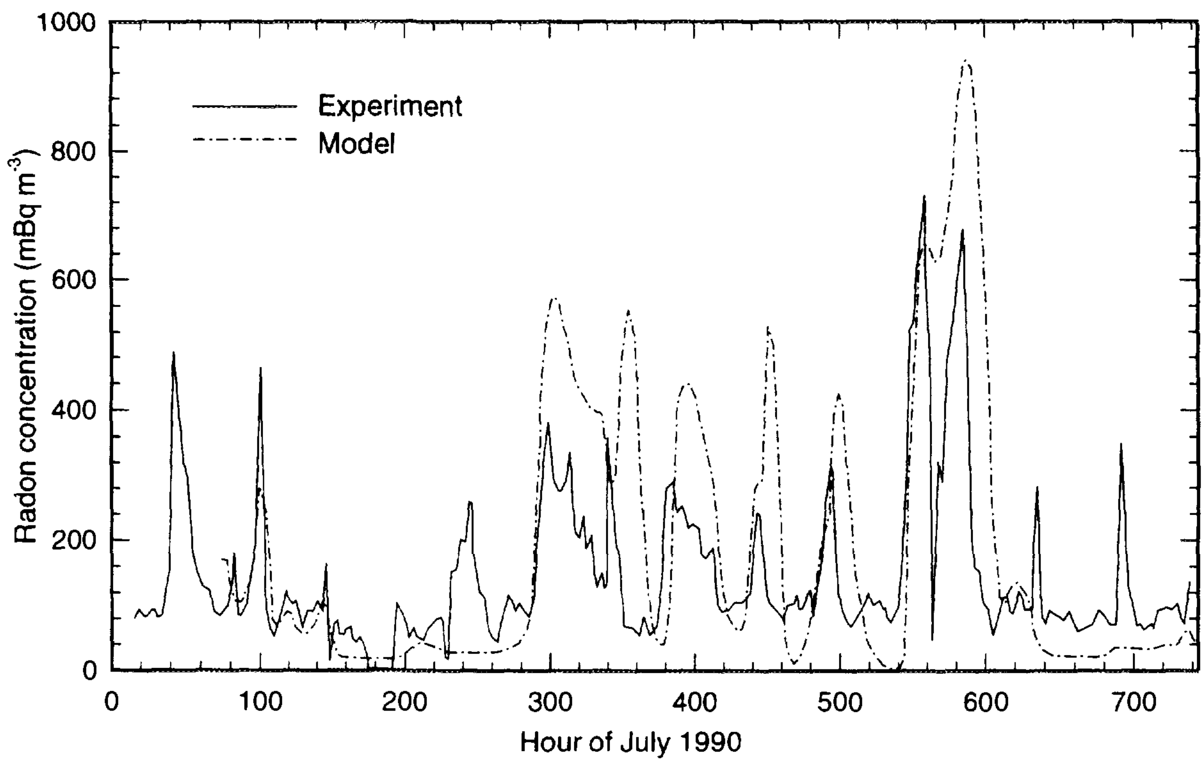

FIG. 6. Radon concentrations at Maquarie Island in July 1990; experiment and model calculations.

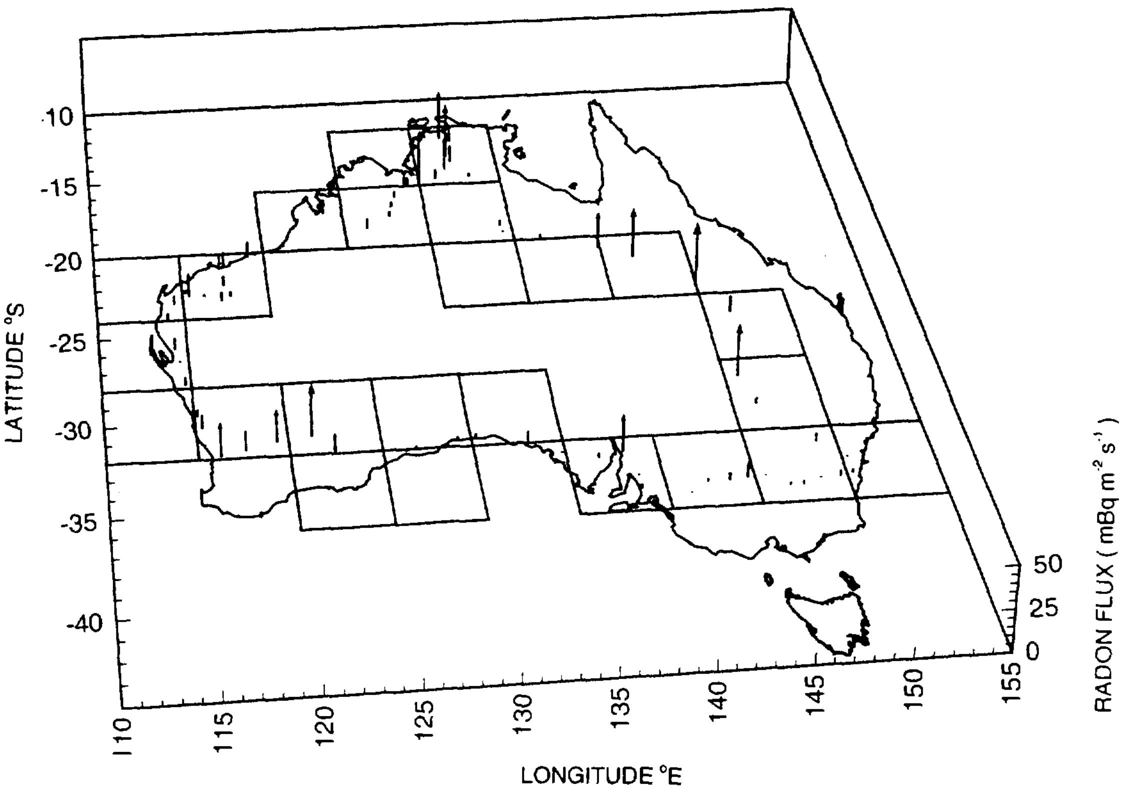

FIG. 7. Radon flux measurements in Australia.

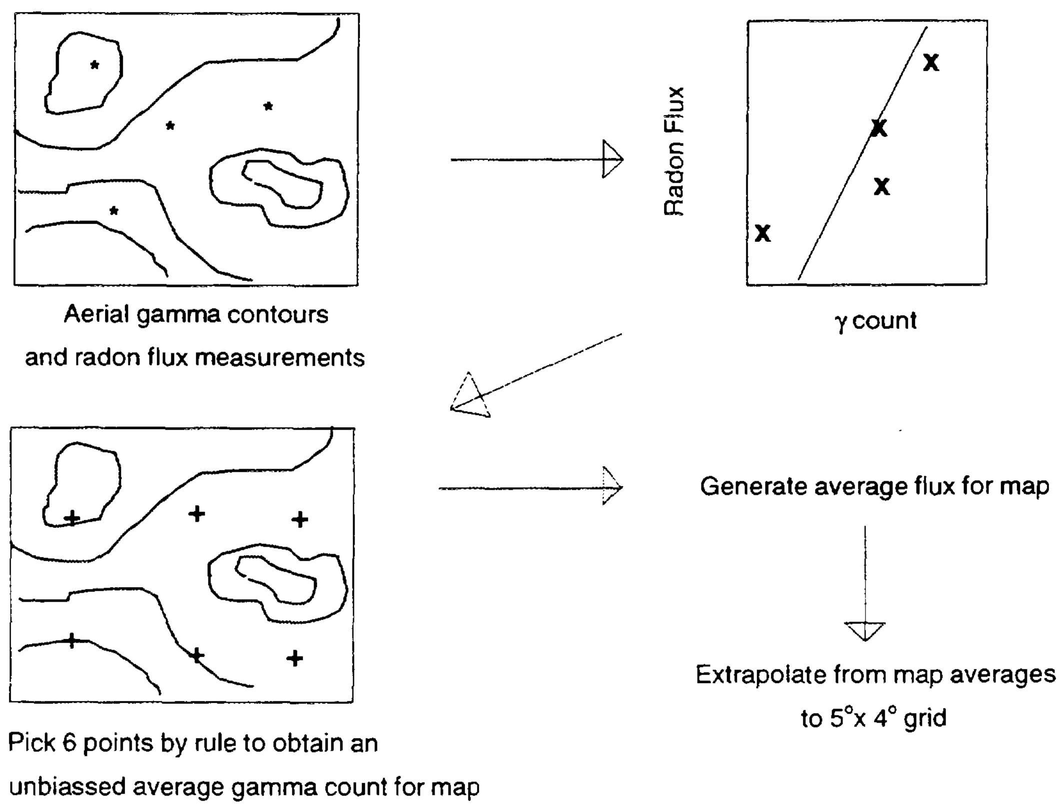

FIG. 8. Procedure for finding map average radon flux from spot flux measurements and airborne gamma survey.

The results of the flux evaluation procedure is shown in Fig. 9. The grids shown are the same as in Fig. 7. The top number in a cell is derived from spot radon measurements and the bottom from airborne gamma data. The two values, when both are available for a cell, agree quite well. It is also clear that the 1 atom cm^{-2} s^{-1} assumption is very inaccurate even on the 5 × 4° grid basis. From the bottom half of Australia, the average is 1.1, but for the upper, northern half it is 1.8. The whole data set gives 1.4 atom cm^{-2} s^{-1}. The extrapolation procedure can be improved in many ways starting with the use of more representative averages from digital airborne gamma maps.

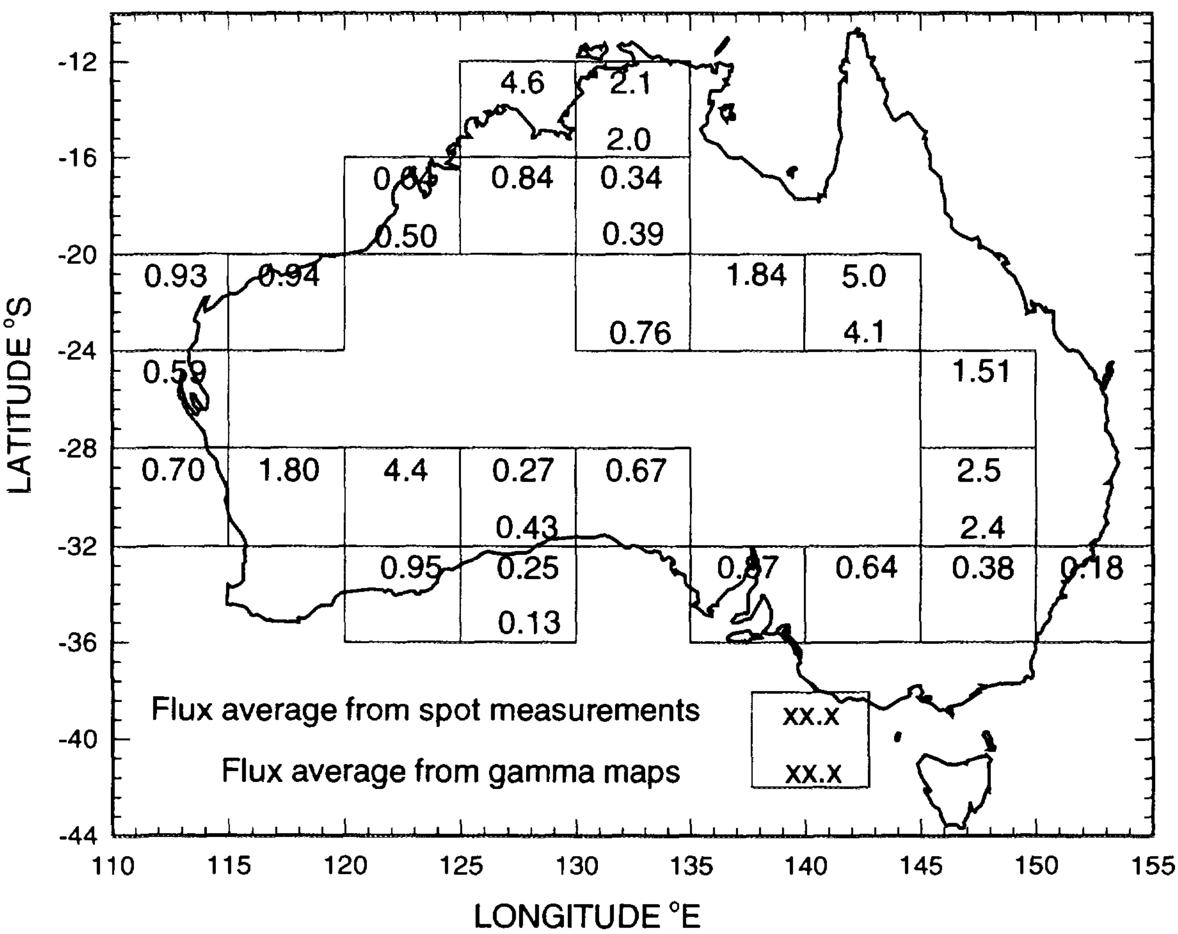

FIG. 9. Average radon fluxes from 5° × 4° grid boxes (one atom cm^{-2} s^{-1} corresponds to about 21 mBq m^{-2} s^{-1}).

3.2. Fast measurements of radon environmental fluxes

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.

Figure 10 shows the essential features of a fast radon and thoron emanometer used in the survey summarized in Fig. 7. The principle of operation of the emanometer is as follows [7]. At the start of the measurement the accumulation chamber (A) is placed over the ground. Air is drawn from the chamber 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 99% of the thoron decays, into the second cell (SC2) which records essentially counts from radon. The computer controlling the device stores the counts and resets the counts each 6 minutes. After 3–4 counting cycles it switches the inlet of the scintillation cells to ambient air. There is a small fan to ensure that thoron is well mixed in the emanometer chamber.

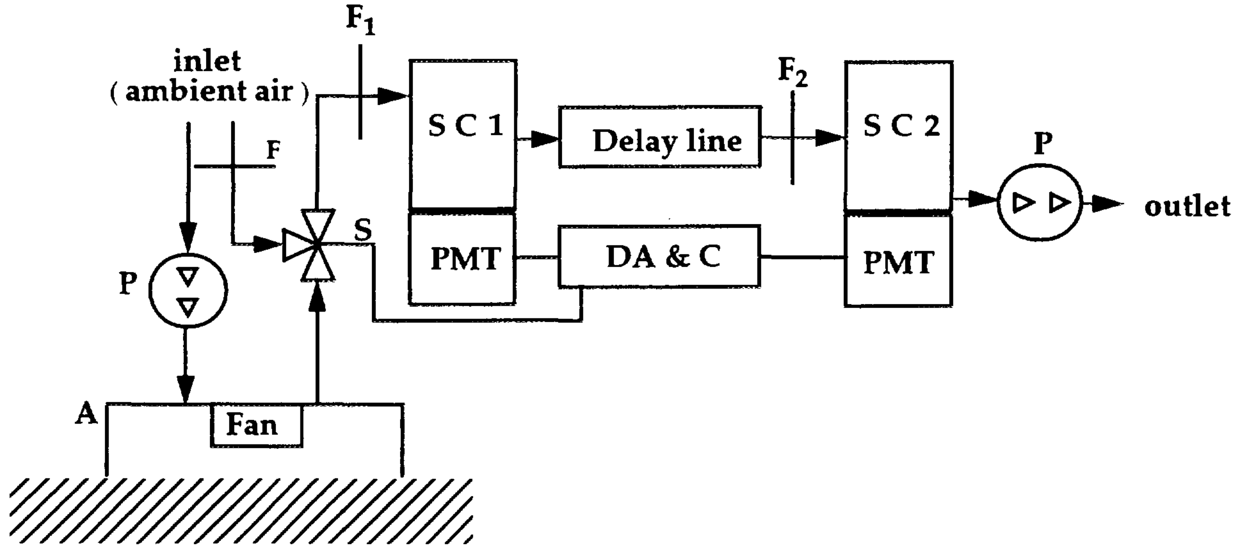

FIG. 10. Diagram of fast radon and thoron emanometer.

In practice, one can perform one measurement per hour providing that the next destination is within 30 minutes drive from the previous one. Table II shows the emanometer's sensitivity and Table III its lower limit of detection. It is clear that the 24 min count is adequate for a fast and precise evaluation of most naturally occurring radon and thoron fluxes. The sampling frequency per day of field work is typically less or equal to 10 sites, depending on site preparation time and distance between adjacent sites.

3.3. Local radon and thoron sources

Pb-212 is a decay product of thoron (Rn-220) which, like Rn-222, emanates from all land surfaces. Thoron is inert, but its very short half life of 55.6 s prevents it from being used as a tracer of air masses over distances more than a kilometre. Pb-212, with a half life of 10.64 h, can become attached to aerosol particles, and remain in air less than about 1000 km away from the thoron's source. The tracer would detect periods when air samples have been influenced by passage close to Hawaiian land. This complements radon measurements which do not distinguish between local and distant land contact.

In baseline air monitoring stations located at island or coastal sites with continental land masses being away more than 1000 km, only air trajectories close to local land will contain lead-212. The baseline air pollution station at the Mauna Loa Observatory in Hawaii is an example of such a station [8].

Meteorological data will indicate that an air parcel has approached local land, but lead-212 may gives a quantitative measure of the degree of contact. This can be done providing that a detailed mapping of the local thoron source is available. Two conditions have to be fulfilled. First, thoron fluxes are to be measured over as wide a range of soils as possible. Second, a selection of sites has to take into account all major air trajectories over Hawaiian land to the MLO.

A survey aimed at measuring the thoron source term to evaluate the use of Pb-212 as an atmospheric tracer was done on Hawaii. Figure 11a shows the measurement sites and the height contours. Fig.11b includes average rainfall isopleths and the measured thoron fluxes which are quantified by a subdivision into values below and above 300 mBq m^{-2} s^{-1}.A detailed discussion [9] of the survey reveals that the observed fluxes are strongly dependent on rainfall, soil depth, soil

TABLE II. FAST EMANOMETER SENSITIVITY FOR DIFFERENT COUNTING TIMES

Flux sensitivity (counts/mBq m^{-2} s^{-1})					
24 min count		60 min count		180 min count	
Radon	Thoron	Radon	Thoron	Radon	Thoron
8	0.8	39	2.1	188	6.5

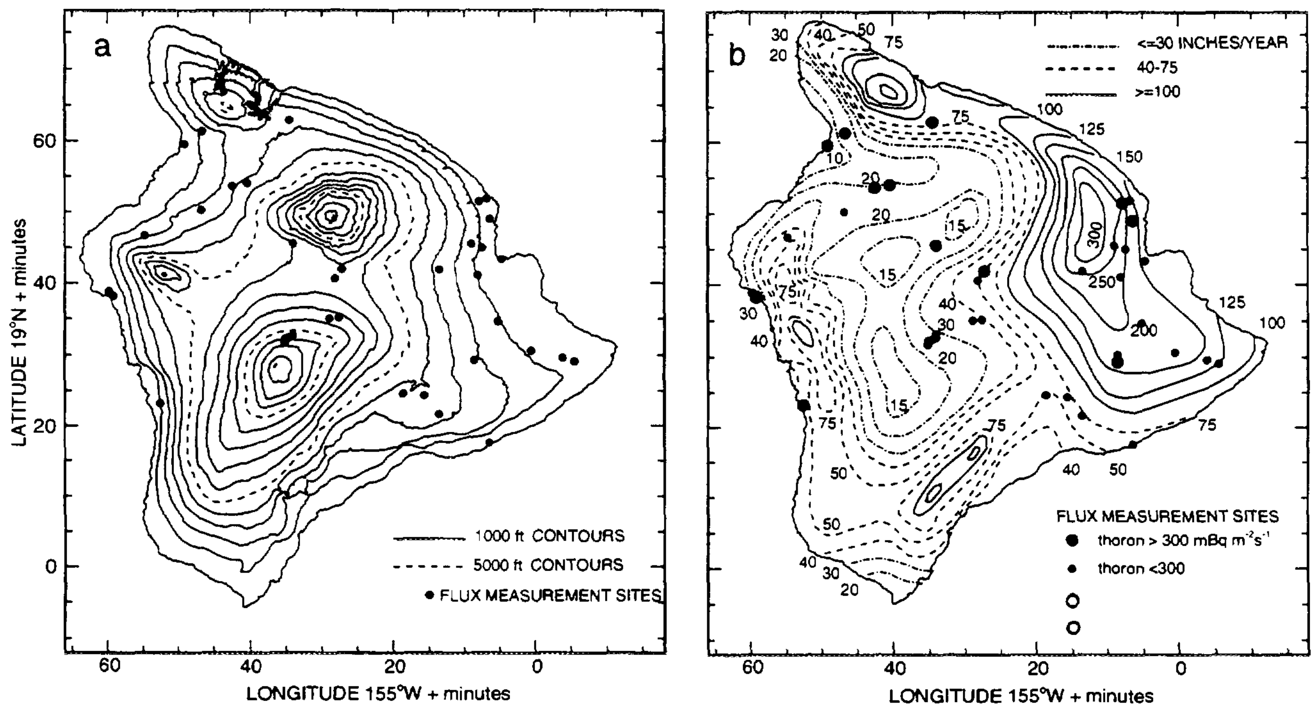

FIG. 11. Map of Hawaii showing thoron flux measurement sites.

particle size and land use. While the soil depth and particle size are not greatly different between the east and west sides of the island, the rainfall is much greater on the east side, resulting in distinctively different average fluxes. The estimated average fluxes from the dry western side of the island were a factor 5 higher than those from the wet eastern side.

3.4 High sensitivity thoron emanometer

Most of the thoron fluxes shown in Fig. 11 were determined by the fast emanometer described above. Fluxes from barren lava, ranging from 5 to 50 mBq m^{-2} s^{-1}, located above and immediately below the MLO were measured with a high sensitivity thoron emanometer which was assembled specifically for the survey [5,8]. The principle of operation is shown in Fig. 12. A large sampling area was obtained by taking a 2.7 × 3 m tarpaulin and spacing the central 6 m^2 a distance of 9 cm from the ground with rows of plastic cups. Air from the centre of the chamber was drawn at 400 L/min through a filter to a 400 L delay chamber where about half of the thoron decayed. The resulting lead-212 was collected on a nylon mesh.

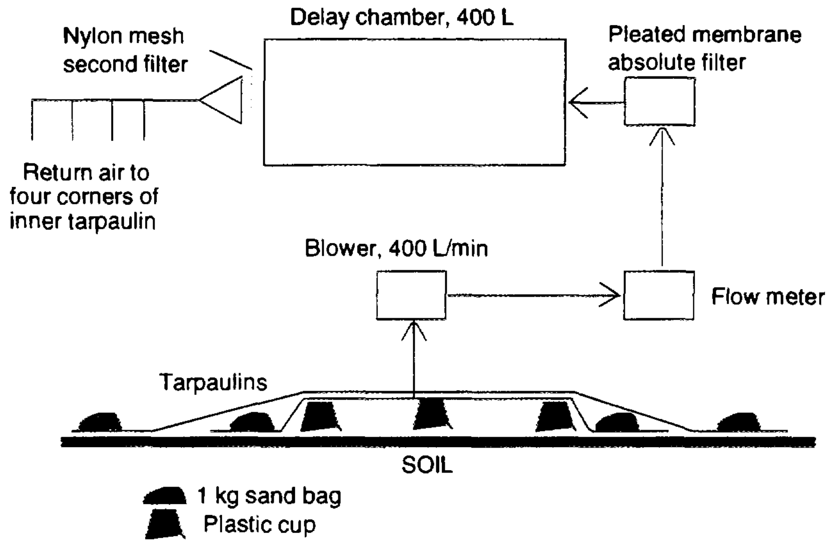

FIG. 12. High sensitivity thoron emanometer.

Air from the delay chamber was recycled to the four corners of the chamber. The concentration of thoron in the volume under the tarpaulin is inversely proportional to the height of the spacers. It is therefore possible to improve the sensitivity of the measurement by lowering the spacing as far as possible. On the other hand, the accuracy of the measurement is dependent on how well the volume is defined. On rough ground, a height greater than the roughness is needed. It is clear that at least some constructional parameters depend strongly on measurement sites.

Calibration of the instrument was performed by making side-by-side measurements with the fast emanometer which resulted in an agreement within 30% of a calculated value. It was estimated that the high sensitivity emanometer was able to detect thoron flux down to 1 mBq m^{-2} s^{-1}.

TABLE III. FAST EMANOMETER LOWER LIMIT OF DETECTION FOR DIFFERENT COUNTING TIMES

Lower level of detection (mBq m^{-2} s^{-1})					
24 min count		60 min count		180 min count	
Radon	Thoron	Radon	Thoron	Radon	Thoron
4.0	40	1.0	20	0.35	10

4. CONCLUSIONS

Radon-222 and lead-212 recorded at baseline air monitoring stations are effective indicators of contact of the air with distant and local land, respectively. Radon-222 concentrations measured at baseline stations can be modelled using global atmospheric transport codes. Hence the radon record enables an independent validation of global atmospheric transport models. The validation can be done for the response of the models to radon concentration changes on time scales as short as 1 hour. The use of radon as a tracer requires a better knowledge of the radon global source term if such comparisons are to become more precise.

It is possible to construct a map of radon flux averages from large land masses based on radon flux spot measurements and airborne gamma maps. Unique radon instrumentation had to be developed to measure baseline radon concentrations and radon and thoron fluxes to meet the requirements for the lower limit of detection and response in time.

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