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Seasonal variability of the radon-222 flux density from the Southern Ocean derived from atmospheric radon-222 measurements at the Cape Grim baseline station in Tasmania

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We present and discuss an estimate of the radon-222 (radon) flux density from the Southern Ocean and its seasonal variability. The flux estimate was based on selected hourly atmospheric radon concentration measurements in the Baseline wind sector of the Cape Grim station in Tasmania (40^{o}41'S, 144^{o}41'22"E) from 2001 to 2008.

The aim of the selection process was to define a subset of hourly radon observations corresponding to oceanic air parcels that were least perturbed by land emissions and exhibiting minimal exchange with the free troposphere, so that an equilibrium could be assumed between the measured radon concentrations and their oceanic source. The initial dataset included all events in the Cape Grim Baseline sector, defined solely by local wind direction (190º$<\theta<$280º). In this set, more than 75% of the hourly radon observations were already below 100 mBq m^{-3}.

We demonstrate that the strongest perturbation due to land emissions occurs at the beginning and end of each baseline period, with the duration of these transitional periods being 24 and 12 hours, respectively. Additional statistically significant terrestrial radon emissions and near-shore influences were identified and quantified by analysing special features of 10-day back trajectories, calculated using the HYSPLIT model, and the associated radon distributions. We show that the Australian terrestrial influence leads to an upward shift of the corresponding radon distributions, with the converse being true for Antarctic terrestrial influence. Statistically significant near-shore influences were attributed to horizontal radon gradients extending from the coast over the ocean, south from the Australian mainland and north from the Antarctic sea ice boundary.

Progressive application of the selection criteria contracts radon distributions of the resulting subsets, with the higher percentile concentrations undergoing the most pronounced reductions. For example, the concentration ranges of 90% of baseline observations (ie. from the 5^{th} to 95^{th} percentiles) were 483, 165, 126, 90, and 88 mBq m^{-3} for the five major radon sub-sets considered with progressively stringent selection criteria. The progressive reduction in concentration range for each category confirms the efficacy of the selections made, since the narrower the concentration range, the more homogenous the radon source probed by the observations. We also found that the more that the terrestrial influences could be reduced, the more clearly revealed was the seasonal variability in flux estimates. The ratio of summer to winter median radon concentrations increases from 1.13 for all baseline observations, to 1.6 for the final subset of baseline observations (considered to include only those radon observations corresponding to air parcels most closely in equilibrium with their oceanic radon source).

The final dataset included 900 hourly radon observations, about 3% of all baseline events recorded in the 2001-2008 period.

The marine boundary layer heights required for the flux estimates were derived from an ECMWF operational model reanalysis on a 1.5^o grid. We compared the reanalysis for 1998 with mini-lidar measurements gathered at Cape Grim and found that on average the lidar estimates were approximately 11% lower. Assuming that the application of our stringent selection criteria also excluded meteorological events such as frontal passage and significant boundary layer venting by deep active cumulus, we employed an entrainment velocity typical of the Southern Ocean marine boundary layer in the study region (0.004 ms^{-1} $\pm$0.002 ms^{-1}). The flux density means calculated using the above assumptions, expressed in units of mBq m^{-2}s^{-1} were 0.27 $\pm$0.10, 0.30 $\pm$0.11, and 0.19 $\pm$0.07 for the 2001-2008 composite year, winter, and summer, respectively, with the stated uncertainties resulting from uncertainties in the estimates of marine boundary layer heights at Cape Grim, and of the assumed entrainment

velocity.