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An estimate of the global distribution of radon emissions from the ocean

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[1] There is a need for improved estimates of the radon (^{222}Rn) flux density from the ocean for use in the modeling and interpretation of atmospheric radon in global climate and air pollution studies. We use a modification of a frequently used model of gas transfer to generate global predictions of ocean radon flux density for each month of the year (climate averaged) on a 192 by 94 global grid. Compared with the often-used approximation of a constant radon flux from the ocean, the model's predictions indicate large variations over regions of the ocean (a factor of ten is not uncommon). For example, latitude bands near the equator and Southern Ocean are predicted to emit relatively high average radon flux compared with other latitude bands. The predicted annually-averaged flux density from the ocean is 0.0382 mBq m^{-2} s^{-1} (0.00182 atoms cm^{-2} s^{-1}), smaller than some commonly-used estimates. *INDEX TERMS:* 0312 Atmospheric Composition and Structure: Air/sea constituent fluxes (3339, 4504); 1610 Global Change: Atmosphere (0315, 0325); 4860 Oceanography: Biological and Chemical: Radioactivity and radioisotopes. **Citation:** Schery, S. D., and S. Huang (2004), An estimate of the global distribution of radon emissions from the ocean, *Geophys. Res. Lett.*, *31*, L19104, doi:10.1029/2004GL021051.

1. Introduction and Methodology

[2] There has recently been an increased interest in modeling radon (^{222}Rn) in the atmosphere as a part of programs directed at improving climate and air quality studies and the interpretation of radon signals at new monitoring stations [*Barrie and Lee*, 2004]. The relatively poor specification of the radon source term (the distribution of radon flux over the earth's surface) has been identified as an important factor limiting the understanding of atmospheric radon and its use to test atmospheric transport models [*Barrie and Lee*, 2004]. Lack of information on radon flux from the ocean has been a factor limiting interpretation of baseline radon signals at coastal monitoring stations where, for certain wind trajectories, radon from the ocean can dominate that from land. Current estimates of the global radon source term used for atmospheric modeling either tend to ignore radon flux from the ocean or assign it a small constant value [*Jacob et al.*, 1997; *Schery and Wasiolek*, 1998; *Taguchi et al.*, 2002]. On the other hand, much progress has been made in the study of gaseous exchange at the sea's surface and interest remains high in this subject [*Donelan et al.*, 2002]. Good-quality global datasets are now available for many meteorological variables over the earth's surface [*Kalnay et al.*, 1996]. Given the above considerations, it seems an appropriate time to carry out a more sophisticated estimate of the radon source term for the ocean.

[3] The starting point of our predictions for ocean radon flux is the frequently used model of *Wanninkhof* [1992] for gas transfer velocity containing a quadratic dependence on wind speed. The Wanninkhof model is meant to apply to longer-term, time-averaged fluxes. Its proportionality coefficient is sensitive to the time period (both length and lapse interval) over which its input wind speed is averaged, in part due to the model's non-linear dependence on wind speed. In the present case, in order to generate global estimates of ocean radon flux with some generality for a first look by atmospheric radon modelers, we decided to use monthly climate averages for the global wind speed (from the National Centers for Environmental Protection (NCEP) reanalysis project [*Kalnay et al.*, 1996], dataset: flx.gau.grib.mean.clim.y1979-1995b.y1979-1995). The averaging protocol for these winds is different from the cases studied in the Wanninkhof paper, so we anticipate a change in the value of the model's coefficient. In addition, in order to apply the model to radon flux from the ocean, even under the simplifying assumptions of deep ocean (sea bottom radon flux not a factor) and locations far from shore (no back diffusion from higher atmospheric radon concentrations), it is necessary to apply the transfer velocity to the radon concentration in surface water. Variation in this concentration is not well mapped for the ocean's surface. As a surrogate, we chose to use the concentration of the more extensively studied ^{226}Ra in the surface mixing layer. Radon is the direct decay product of this radium. The ratio between activity concentrations of ^{222}Rn and ^{226}Ra is not necessarily constant (dependent on such factors as transfer rate to the atmosphere and depth of the mixing layer), but study of surface radon and radium profiles [e.g., see *Smethie et al.*, 1985] suggests that variation of this ratio in the surface mixing layer, [Rn]/[Ra] $\sim$ 0.7, is usually small compared with the much larger variation in the transfer velocity itself. Use of this surrogate is a second factor that would be expected to change the magnitude of the coefficient in the Wanninkhof model. Finally, since we are interested in actual radon flux for various conditions of

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the ocean and atmosphere, we omit any normalization convention to a reference temperature or gas.

[4] In summary, the model we use for estimating the radon flux density from the ocean's surface takes the following form

$$F_{av} = a_{Rn} u_{av}^2 (Sc)^{-1/2} [Ra], \quad (1)$$

where F_{av} is the average radon flux in milliBequerels per square meter per second (1 mBq m^{-2} s^{-1} = 0.0477 atoms cm^{-2} s^{-1}), a_{Rn} is a constant coefficient (dimensions of s m^{-1}) whose value must be determined, u_{av} is the wind speed in meters per second 10 m above the sea surface, and [Ra] is the radium concentration in the surface mixing layer in units of milliBequerel per cubic meter. The variable Sc is the dimensionless Schmidt number for radon in seawater which gives a temperature (t) dependence to the flux. We needed a wider temperature range than given in the Wanninkhof paper, so we used a similar procedure to derive a new polynomial estimate. The result, for $0°C \leq t \leq 35°C$, was $Sc = A + Bt + Ct^2 + Dt^3 + Et^4$ where A = 3412.8, B = −235.6, C = 8.8563, D = −0.1951, and E = 0.0018652. For the ^{226}Ra concentration [Ra] in the surface layer of the ocean, we divided the ocean into sectors and estimated constants or simple linear dependences for these sectors using data in *Peng et al.* [1979]. The result (in mBq m^{-3}) was 1180 for latitudes between −40° and +40°, (1180 + [−40° − latitude]*60.8) for latitudes between −40° and −70°, 3000 for latitudes less than −70°, 1180 for latitudes greater than 40° in the North Atlantic, 1180 + (latitude − 40°)*42.9 for latitudes between 40° and 70° in the North Pacific, and 2470 for latitudes greater than 70° in the North Pacific. The coefficient a_{Rn} in equation (1) was then determined by adjusting it to give best prediction for the radon flux data (obtained by the profile deficiency method) at ninety stations for the Atlantic and Pacific oceans in the same paper. These radon data were chosen because they provide the most varied coverage of position and season under a consistent calibration methodology. The magnitude of the wind at 10 m (from its horizontal components), and the sea surface temperature (used to calculate the Schmidt number), were taken from the NCEP's dataset for the same (climate-averaged) month of the year and sector covering the position. The result was $a_{Rn} = 4.15 \times 10^{-5}$ s m^{-1}, which provided an average of the ninety model predictions equal to the average of the ninety measurements.

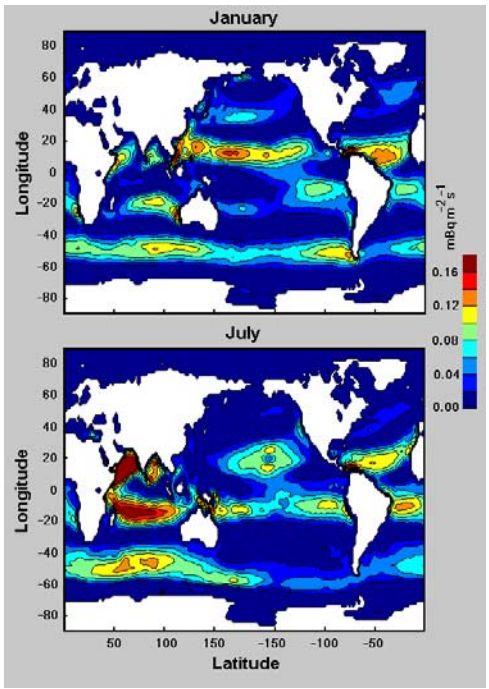

Figure 1. Model predictions for radon flux density for January and July.

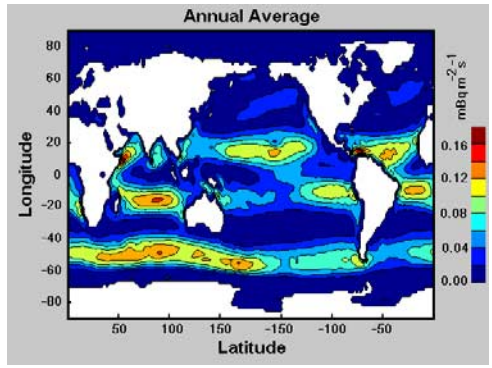

Figure 2. Annually-averaged model predictions for radon flux density.

2. Results and Discussion

[5] Using NCEP's monthly climate averages for the years 1979–1995 for surface wind speed at 10 m (from the horizontal components), surface temperature, and ice, global predictions of ocean radon flux density were made from equation (1) with the above [Ra] distribution over a 192 × 94 grid. Predictions were made for each month, as well as a year average obtained by averaging the results for each month. The flux density of radon from areas of sea ice, which are a small proportion of the total ocean area, were arbitrarily reduced by a factor of 10 from the unfrozen prediction. We could find no information on radon flux from sea ice, but based on molecular and turbulent diffusion coefficients, a reduction by 10 is probably still an upper bound on the actual flux reduction.

[6] The individual thirteen files for radon flux density are available as auxiliary material[1]. We here show some representative cases and provide discussion. Figure 1 shows predictions for the months of January and July. Compared with the often-used assumption by atmospheric radon modelers of a constant flux density from the oceans, the predictions of the present model clearly show major variation. A variation in average flux density by a factor of ten or more is not uncommon between major areas of the ocean. Variation from month to month is not as strong, but still very significant. For example, there is a large variation between January and July for the Southern Pacific ocean west of the southern part of South America. The model of

[1]Auxiliary material is available at ftp://ftp.agu.org/apend/gl/2004GL021051.

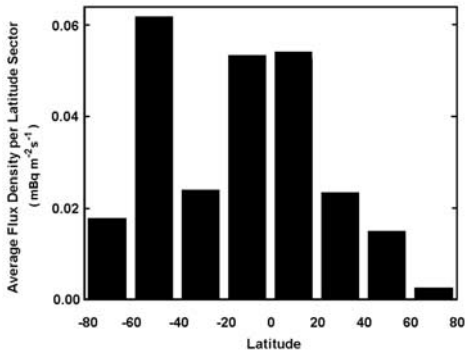

Figure 3. Annually-averaged radon emissions for latitude bands.

equation (1) is strongly dependent on surface winds over the ocean. Most of the variation seen in predictions like that of Figure 1 is due to variation in the speed of the wind, not sea surface temperature or radium concentration. Given the known large variation in wind magnitude over the ocean's surface, these predictions of large radon flux variation are not surprising.

[7] Figure 2 shows the prediction for the yearly average. The variation between large areas of the ocean still remains strong. We calculated a yearly average flux density for the ocean, weighting the value for each cell of the grid by its latitude-dependent physical area. The result was 0.0382 mBq m^{-2} s^{-1} for the entire ocean. For latitudes between 60°N and 60°S, which is a sector important for some modelers, the corresponding value is 0.0415 mBq m^{-2} s^{-1}. These numbers are smaller than some previously assumed values, for example 0.14 mBq m^{-2} s^{-1} for the entire ocean [*Schery and Wasiolek*, 1998], 0.105 mBq m^{-2} s^{-1} for 60°N and 60°S [*Jacob et al.*, 1997], and 0.21 mBq m^{-2} s^{-1} for 60°N and 60°S [*Taguchi et al.*, 2002]. The corresponding averages from our model for each month did not differ a great deal from the annual averages (<±10%). Figure 3, a plot of the annually-averaged flux density for twenty-degree-wide latitude bands, provides more detail of variation with latitude. Latitude bands around the equator and the Southern Ocean (near 50°S) are predicted to have comparatively higher radon emissions per unit ocean area, again primarily due to higher average winds. For the Southern Ocean latitudes, there is also some contribution from higher surface-water radium and radon (present due to upwelling). Our average flux density for the entire ocean, 0.0382 mBq m^{-2} s^{-1}, is much smaller than the common estimates for the average flux density from land, which are in the approximate range 20 to 35 mBq m^{-2} s^{-1} [*Schery and Wasiolek*, 1998].

[8] Perhaps a central question with our predictions is just how accurate are they? There are well known major uncertainties with models of gas transfer such as that underlying equation (1) [*Wanninkhof*, 1992; *Donelan et al.*, 2002], and the evidence for a specific functional dependence of gaseous flux from the ocean on wind speed is less than convincing. Our use of equation (1) includes additional approximations such as use of radium concentration in place of radon concentration, and climatological, rather than short-term, averages for wind and temperature. As a step in the direction of validation analysis, we have applied our model to some of the major cases of radon flux measurement from the ocean done independently of those used for our calibration of equation (1). Table 1 shows the result. For this comparison, we have tried to match our model predictions to the same locations and months of measurement, if known. However, we can only predict the climate-based monthly average over a sector of about 1.9 by 1.9 degree. We cannot give a prediction for a specific date and point location, so this is one source of uncertainty in the comparison. Given these qualifications, a quick perusal of Table 1 would suggest that an uncertainty of at least a factor of two, either large or small, is present in our predictions. Atmospheric modelers wishing radon flux density predictions averaged over specific, presumably shorter, time periods, would need to redetermine the coefficient in equation (1) using specific wind and temperature data averaged over the period of interest. However, the results provided here should still be a significant improvement over many of the previous simpler estimates that have been used by modelers.

3. Conclusions

[9] 1. A radon flux model using the wind and temperature dependence of *Wanninkhof* [1992], and assuming sea-surface radon concentration proportional to sea-surface radium concentration, gave an annually averaged global flux density from the ocean of 0.0382 mBq m^{-2} s^{-1}. This result is smaller than values that have often been assumed for the ocean source term in global model predictions of atmospheric radon. It is much smaller (about 0.1% to 0.2%) compared with common estimates of the average flux density from land, but could still be a significant source of atmospheric radon at coastal and marine locations for wind trajectories that cover long stretches of ocean.

[10] 2. The model predicts significant variation in the average radon flux density for large regions of the oceans,

Table 1. Comparison of Model Predictions for Radon Flux Density With Measurements

Model Average (mBq $m^{-2}s^{-1}$)	Measurement Average (mBq $m^{-2}s^{-1}$)	Reference	Comments
0.0796	0.0432	*Smethie et al.* [1985]	21 measurements, tropical Atlantic, radon deficiency technique
0.0315	0.0844	*Kawabata et al.* [2003]	13 measurements, NW Pacific, radon deficiency technique
0.0473	0.155	*Wilkening and Clements* [1975]	1 measurement, offshore Hawaii, accumulator technique
0.0837	0.0503	*Chambers et al.* [2002]	analysis of atmospheric radon from the Southern Ocean, 40S to 60S by 70E to 140E for the model annual average calculation

primarily due to its strong dependence on surface wind speed. A factor of ten variation is not uncommon. Latitude bands near the equator and Southern Ocean have relatively high average radon emissions.

[11] 3. Comparison of model predictions with independent measurements of radon flux density suggests a factor of about two is a lower bound on the accuracy of its predictions. Predictions are particularly sensitive to the wind dependence of the underlying gas transfer model. Any inaccuracies in this underlying model would carry over to the present radon flux density predictions.

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