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Radon-222 and radium-226 in southeastern Bering Sea shelf waters and sediment

DAVID M. GLOVER*† and WILLIAM S. REEBURGH*

(Received for publication 11 April 1986)

Abstract—Radon-222 and ^{226}Ra activities were measured in the waters and sediment of the southeastern Bering Sea shelf to evaluate the use of radon as a tracer of gas exchange, water column mixing and sediment–water exchange. Cross-shelf distributions of ^{222}Rn and ^{226}Ra are presented. Gas transfer coefficients were estimated using near-surface ^{222}Rn deficiency measurements. A statistically significant linear relationship between averaged wind speed and transfer coefficient was found. Vertical eddy diffusivities were evaluated by applying a one-dimensional model to near-bottom excess ^{222}Rn distributions; these diffusivities were compared to independently determined values. The one-dimensional model applied to the near-bottom ^{222}Rn data was found to be inadequate and a two-dimensional model was applied to improve the fit between model and data. Exchange across the sediment–water interface was computed from the deficiency of ^{222}Rn measured in sediment cores, standing crop estimates of excess ^{222}Rn in the water column and ^{222}Rn production rates of sediment surface grab samples. Biological irrigation of the sediments appeared to be the primary exchange mechanism between the sediment and water columns. Distributions in the water column showed finestructure reported previously and suggested biological removal of ^{226}Ra .

INTRODUCTION

WE PRESENT cross-shelf and sediment distributions of ^{222}Rn and ^{226}Ra from the southeastern Bering Sea shelf. These measurements were undertaken late in the PROBES (Processes and Resources of the Bering Sea shelf) project. They provide information on (1) gas exchange rates, which constrain carbon dioxide budgets; (2) near-bottom mixing rates, which provide information on the supply of regenerated nutrients from waters below the photic zone and (3) chemical fluxes from the sediments to the water column. The PROBES project focused on physical and biological interactions in the southeastern Bering Sea and provided an opportunity for extensive sampling. As far as we know, these ^{222}Rn and ^{226}Ra data are the most extensive shelf data presented to date.

Radon-222 is the inert gas, short-lived (half life = 3.83 days), radioactive daughter of ^{226}Ra (half life = 1620 years), a partially soluble product from the decay of insoluble ^{230}Th (half life = 7.5×10^4 y). These differences in chemical and radioactive properties and concentrations detectable at natural levels make ^{222}Rn and ^{226}Ra an attractive tracer pair for evaluating sediment chemical flux, near-bottom mixing intensity and gas exchange rate across the air–sea interface. The ^{238}U (half life = 4.5×10^9 y) decay series in Fig. 1 shows the tendency of ^{226}Ra to be incorporated largely in the sediments

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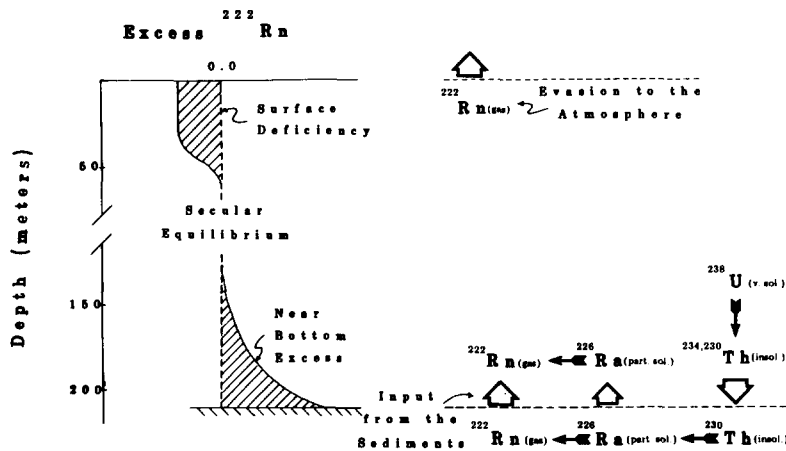


Fig. 1. Ideal radon profile (adapted from HAMMOND *et al.*, 1975). Over most of the water column, ^{222}Rn is in radioactive equilibrium with its parent, ^{226}Ra . Near the surface, the deficiency in ^{222}Rn is caused by evasion to the atmosphere. Near the bottom, an excess of ^{222}Rn is caused by input from the sediments. A portion of the ^{238}U decay series and the important properties of the members are also shown.

(BROECKER, 1974). About 10% of the ^{226}Ra in the sediment diffuses into the water column (KEY *et al.*, 1979a), giving rise to a supported level of ^{222}Rn . The ^{226}Ra remaining in the sediment decays to ^{222}Rn , which then engages in a unidirectional flux toward the atmosphere. Radium-226 radioactivities are subtracted from ^{222}Rn radioactivities to obtain the activity of excess or deficit ^{222}Rn .

Radon-222 is in secular equilibrium with seawater ^{226}Ra in locations removed from either the sediments or the atmosphere. Disequilibria between ^{222}Rn and ^{226}Ra at the sediment-seawater or air-sea interfaces lead to distributions similar to those shown in Fig. 1. There is a deficiency of ^{222}Rn with respect to dissolved ^{226}Ra due to the escape of ^{222}Rn into the atmosphere near the air-sea interface. The integrated deficiency of ^{222}Rn in the near-surface waters is related to the rate of gas exchange (transfer coefficient) (PENG *et al.*, 1979). Several models have been presented to explain the mechanism of gas exchange (DANCKWERTS, 1970); the integrated ^{222}Rn deficiency gives a net flux and is a good test for the models (TORGERSEN *et al.*, 1982; HOLMÉN and LISS, 1984). Excess ^{222}Rn is present near the sediment-seawater interface due to ^{222}Rn escaping from the interstitial waters of the sediment. The distribution of this excess ^{222}Rn above the bottom can be used to calculate vertical eddy diffusivities by fitting a one-dimensional exponential model to the distribution (BROECKER, 1965). The flux of ^{222}Rn from the interstitial waters of the sediments results in a radon deficiency in the near-surface sediments due to the unidirectional flux of radon. The sediment ^{222}Rn flux can be calculated from the sediment ^{222}Rn deficiency by equating the two (SMETHE *et al.*, 1981).

The gas transfer coefficient can be related to a hypothetical stagnant boundary film thickness (DANCKWERTS, 1970). Radon-222 has been used in gas exchange studies and film thickness values for a wide range of locations have been reported (EMERSON, 1975; BROECKER and PENG, 1971, 1974; PENG *et al.*, 1974). These film thicknesses have been summarized in BROECKER and PENG (1982) and range from 10 to 120 μm in the oceans.

Although a relationship between wind speed and gas exchange rate is reported from laboratory experiments and theoretical considerations (DEACON, 1977; LISS, 1973; BROECKER *et al.*, 1978; KANWISHER, 1963), a consistent relationship is not always found in the field (PENG *et al.*, 1979; BROECKER and PENG, 1982; HASSE and LISS, 1980; LISS, 1983). Empirical linear relationships between transfer coefficient and average wind speed give the best results to date (HARTMAN and HAMMOND, 1984; WANNINKHOF *et al.*, 1985; SMETHIE *et al.*, 1985).

Radon-222 has been used to obtain information about vertical eddy diffusivities in near-bottom waters in fjords (SMETHIE, 1981), offshore basins (CIJUNG, 1973; BERELSON *et al.*, 1982), lakes (IMBODEN and EMERSON, 1978) and the open ocean (BROECKER and KAUFMAN, 1970). The distributions of ^{222}Rn appear to vary widely with time in nearshore environments, such as estuaries and continental shelves, and provide less clear results (HAMMOND *et al.*, 1977). BISCAYE *et al.* (1978) found that inhomogeneities in sediment ^{222}Rn production rates in the New York Bight made the interpretation of their excess ^{222}Rn profiles difficult.

The distribution of ^{222}Rn and ^{226}Ra in the sediments of the Washington continental shelf (SMETHIE *et al.*, 1981) and the San Francisco Bay (HARTMAN and HAMMOND, 1984) has been used to investigate sediment mixing processes. They find molecular diffusion is insufficient; biological irrigation (bio-irrigation) is needed to explain the deficiency of ^{222}Rn present. Biological irrigation is the process by which organisms living within the upper centimeters of the sediment move water through the interstitial spaces without disturbing the sediment grains. GRUEBEL and MARTENS (1984), however, find only molecular diffusion is necessary to explain the ^{222}Rn deficiency in the sediments of the White Oak River estuary (NC). However, their cores failed to attain secular equilibrium between ^{222}Rn and ^{226}Ra possibly due to lost radon or the "slurry effect" (KEY *et al.*, 1979a).

STUDY AREA

The southeastern Bering Sea shelf is unusually wide (500 km), has a stepwise uniform depth gradient and graded sediments (SHARMA, 1979). The circulation is reasonably well understood (COACHMAN and CHARNELL, 1979; COACHMAN and WALSH, 1981; COACHMAN, 1986) and hydrographic fronts lead to intense and sustained primary production during summer. These fronts occur at breaks in the shelf topography and effectively divide the shelf waters into three domains by water depth (H) (SCHUMACHER and KINDER, 1983; COACHMAN, 1986): inner shelf ($H < 50$ m), middle shelf ($50 \text{ m} < H < 100$ m) and outer shelf ($100 \text{ m} < H < 150$ m). The outer and middle fronts can be identified as zones of enhanced horizontal gradients of properties; the inner front appears as a zone of transition in water column structure (COACHMAN, 1986). Net horizontal advection in the outer and inner shelf domains is small, approximately 5 cm s^{-1} and $1\text{--}5 \text{ cm s}^{-1}$ to the northwest, respectively (COACHMAN, 1986); the middle shelf domain has no net horizontal advection although the tidal excursions are large (SCHUMACHER and KINDER, 1983). The mid-depth waters of the outer shelf exhibit mixing between cold, near-bottom middle shelf and warmer, upper layer off-shelf waters along and between finestructure layering (COACHMAN, 1986). The physical and hydrographic characteristics of the southeastern Bering Sea shelf, combined with the intensity of our sampling program provided an excellent opportunity to use ^{222}Rn and ^{226}Ra as tracers of air-sea gas exchange, mixing intensity and sediment chemical flux in a shelf environment.

SAMPLING AND ANALYTICAL METHODS

The near-surface waters, mid-depth waters, near-bottom waters and sediments were sampled during October 1980, June–July 1981, June 1982 and July 1983 using 30-l Niskin bottles, a Van Veen grab sampler and a Benthos gravity corer. Five water column profiles were measured in 1980, 27 in 1981 and 8 in 1982 for a total of 20 near-surface, 19 mid-depth and 20 near-bottom profiles, several of which were multi-purpose profiles. Fifteen sediment surface grab samples were collected in 1979 and 13 in 1981; sediment cores were obtained in 1982 and 1983. The sample locations from 1980 (○), 1981 (+) and 1982 (X) are shown in Fig. 2a; several station locations overlay each other. Figure 2b shows the locations of the sediment surface grab samples from 1979 (+) and 1981 (X) and the two sediment cores (○).

Radon-222 analysis

Radon-222 was extracted from 19-l seawater samples using the methods outlined by MATHIEU (1977). Briefly, water samples were transferred through Tygon tubing to evacuated 19-l glass carboys. The radon was stripped from the sample with a circulating helium stream and trapped on an activated charcoal column kept at -55°C by a Cryocool unit (Neslab model CC-60). The radon was then baked off the trap at 400°C and transferred to an alpha scintillation counting cell. With the stripping and transfer boards (Applied Science of Piermont), 4 samples could be run simultaneously with a 140 min turnaround time.

Radium-226 analysis

Radium-226 was determined by the radon ingrowth method (KEY *et al.*, 1979b), so the terminal analysis for ^{222}Rn and ^{226}Ra was identical. Immediately after the radon extraction the water sample was passed through a 15.24×2.54 cm PVC pipe packed with Mn-impregnated acrylic fiber (MOORE, 1976). These fibers quantitatively remove Ra from seawater (MOORE, 1976). The fibers were returned to the laboratory and ^{222}Rn was allowed to grow into Mason jars with their metal lids modified with inlet and outlet valves. We compared ^{222}Rn ingrowth from water left standing in the glass carboys and from the untreated fibers in Mason jars and obtained no statistically significant difference.

Sediment ^{222}Rn and ^{226}Ra analysis

Sediment samples were stripped in glass Mason jars identical to the ones used for the fiber analysis, but with a known amount of water added to create a slurry. Distilled, deionized water was used to form the slurry in 1982; it had a very low ^{226}Ra background but the sediment core segments did not attain secular equilibrium between ^{222}Rn and ^{226}Ra with depth. The slurry was formed with seawater in 1983 after passing through a Mn-impregnated fiber filter to remove ^{226}Ra . The resulting sediment core segments attained secular equilibrium within the accuracy of the ^{226}Ra analysis. The 1979 grab samples and 1983 core were processed with Ra-free seawater. The grab samples were collected only for determining ^{226}Ra levels at the sediment surface, so shipboard ^{222}Rn extractions were necessary only for the cores. After the analysis was complete the sediments were dried in an oven at 70°C to determine their dry weight.

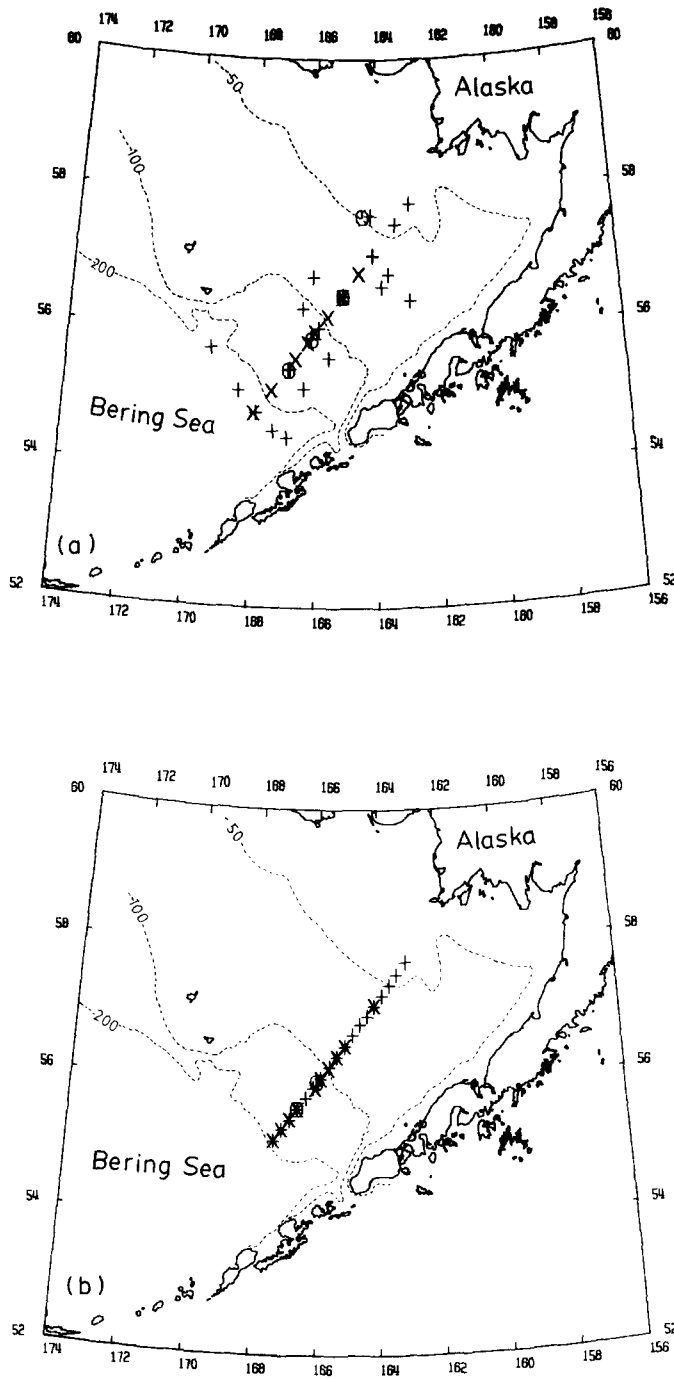


Fig. 2. (a) Radon station locations from cruises in 1980 (○), 1981 (+) and 1982 (×). Several station locations overlay each other. (b) Sediment sampling sites. Benthos corer samples are shown by (○), 1979 grab samples by (+) and 1981 grab samples by (×). Isobaths shown are 50, 100 and 200 m depths.

Alpha scintillation counting

Radon-222 activity was measured using two dual channel alpha scintillation counters (Applied Techniques Co). The counting cells were constructed either from Pyrex glass or from quartz tubes and coated on the inside with silver-activated ZnS. The counting cells were counted for 2000 min or 2000 counts, whichever was shorter. Background histories were kept for the counting cells, stripping carboys, extraction boards and radon ingrowth jars. Counting cell efficiencies were monitored by periodically stripping sealed ^{226}Ra standards (480.8 dpm). The calculations and statistics for each sample were determined as in SARMIENTO *et al.* (1976). The overall accuracy and precision (1σ) of the system is approximately $\pm 10\%$.

RESULTS

Cross-shelf distribution of ^{222}Rn and ^{226}Ra

Figure 3 are cross-shelf sections of total ^{222}Rn (Fig. 3a), ^{226}Ra (Fig. 3b), excess ^{222}Rn (Fig. 3c) and σ , (Fig. 3d) from a 2 month period. To avoid confusion due to the possibility of interannual variability only 1981 data were used. Stations not on the PROBES main station line were projected along isobaths to one section since along-shelf variability is small compared to cross-shelf variability (COACHMAN and WALSH, 1981; SCHUMACHER and KINDER, 1983; COACHMAN, 1986). The measured data were recast into a regular grid with universal kriging (OLEA, 1974) and contoured by SURFACE-II (SAMPSON, 1978) with a piecewise Bessel interpolation smoothing routine. The regular grid was retained for subsequent two-dimensional modeling. Figure 4 shows three representative measured ^{222}Rn and ^{226}Ra profiles, one from each hydrographic domain.

The cross-section of total ^{222}Rn (Fig. 3a) showed a distribution of ^{222}Rn indicative of two-dimensional mixing. The isopleths of ^{226}Ra across the shelf (Fig. 3b) increased towards the sediments and showed an intrusion of ^{226}Ra -rich water onto the shelf from off the shelf. The cross-section of excess ^{222}Rn (Fig. 3c) reflected the features shown in Fig. 1; namely near-surface deficiencies and higher activities of ^{222}Rn near the sediment interface. Shoaling of the deficiency layer was evident in both the section (Fig. 3c) and the representative station profiles (Fig. 4). Radon-222 and ^{226}Ra distributions in the vicinity of the middle front (IVERSON *et al.*, 1979) (30–60 m depth and 327.5 km offshore in Figs 3a–c) suggested the exchange of outer and off-shelf waters with middle shelf waters. We suspect that the ^{222}Rn depleted waters beneath waters containing excess ^{222}Rn (Figs 3c and 4a) reflected the presence of finestructure reported in COACHMAN and CHARNELL (1979) and COACHMAN (1986) and suggested ventilation of near-bottom waters along isopycnal surfaces that intersect both the bottom and the surface (Fig. 3d).

Gas transfer coefficients

The flux of ^{222}Rn across the air–sea interface can be calculated from the size of the radon deficiency in the upper water column (PENG *et al.*, 1979). The transfer coefficient is modeled on a balance of the radon production, decay and evasion to the atmosphere (EMERSON *et al.*, 1973). This one-dimensional model considers flux from below or from the side negligible. The transfer coefficient, J (cm s^{-1}), is calculated from the following equation:

$$J = D_w/Z = \lambda \bar{h}(A_{\text{Ra}}/A_{\text{Rn}} - 1), \quad (1)$$

where D_w is the radon diffusion coefficient in water ($\text{cm}^2 \text{s}^{-1}$) corrected for temperature; λ is the radon decay constant ($2.1 \times 10^{-6} \text{s}^{-1}$); \bar{h} is the integrated ^{222}Rn deficiency divided by the surface deficiency, i.e. the average integrated depth of radon deficiency (cm); A_{Ra} is ^{226}Ra surface activity; and A_{Rn} is ^{222}Rn surface activity. The value reported as Z in Table 1 is the hypothetical stagnant film thickness which is given by

$$Z = D_w/J. \quad (2)$$

Hypothetical film thicknesses and transfer coefficients from 1980, 1981 and 1982 are presented in Table 1. The film thickness from Sta. TT159-4103 was large relative to the

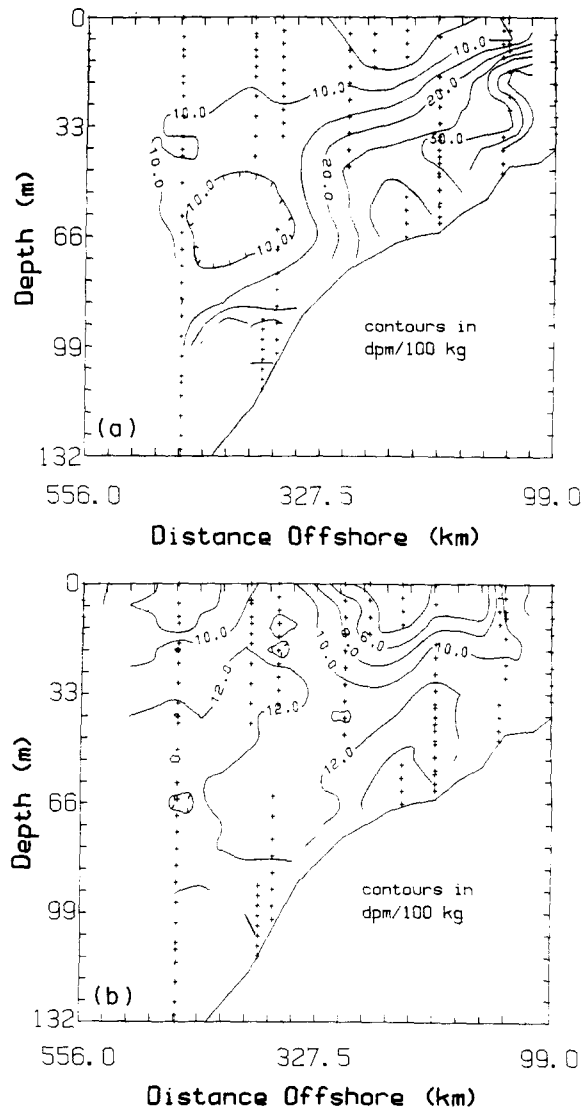


Fig. 3a,b.

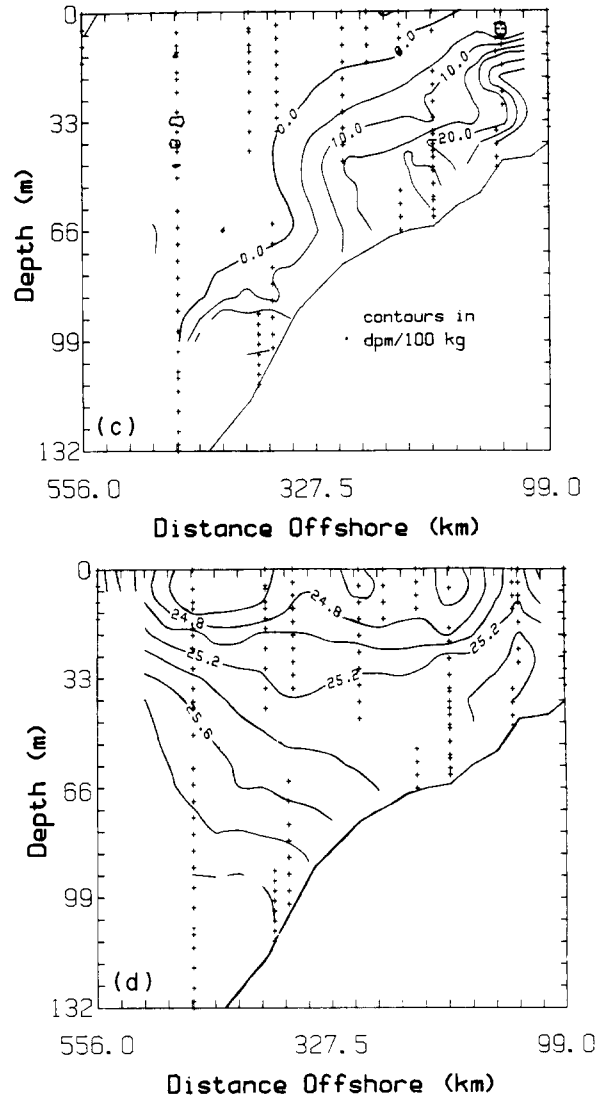


Fig. 3. (a) Cross-shelf section of total ^{222}Rn . Samples are from June–July 1981. Stations are projected along isobaths to form a single section. Contours are in dpm/100 kg. Crosses indicate sample depths. (b) Cross-shelf section of ^{226}Ra . Section description same as (a). (c) Cross-shelf section of excess ^{222}Rn . Section description same as (a). (d) Cross-shelf section of σ_t . Contours are in kg m^{-3} . The remainder of section description same as (a).

range of values reported in BROECKER and PENG (1982). Additionally, Fig. 5 suggests that the gas exchange rate decreased as the water column shoaled. This appeared to be due to flux overlap between radon gas leaving the water column at the surface and radon gas leaving the sediments at the bottom of a shallow water column. Excluding these anomalies, the average transfer coefficients were $2.2 \pm 0.4 \text{ m d}^{-1}$ in 1980 ($n = 2$), $3.3 \pm 1.1 \text{ m d}^{-1}$ in 1981 ($n = 6$) and 2.6 m d^{-1} in 1982 ($n = 1$).

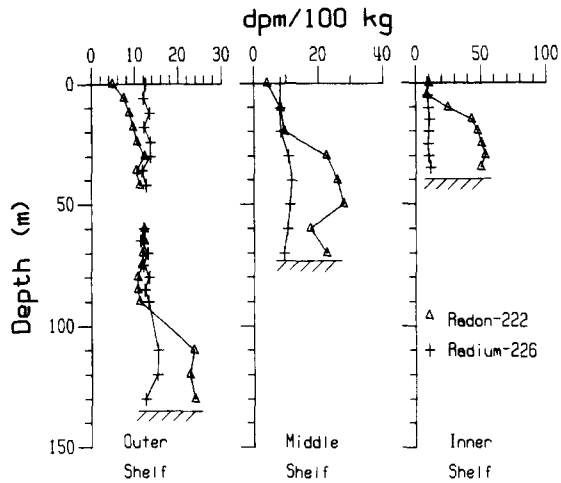


Fig. 4. Representative ^{222}Rn and ^{226}Ra depth distributions from each of the hydrographic domains in the PROBES area. (a) Outer shelf: Stas TT159-3115 and TT159-3119, 135 m. (b) Middle shelf: Sta. HX028-0041, 73 m. (c) Inner shelf: Sta. TT159-4021, 40 m.

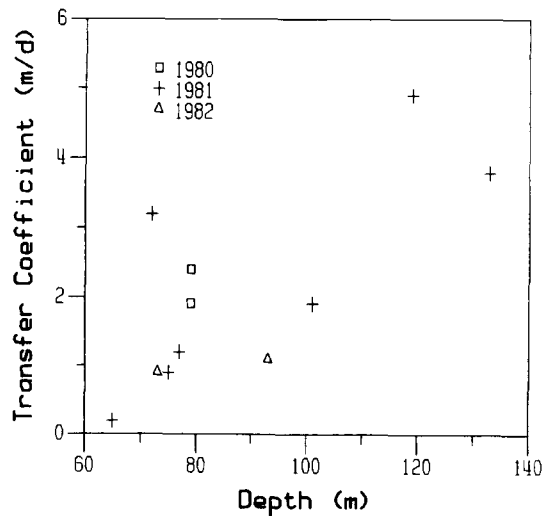


Fig. 5. Depth of water column vs transfer coefficient from 1980, 1981 and 1982. As the water column shoaled the transfer coefficient decreased.

Table 1. Gas transfer coefficients

Station	Date	N Lat.	W Long.	Depth (m)	Domain	Mixed layer depth (m)	H-Bar (m)	Z (μm)	J (m day^{-1})
HX009-0012	6.10.80	56 32.1	165 08.5	79	Middle	51	26	39	1.9
HX009-0031	9.10.80	56 32.1	165 08.5	79	Middle	51	151	30	2.4
TT159-3042	5.06.81	56 22.1	166 27.1	101	Middle	20	81	38	1.9
TT159-4033	3.07.81	56 28.0	162 55.0	77	Middle	17	7	64	1.2
TT159-4068	11.07.81	56 33.1	165 08.2	75	Middle	8	34	88	0.9
TT159-4093	15.07.81	56 41.0	163 51.0	72	Middle	12	38	25	3.2
TT159-4103	16.07.81	57 08.1	164 09.0	65	Middle	15	2	400	0.2
HX028-0041	26.06.82	56 52.0	164 37.0	73	Middle	5	5	78	0.9
HX028-0112	3.07.82	56 15.2	165 38.0	93	Middle	29	40	64	1.1
TT159-3119	18.06.81	55 56.6	166 08.7	119	Outer	16	17	16	4.9
TT159-4083	14.07.81	55 12.0	166 23.0	133	Outer	13	26	21	3.8
TT159-3103	15.06.81	54 51.0	167 53.0	1440	Off	12	13	34	2.2
TT159-4124	19.07.81	55 46.0	169 22.1	1829	Off	12	9	21	3.8
HX028-0030	24.06.82	54 50.3	167 57.4	1500+	Off	6	16	27	2.6

Near-bottom mixing rates

A steady-state model (BROECKER, 1974) assuming no lateral transport of radon was applied to the near-bottom excess ^{222}Rn profiles. An exponential decay curve was fit to the near-bottom excess ^{222}Rn distribution by a non-linear, least squares cubic regression (YORK, 1966). With this regression routine the error associated with each radon measurement and each depth measurement was incorporated into the calculations. The equation fitted is of the form:

$$\text{Rn}_y = \text{Rn}_o e^{-ay}, \quad (3)$$

where Rn_y is the excess ^{222}Rn activity (dpm/100 kg) at depth y ; Rn_o is the excess ^{222}Rn activity (dpm/100 kg) at the sediment-seawater interface; $a = \sqrt{\lambda/K_v}$, with K_v = vertical eddy diffusivity ($\text{m}^2 \text{s}^{-1}$), λ = radon decay constant ($2.1 \times 10^{-6} \text{ s}^{-1}$); and y is depth above bottom (m). A propagation of errors calculation was used to estimate the uncertainty (shown as sigma in Table 2) in the vertical eddy diffusivity. A correlation coefficient for each least squares cubic fit was also calculated (correlation coefficients statistically significant at the 95% confidence level are marked with an asterisk in Table 2).

Table 2 presents near-bottom vertical eddy diffusivities from 1980, 1981 and 1982. Of the statistically significant values reported, two were from the inner shelf, two from the middle shelf and two from the outer shelf domain. After propagation of errors, the significant values were (± 1 S.D.) $17.9 \pm 6.6 \text{ cm}^2 \text{ s}^{-1}$ for the inner shelf, $541 \pm 3080 \text{ cm}^2 \text{ s}^{-1}$ for the middle shelf and $759 \pm 6600 \text{ cm}^2 \text{ s}^{-1}$ for the outer shelf domain. It should be noted that the model used to obtain these vertical eddy diffusivities did not consider lateral mixing terms and in our opinion overestimated these values. We believe this overestimation resulted from excess ^{222}Rn being transported horizontally into the water column. Nevertheless, these values were considered upper limits in the development of a two-dimensional model.

The horizontal transport of ^{222}Rn was accounted for with a two-dimensional mixing model (GLOVER, 1985) of the cross-shelf distributions of total ^{222}Rn , ^{226}Ra and σ_t , shown in Fig. 3. The development of this two-dimensional model is the subject of another

Table 2. Near-bottom vertical eddy diffusivities

Station	Date	N Lat.	W Long.	Depth (m)	Domain	No. samples	K (cm ² s ⁻¹)	Sigma	r
HX009-0056	16.10.80	57° 41.8'	164° 28.5'	50	Inner	5	25.3	8.07	-0.962*
TT159-3033	4.06.81	57° 43.3'	165° 12.0'	48	Inner	6	10.5	4.68	-0.944*
TT159-4021	30.06.81	57° 52.9'	162° 53.2'	40	Inner	6	11.9	7.91	-0.786
HX009-0012	6.10.80	56° 32.1'	165° 08.5'	79	Middle	3	71.4	18.3	-0.932
TT159-3066	9.06.81	56° 51.9'	163° 37.9'	67	Middle	4	999	6550	+0.099
TT159-3071	10.06.81	57° 08.0'	164° 09.0'	65	Middle	4	173	472	-0.562
TT159-4025	1.07.81	57° 08.0'	164° 09.0'	64	Middle	5	130	113	-0.696
TT159-4103	16.07.81	57° 08.1'	165° 09.0'	65	Middle	7	353	1750	-0.709
HX028-0010	21.06.82	56° 32.5'	165° 08.7'	80	Middle	6	12.5	9.42	-0.856*
HX028-0041	26.06.82	56° 52.0'	164° 37.0'	73	Middle	7	1070	4360	-0.817*
HX028-0112	3.07.82	56° 15.2'	165° 38.0'	93	Middle	9	790	938	-0.526
HX009-0005	4.10.80	55° 28.4'	166° 53.1'	139	Outer	6	7260	16100	-0.445
HX009-0008	5.10.80	55° 55.3'	166° 07.5'	121	Outer	6	1490	9330	-0.838*
TT159-3058	8.06.81	55° 39.0'	165° 35.9'	112	Outer	8	5170	38700	-0.185
TT159-3115	17.06.81	55° 29.0'	166° 54.0'	133	Outer	3	62.8	109	-0.688
TT159-4055	8.07.81	55° 29.0'	166° 54.0'	133	Outer	8	469	1260	-0.401
TT159-4112	17.07.81	56° 05.0'	165° 54.0'	104	Outer	7	27.1	11.4	-0.879*
HX028-0046	28.07.82	55° 10.0'	167° 24.0'	160	Outer	4	37.1	84.1	-0.497
HX028-0096	1.07.82	55° 38.0'	166° 40.0'	133	Outer	5	139	1540	-0.312
TT159-3050	6.06.81	55° 10.1'	168° 26.9'	1682	Off	4	15.7	5.82	-0.872

* Significant at 95% level.

paper; however, some results from it are presented here. The equation used to describe the steady-state distribution of ²²²Rn on the southeastern Bering Sea shelf is:

$$\partial C/\partial t = 0 = K_H(\partial^2 C/\partial x^2) + K_V(\partial^2 C/\partial z^2) + P - \lambda C, \quad (4)$$

where C is ²²²Rn concentration (atoms cm⁻³); P is ²²²Rn production rate (atoms s⁻¹ cm⁻³); λ is the ²²²Rn decay constant (2.1×10^{-6} s⁻¹); K_H is apparent horizontal eddy diffusivity (cm² s⁻¹); and K_V is apparent vertical eddy diffusivity (cm² s⁻¹). A numerical finite differencing approximation of this differential equation was fit to the uniform grid of ²²²Rn obtained from the universal kriging exercise. The study area was divided into subregions according to temperature and density gradients and Dirichlet boundary conditions were used. The fit was accomplished by an optimization search of the chi-square (χ^2) "hypersurface" described by K_H and K_V variations (BEVINGTON, 1969) with the "best fit" defined as the global χ^2 minimum. The resultant statistically significant fits to the data gave horizontal eddy diffusivities of 10⁶-10⁸ cm² s⁻¹ and vertical eddy diffusivities of 0.5-5 cm² s⁻¹ depending on the region of the water column investigated (upper vs lower water column).

Sediment ²²²Rn flux

The profiles of ²²²Rn and ²²⁶Ra for the cores taken in 1982 (HX028-0120) and 1983 (HX048-0014) are shown in Fig. 6. Biological irrigation rates were calculated for these cores using a model similar to those applied by SMETHIE *et al.* (1981) and HAMMOND and FULLER (1979) and are shown as V_B in Fig. 6. The model assumed that the radon flux out of the sediments was due to molecular diffusion and biological irrigation. The pertinent equation is:

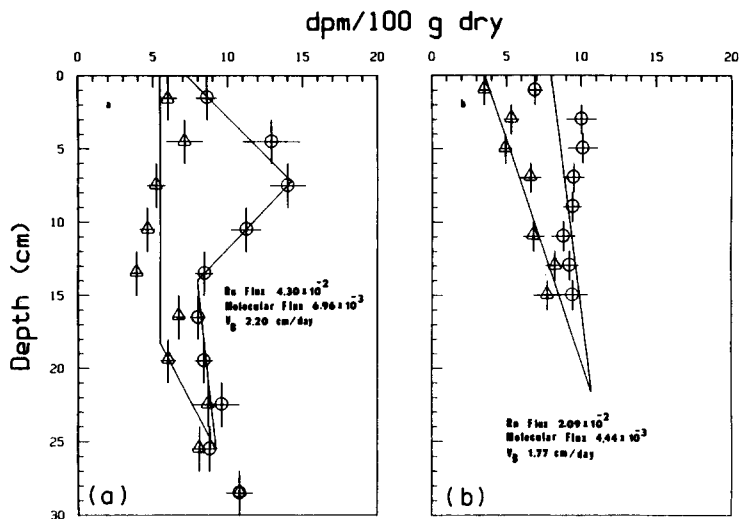


Fig. 6. ^{222}Rn and ^{226}Ra depth distributions in sediment cores. The actual flux exceeds the molecular diffusive flux ($\text{atoms s}^{-1} \text{cm}^{-2}$). (a) Station HX028-0120, 110 m. (b) Station HX048-0014, 129 m. Δ , ^{222}Rn ; \circ , ^{226}Ra .

$$V_B = \frac{\Sigma D - D_T[(\Delta[\text{Rn}_s]/\Delta x)]_o}{[\text{Rn}_{iw}^o]}, \quad (5)$$

where V_B is the biological irrigation rate (cm s^{-1}); ΣD is integrated radon deficiency in the core ($\text{atoms s}^{-1} \text{cm}^{-2}$); D_T is radon diffusivity in the sediment ($\text{cm}^2 \text{s}^{-1}$) corrected for temperature and tortuosity (SMETHIE *et al.*, 1981); $[(\Delta[\text{Rn}_s]/\Delta x)]_o$ is radon concentration gradient in wet sediment (atoms cm^{-4}) at the sediment-seawater interface; and $[\text{Rn}_{iw}^o]$ is radon concentration in the interstitial water 1 cm below the interface (atoms cm^{-3}). Unlike the cores examined by SMETHIE *et al.* (1981), there were no homogeneous zones of ^{210}Pb in our cores (GLOVER, 1985), so only one zone of deficiency was assumed. Since the 1981 core segments (Fig. 6a) did not attain secular equilibrium with depth, the ^{226}Ra profile was adjusted by multiplying each data point by the $^{222}\text{Rn}:^{226}\text{Ra}$ ratio of the deepest core segment as in SMETHIE *et al.* (1981). The line segments in Fig. 6 smoothed the data and their intersection determined the base of the radon deficiency. These line segments were also fitted with the least squares cubic routine of YORK (1966).

Estimates of ^{222}Rn flux from the sediments were given directly from the standing crop depth integration of excess ^{222}Rn in the water column. Figure 7a shows standing crop vs distance offshore for all appropriate near-bottom profiles measured. There is a statistically significant linear trend to this data ($n = 20$, $r = -0.70$) at the 99.9% confidence level. These data suggested that the flux of ^{222}Rn from the sediments was increasing in the onshore direction.

Given undisturbed sediments, the molecular diffusion of ^{222}Rn would be the only flux of ^{222}Rn from the sediments. The molecular fluxes of ^{222}Rn shown in Fig. 7b were calculated by applying the model of IMBODEN and JOLLER (1984) to the sediment surface

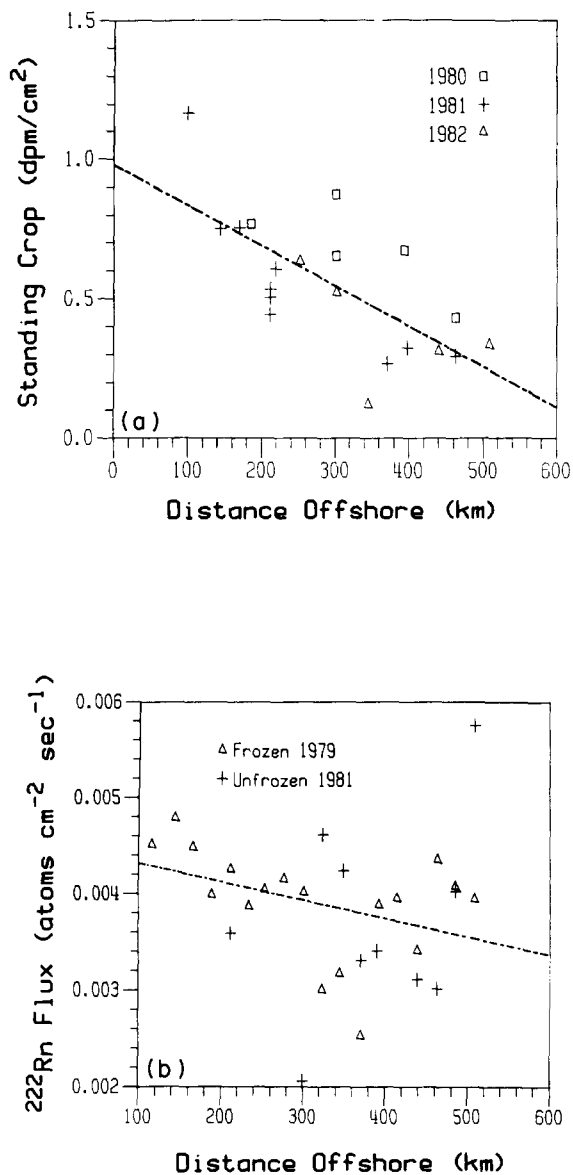


Fig. 7. (a) Distance offshore vs excess ^{222}Rn standing crop in the water column. The dot-dash line shown is statistically significant at the 99.9% confidence level ($r = -0.70$, $n = 20$). (b) Distance offshore vs molecular diffusive flux of ^{222}Rn from sediment. Flux was estimated from grab sample ^{222}Rn production rates and porosities. The dot-dash line shown is statistically significant at the 90% confidence level for only the 1979 samples stored frozen ($r = -0.41$, $n = 18$); the unfrozen 1981 samples may have had unreliable porosity estimates due to desiccation.

grab samples collected in this study. The flux, F (atoms $\text{s}^{-1} \text{cm}^{-2}$), was calculated from the following equation:

$$F = (1 - \phi)\rho_s S_E \sqrt{D_T \lambda}, \quad (6)$$

where ϕ is sediment porosity; ρ_s is sediment density (2.64 g cm^{-3}); S_E is sediment ^{222}Rn production rate (atoms $\text{g}^{-1} \text{dry}$); D_T is molecular diffusivity of ^{222}Rn ($\text{cm}^2 \text{s}^{-1}$), corrected for temperature and tortuosity (SMETHIE *et al.*, 1981); and λ is the ^{222}Rn decay constant ($2.1 \times 10^{-6} \text{ s}^{-1}$). Molecular diffusive flux of ^{222}Rn slightly increased in the onshore direction (Fig. 7b). This increase is statistically significant at the 90% confidence level for samples stored frozen ($n = 18$, $r = -0.41$); these were the lowest ^{222}Rn flux estimates obtained.

DISCUSSION

Gas transfer coefficients

The processes controlling gas exchange at the air-sea interface (LISS, 1983) have received a great deal of attention recently. Theoretical studies postulate various relationships between wind speed and transfer coefficient; BRITKO and KABEL (1978) propose a square root relationship, DEACON (1977) proposes a linear relationship. Laboratory investigations have yielded other possible mathematical relationships between wind speed and transfer coefficient. KANWISHER (1963) obtains a quadratic relationship while BROECKER *et al.* (1978) find a bilinear relationship in their wind tunnel experiments. However, field studies fail to find any kind of consistent correlation between wind speed and transfer coefficient (PENG *et al.*, 1979; HASSE and LISS, 1980; BROECKER and PENG, 1982; LISS, 1983). Empirical relationships are found (HARTMAN and HAMMOND, 1984; WANNINKHOF *et al.*, 1985; SMETHIE *et al.*, 1985) for specific study areas. Since the mechanism of gas transfer is poorly understood (HOLMÉN and LISS, 1984), SMETHIE *et al.* (1985) argue that the radon method is the only method capable of establishing the relationship between average wind speed and transfer coefficient applicable to field studies in the ocean.

The wind velocity (at 10 m elevation) was recorded on the bridge of the R.V. *T.G. Thompson* every 2 h during the 1981 cruise. Figure 8 shows an 80 day period of wind speeds and transfer coefficients derived from radon distributions suitable for calculating gas exchange rates during the 1981 field season. Wind speed gradients in the study area were estimated from surface pressure maps (NIEBAUER, 1982) for the 80 day period of Fig. 8; in the worst case (24 May 1981, day 144 of Fig. 8) they indicated a gradient of $\sim 1.2 \times 10^{-2} \text{ m s}^{-1} \text{ km}^{-1}$ along the PROBES line, generally the gradients were much lower ($\sim 2.5 \times 10^{-3} \text{ m s}^{-1} \text{ km}^{-1}$). Chauvenet's criterion was applied to the nine 1981 gas exchange parameters and the six remaining values were used in the analysis that follows.

A regression optimization analysis was conducted to determine the optimal duration for averaging the wind speed that gave the best least squares correlation between mean wind speed and transfer coefficient. Starting at the time of sample collection this optimization program considered wind speeds at 2 h intervals up to 20 days prior to radon sample collection. Figure 9 shows the optimum averaging periods for wind speeds vs transfer coefficients from the linear regression optimization (Figs 9a and b), the correlation coefficient spectra for all the averaging periods (Fig. 9c) and the autocorrelation spectra of the wind speeds (Fig. 9d). As seen in Fig. 9c the correlation coefficient

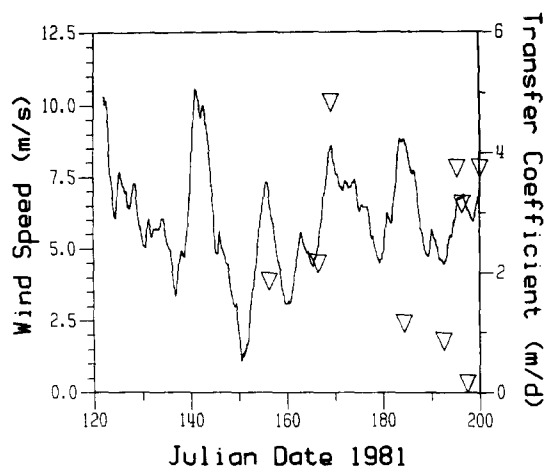


Fig. 8. Day of year, 1981 vs wind speed and transfer coefficient in the PROBES area. Wind speeds were averaged over 96 h prior to sampling. Wind speed is the solid line; transfer coefficients are the inverted triangles.

has two distinct peaks significant at the 95% confidence level, the first at 2.9 days and the second at 9.7 days. An autocorrelation analysis (Fig. 9d) revealed the synoptic time scale of the wind speeds was between 2.0 and 2.5 days. We concluded that longer averaging periods did not improve the relationship between wind speed and transfer coefficient.

There are at least five caveats with the application of ^{222}Rn deficit profiles to gas exchange rate determination in this study. LISS (1983) suggests that the "shape" of the radon deficiency profile may change with changing wind conditions. An increase in mixed layer depth with increasing wind speed may cause the radon deficiency to become deeper and less wide. In terms of the model in this paper, \bar{h} became larger but $A_{\text{Ra}}/A_{\text{Rn}}$ became smaller and they tended to cancel. This may have overestimated the gas transfer coefficient. Second, ^{222}Rn , with a decay half-life of 3.83 days, may not respond quickly enough to the wind conditions to be useful except in situations with relatively constant wind velocity (BROECKER and PENG, 1971; PENG *et al.*, 1979; DEACON, 1981). Third, isopycnal surfaces in the outer and off-shelf domains intersect both the surface and bottom (Fig. 3d); however, a time scaling argument showed that along isopycnal venting of these near-bottom waters did not substantially violate the no-bottom-flux assumption of the one-dimensional gas exchange model. Fourth, the second peak in the correlation coefficient spectra (Fig. 9c) may have been generated by the choice of a linear weighting function to average the wind speeds; a weighting function that decayed, backwards in time, with a half-life of ^{222}Rn may have been more appropriate. Finally, HOLMÉN and LISS (1984) present the results of an experiment designed to distinguish between the stagnant surface film, surface replacement and boundary layer models of gas exchange. They present evidence that the stagnant film model not only seems physically unrealistic, but that it is not the model indicated by the experimentally determined relationship between transfer coefficient and molecular diffusivity. Nevertheless, they cannot determine whether the film replacement or the boundary layer model is correct due to uncertainties in the molecular diffusivities used in their experiments. Because of the above, the

average film thickness reported here can only be considered conceptually and the transfer coefficient was a better measure of gas exchange since it was model independent.

Film thickness, however, was useful for computing annual gas exchange budgets and possibly seasonal budgets. Four methods were used in PROBES to estimate the CO_2 gas exchange rate: (1) ^{222}Rn deficiency measurements and infrared $p\text{CO}_2$ measurements of the atmosphere and seawater (CODISPOTI *et al.*, 1986), (2) net productivity and $^{15}\text{NO}_3$ uptake experiments (R. SAMBROTTO, personal communication), (3) wind speed records

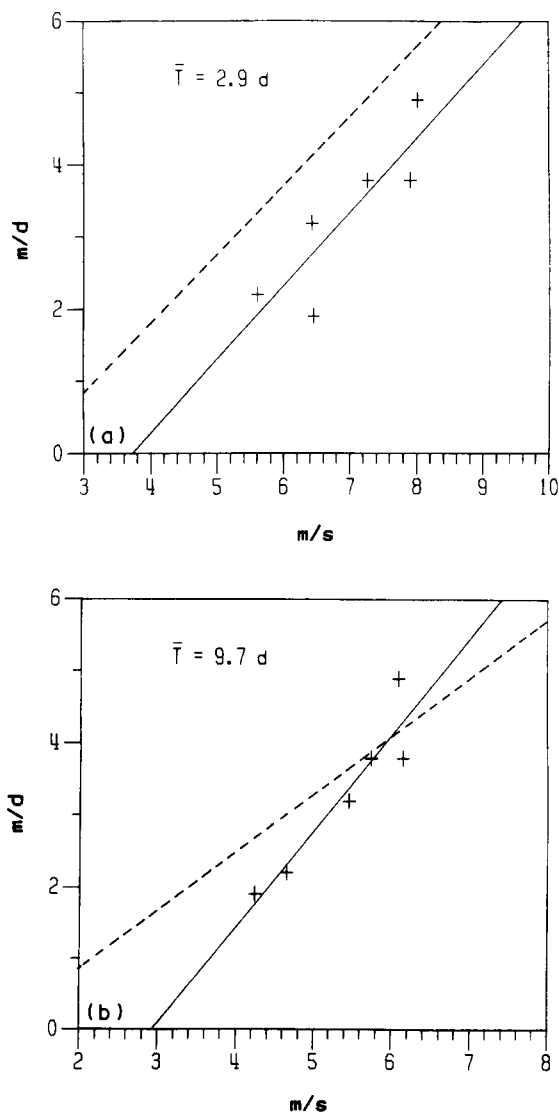


Fig. 9a,b.

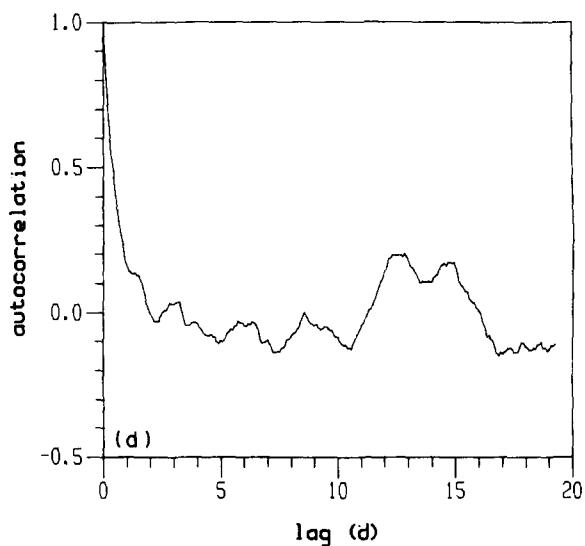
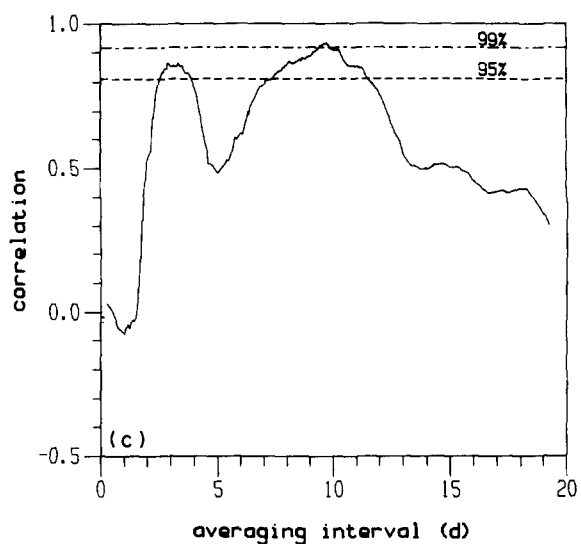


Fig. 9. Linear regression optimization analysis of averaged wind speed vs transfer coefficient. Winds were averaged over (a) 2.9 days and (b) 9.7 days. Data points and wind tunnel results normalized to $Sc = 1800$ for ^{222}Rn at 8.5°C ($Sc = \text{Schmidt number} = \text{kinematic viscosity}/\text{molecular diffusivity}$) by assuming transfer coefficient proportional to $Sc^{-1/2}$. Wind speed vs transfer coefficient relationship of BROECKER *et al.* (1978) shown as a dashed line. (c) Correlation coefficient spectra for averaging periods from 2 h to 20 days. Confidence levels of 95 and 99% are shown. (d) Autocorrelation coefficient spectra of wind speeds from 2 h to 20 days. Synoptic time scale of wind speeds was between 2.0 and 2.5 days.

and the relationship given by BROECKER *et al.* (1978) coupled with the $p\text{CO}_2$ measurements of CODISPOTI *et al.* (1986), and (4) a floating chamber in which the evolution of $p\text{CO}_2$ was monitored (D. HOOD, personal communication). Methods 1 and 3 used in PROBES averaged to about the same gas exchange rate over approximately month-long periods (Table 3), suggesting that the radon-derived film thicknesses have use as long-term averages.

Near-bottom mixing rates

Applying the one-dimensional, vertical eddy diffusion model (BROECKER, 1965; BROECKER *et al.*, 1968) to the near-bottom excess ^{222}Rn distributions measured in this study gave only six statistically significant fits (see Table 2). Horizontal transport of ^{222}Rn has been invoked in the past to explain non-linearities in the transformed data. SMETHIE (1981) includes rapid horizontal mixing of radon from the sides of the fjords to improve the fit to his data. BROECKER and KAUFMAN (1970) find maxima in their near-bottom profiles that are explained as horizontal inputs of excess ^{222}Rn from surrounding "hills". LIETZKE and LERMAN (1975) use a two-dimensional numerical model to describe the distribution of ^{228}Ra on the eastern United States continental shelf. A similar approach for ^{222}Rn in a lake is used by IMBODEN and EMERSON (1978) and in an offshore basin by BERELSON *et al.* (1982). Horizontal and vertical eddy diffusivities have been calculated based on salt balance arguments; horizontal eddy diffusivities were found to be 5–6 orders of magnitude larger than vertical eddy diffusivities (COACHMAN and WALSH, 1981; COACHMAN, 1986). The one-dimensional model assumed the radon could only be transported vertically in the water column. Horizontal fluxes of ^{222}Rn were important on the southeastern Bering Sea shelf because where the bottom slope was steep enough, any horizontal gradient of radon caused a horizontal flux of radon in the water column. This horizontally added ^{222}Rn caused the one-dimensional model to overestimate the vertical eddy diffusivity. Our statistically significant outer and inner shelf values for vertical eddy diffusivity were about a factor of seven and two higher than that of COACHMAN (1986), implying horizontal flux was important there; our significant middle shelf values were approximately three orders of magnitude higher, indicating that horizontal transport was very important in this domain. SCHUMACHER and KINDER (1983) suggest that interaction

Table 3. Comparison of methods of estimating CO_2 gas exchange

	Radon deficiency	Net productivity	$p\text{CO}_2$ + wind	Surface chamber
1980 $\Delta p\text{CO}_2 = -169 \mu\text{atm}$				
Max.	1.50	7.90	–	1.8
Min.	0.18	0	–	0.32
Avg.	0.46	2.2	0.5	1.03
1981 $\Delta p\text{CO}_2 = -93 \mu\text{atm}$				
Max.	0.83	2.9	0.57	–
Min.	0.10	0.59	0.29	–
Avg.	0.25	1.58	0.43	–

Net productivity values from R. SAMBROTTO (personal communication).

Surface chamber values from D. HOOD (personal communication).

$p\text{CO}_2$ + wind values from CODISPOTI *et al.* (1985).

Values reported are CO_2 flux in $\text{g C m}^{-2} \text{d}^{-1}$.

between bottom slope variation and tidal action drives horizontal mixing on this shelf; our data were consistent with this notion.

Applying a two-dimensional, diffusion–decay model to the cross-shelf distribution of ^{222}Rn (GLOVER, 1985) yielded two levels of statistical significance. The upper water column fits were statistically significant at the 95% confidence level. The horizontal and vertical eddy diffusivities (10^6 and $0.5 \text{ cm}^2 \text{ s}^{-1}$, respectively) were in close agreement with the values reported by COACHMAN (1986). The near-bottom horizontal and vertical eddy diffusivities were 10^8 and $5 \text{ cm}^2 \text{ s}^{-1}$, respectively, and significant at the 50% confidence level. Because the χ^2 statistic described the dispersion of observed frequencies about expected frequencies there was no positive or negative answer to the χ^2 test. Results statistically significant at the 50% confidence level implied that the two-dimensional distribution of ^{222}Rn fit to the observed distribution was better than a random distribution at least half the time. These horizontal and vertical eddy diffusivities were two and one order of magnitude greater than COACHMAN's (1986) estimates, respectively. The vertical eddy diffusivities from the two-dimensional model were one to three orders of magnitude smaller than the one-dimensional model estimates presented above.

Sediment ^{222}Rn flux

We have found that molecular diffusive ^{222}Rn flux did not account for the total sediment ^{222}Rn flux across the entire southeastern Bering Sea shelf. Molecular diffusive ^{222}Rn flux out of the sediments in two cores from the outer shelf accounted for about 20% of the total ^{222}Rn flux (Fig. 6). The total ^{222}Rn flux, as given by the standing crop, and the molecular diffusive ^{222}Rn flux, as given by the grab samples, were estimated across the study area by integrating the straightline fit to the corresponding data set. The average excess ^{222}Rn standing crop calculated in this manner was 0.5 dpm cm^{-2} while the average excess ^{222}Rn standing crop predicted from molecular diffusion alone was 0.2 dpm cm^{-2} . The integrated molecular diffusive ^{222}Rn flux accounted for about 40% of the integrated total ^{222}Rn flux from the sediments. Although this was a very crude approximation of sediment ^{222}Rn flux of the entire shelf, it indicated that molecular diffusive ^{222}Rn flux out of the sediments was insufficient to explain the total amount of ^{222}Rn in the water column and another process(es) must be occurring.

Several processes could be responsible for the flux of radon out of the outer shelf sediments: molecular diffusion, bioturbation, physical mixing, bio-irrigation, physical irrigation and chemical processes (SMETHIE *et al.*, 1981). Molecular diffusion (6.96×10^{-3} and $4.44 \times 10^{-3} \text{ atoms s}^{-1} \text{ cm}^{-2}$, see Fig. 6) was too small to account for the deficiency seen in the two cores. Bioturbation and physical mixing of sediment particles were disregarded based on the appearance of ^{210}Pb profiles for our sediment cores (GLOVER, 1985) and other cores from the PROBES area (BANAHAN and GOERING, 1986). Bioturbation can be present without homogenizing ^{210}Pb (KIPPHUT, 1978), but in that case bioturbation would be too slow to affect sediment ^{222}Rn profiles (IMBODEN and STILLER, 1982). Physical irrigation (wave pumping) was also disregarded because of the small effect this process has at these depths ($>100 \text{ m}$) (RIEDL *et al.*, 1972). The chemical process of radon stripping by methane ebullition (KIPPHUT and MARTENS, 1982) was discounted due to the oxic condition of the sediments. The remaining process, bio-irrigation, appeared to be the most likely possibility because a rather large community of suspension-feeding infauna inhabit this shelf (HAFLINGER, 1978).

Cross-shelf distribution of ^{222}Rn and ^{226}Ra

The cross-shelf sections of total ^{222}Rn , ^{226}Ra , excess ^{222}Rn and σ_t , shown in Fig. 3 are in reality 2-month-long "time exposures". Through this time-averaged view of these distributions, several processes were identified.

The contour plots of ^{222}Rn (total and excess) revealed some interesting features (Figs 3a and c). The shoaling of the near-surface ^{222}Rn deficiency resulted from the overlap of the two exchange fluxes of ^{222}Rn on the shelf. The shallow waters of the shelf allowed the excess ^{222}Rn flux out of the sediments to impinge upon the ^{222}Rn flux out of the water column into the atmosphere. The mid-depth waters of the outer shelf have been identified as a zone of finestructure mixing on the southeastern Bering Sea shelf (COACHMAN, 1986). The presence of a closed contour line surrounding this area (Fig. 3a) implied some sort of interruption to the general from-sediment-to-atmosphere flux of ^{222}Rn on the shelf. Additionally, the presence of ^{222}Rn -depleted waters below waters containing excess ^{222}Rn (Figs 3c and 4a) suggested ventilation of near-bottom waters along isopycnal surfaces (Fig. 3d) in the finestructure region of the shelf. COACHMAN (1982) proposes a weak flow convergence in the middle of this broad continental shelf driving the finestructure mixing, but the physics of this process is still unclear. Along with other data (Figs 7a and b) the contours of ^{222}Rn implied an increasing source of ^{222}Rn from the sediments in the onshore direction. SHARMA (1979) reports increasing average grain size and we measured decreasing sediment porosity going onshore. Increased permeability (LERMAN, 1979) may have allowed more of the ^{222}Rn produced within the inner shelf sediments to escape into the overlying water column.

The contour plot of ^{226}Ra also showed some interesting features (Fig. 3b). If ^{226}Ra was derived only from the sediments, then the isopleths of ^{226}Ra would be parallel to the bottom topography. If ^{226}Ra was solely terrestrially derived, then the isopleths of ^{226}Ra would slope away from the shore and intersect the bottom. However, the heterogeneous appearance of the ^{226}Ra activity field and the appearance of a tongue of ^{226}Ra -rich water injecting itself onto the shelf implied that there was another source of ^{226}Ra from off the shelf. The deep off-shelf waters had much higher ^{226}Ra activities (GLOVER, 1985) consistent with this idea. A sink of ^{226}Ra was also indicated in the near-surface waters of the middle shelf by a depression in activity centered at 247.5 km offshore (^{226}Ra activity decreased from ~ 10 to ~ 2 dpm/100 kg). This area of the shelf is reported to have high diatom activity (SAMBROTTO *et al.*, 1986). Figure 10 is a plot of soluble silica concentration vs ^{226}Ra activity for all ^{226}Ra samples measured on the shelf in 1981. The statistically significant linear relationship at the 95% confidence level ($r = 0.66$, $n = 153$) implies an association between silica and ^{226}Ra as found by others (BROECKER *et al.*, 1976; KU and LIN, 1976; CHUNG, 1980; KU *et al.*, 1980).

CONCLUSIONS

We believe that the shoaling landward of the near-surface radon deficiency layer, the standing crop of excess ^{222}Rn increasing landward and the deficiency of ^{222}Rn in the mid-depths of the outer shelf resulted from the interaction between baroclinic flows, tidal currents and bottom slope variations as proposed by SCHUMACHER and KINDER (1983). The variations in the bottom slope occur approximately beneath the hydrographic fronts and their interaction with tidal currents and baroclinic flows are believed to be generating the fronts that divide the shelf into the three regions (SCHUMACHER and KINDER, 1983;

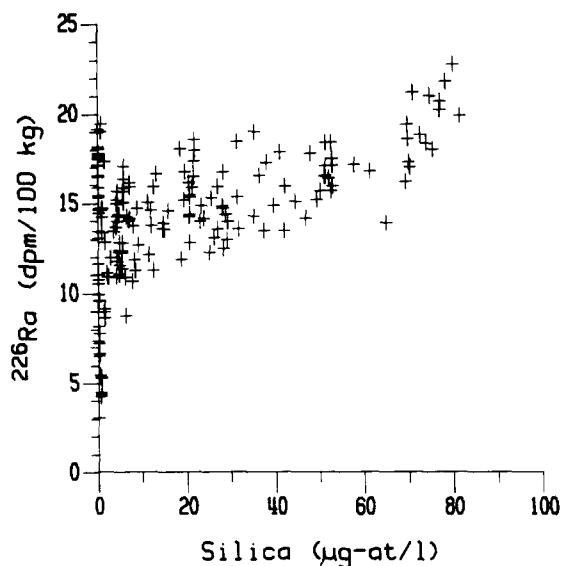


Fig. 10. Soluble silica concentration vs ^{226}Ra activity in the PROBES area, 1981. A correlation coefficient of 0.66 obtained from 153 data points is statistically significant at the 95% confidence level.

COACHMAN, 1986). The following features were seen in the "time exposure" cross-shelf sections (Fig. 3): near-surface ^{222}Rn deficiency due to gas exchange, two-dimensional mixing of ^{222}Rn in the water column, ^{222}Rn deficiency beneath ^{222}Rn excess, biological removal of ^{226}Ra and an apparent higher sediment ^{222}Rn flux on the inner shelf. We concluded the following from this investigation into using ^{222}Rn and ^{226}Ra as tracers of mixing intensity and gas exchange on the southeastern Bering Sea shelf:

(1) The loss of excess ^{222}Rn to the atmosphere was obscured in the shallower water column due to overlap of benthic and near-surface fluxes.

(2) Like other workers, we found an empirical linear relationship between radon-derived transfer coefficients and wind speed if stations exhibiting benthic flux overlap were omitted. The mean film thickness reflected the average gas exchange conditions as shown from agreement with the estimates of CODISPOTI *et al.* (1986).

(3) The overestimation of the vertical eddy diffusivity by the one-dimensional, vertical flux model was caused by the horizontal flux of ^{222}Rn . This flux resulted from the interaction of the horizontal gradient of ^{222}Rn , tidal action, bottom slope variability and baroclinic flow.

(4) ^{222}Rn deficient waters were found beneath waters containing excess ^{222}Rn in the same region of the shelf waters as the finestructure layering of COACHMAN and CHARNELL (1979) and where isopycnal surfaces that intersect both the bottom and surface were found. Radon-222 was one more property that correlated with the presence of finestructure layering.

(5) A two-dimensional, steady-state, diffusion-decay mixing model gave statistically significant fits to the ^{222}Rn data collected on the southeastern Bering Sea shelf during the summer of 1981.

(6) Molecular diffusion was insufficient to account for the ^{222}Rn deficiency in sediment cores taken on the shelf and the excess ^{222}Rn in the water column. The most likely mechanism responsible for the ^{222}Rn deficiency in the sediments was bio-irrigation ($\sim 2 \text{ cm d}^{-1}$).

(7) ^{226}Ra activity was removed in a region of known high biological activity and showed a statistically significant positive correlation with dissolved silica.

(8) The ^{226}Ra activity field showed an heterogeneous nature and strongly suggested a flux of ^{226}Ra enriched water from the deep Bering Sea basin onto the shelf. We concluded that the ^{226}Ra on the shelf was not terrestrially derived.

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