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Excess ^{210}Po in the coastal atmosphere

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ABSTRACT

Many researchers have reported the widespread occurrence of excess ^{210}Po in the global atmosphere and suggested probable sources such as resuspension of top soils, stratospheric aerosols, sea spray of the surface micro-layer, volcanic emission, and bio-volatile ^{210}Po species from the productive ocean. We have observed excess ^{210}Po on aerosols in the coastal atmosphere of the Chesapeake and Delaware Bays. On-board measurements in the Chesapeake Bay atmosphere show that the increase of this excess ^{210}Po is dependent upon wind speed. Simultaneously measured activity ratios of ^{7}Be/^{210}Pb and ^{210}Pb/^{222}Rn argue against either higher altitude air or continental soils as the source of this excess. We hypothesize that the excess ^{210}Po originates mainly from surface waters either by the sea-spray of the surface microlayer, or more likely, by gas exchange. We conclude gas exchange as the mechanism since the polonium excess increases linearly with wind speed over a threshold of 3 m s^{-1} (mean) similar to other gases (i.e., CO_2, SF_6, and DMS). In addition, higher ^{210}Po excess with lower ^{222}Rn is observed in on-shore marine air at Lewes, DE. This suggests sea-air exchange of volatile Po along with other bio-volatile species (i.e., DMS, DMSe, and MMHg) in the coastal productive ocean during high wind speeds.

1. Introduction

As gaseous ^{222}Rn ($t_{1/2} = 3.8$ days) diffuses from earth's surface soils into the lower atmosphere, its daughters ^{210}Pb ($t_{1/2} = 22.3$ years), ^{210}Bi ($t_{1/2} = 5.01$ days), and ^{210}Po ($t_{1/2} = 138$ days) are continuously produced by the radioactive decay in situ and removed via dry and wet deposition. Although these ^{222}Rn daughters are useful for estimating tropospheric aerosol residence times due to their reactivity, the aerosol residence times based on ^{210}Po/^{210}Pb ratios (10–300 days) are thought to be largely overestimated compared with those based on other tracers such as a ^{210}Bi/^{210}Pb model (< 10 days) (Poet et al., 1972; Turekian et al., 1977; Balkanski et al., 1993; Hussain et al., 1998). This leads one to search for another source of ^{210}Po in the troposphere.

Sources of excess ^{210}Po into the atmosphere have been hypothesized by several workers. These include resuspension of humus or fine soil fraction of the regolith component (Vilenskii, 1970), mixing of older stratospheric aerosol (Poet et al., 1972; Tsunogai and Fukuda, 1974; Tokieda et al., 1996), secretion from phytoplankton concentrated in the surface microlayer and subsequent spray from the oceanic surface layer (Turekian et al., 1974, 1977; Bacon and Elzermann, 1980), volcanic emissions (Lambert et al., 1982) and a possible volatilization of alkyl polonium species from the oceans (Hussain et al., 1995). Hussain et al. (1991) reported a summer time activity ratio of ^{210}Po/^{210}Pb in aerosols in and around the mid-Atlantic region nearly 3 times higher than that during fall and winter, indicative of an additional

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^{210}Po source to the marine environment during summer time.

On the other hand, ^{7}Be ($t_{1/2} = 56$ days) is produced in the stratosphere and upper troposphere and often used as a tracer of upper altitude sources such as ozone, stratospheric bomb fallout debris, and stratospherically injected volcanic emissions. Due to differences in source terms for ^{210}Pb and ^{7}Be, simultaneous measurements of these two nuclides can provide a powerful tracer for vertical air mass mixing. For example, the ^{7}Be/^{210}Pb ratio is indicative of vertical mixing between the upper atmosphere and the middle and lower troposphere (Baskaran, 1995; Tokieda et al., 1996; Kim, 1998) and of an upper tropospheric source for ozone (Graustein and Turekian, 1996).

While the levels and sources of ^{7}Be, ^{222}Rn and ^{210}Pb in the atmosphere are well known, the sources of excess ^{210}Po are still not certain. The objective in this study was to assess the origin of excess ^{210}Po in the coastal atmosphere. We measured ^{210}Po, along with its grand parent ^{210}Pb, the cosmogenic ^{7}Be (high altitude source), and ^{222}Rn (continental ground source), in the surface air from the upper Chesapeake Bay and evaluated its dependence on wind speed. Comparison is also made with similar on-shore results from Lewes, DE on the lower Delaware Bay.

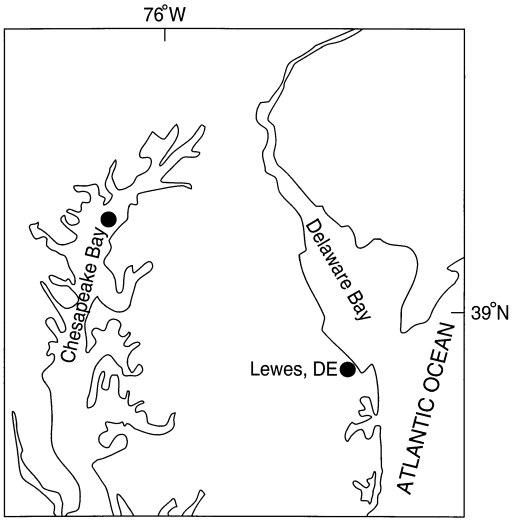

Fig. 1. Location of the sampling stations. The Chesapeake Bay aerosol samples were collected onboard SV Anderson during 10–24 August 1995. The Lewes aerosol samples were collected on top the roof of Cannon lab at the College of Marine Studies, Lewes, DE during 11 July–6 August 1995 (~12 m from the ground).

2. Materials and methods

The ^{7}Be and ^{226}Ra daughters (^{222}Rn, ^{210}Pb, and ^{210}Po) were measured in aerosol and air samples collected at a fixed station in the Chesapeake Bay during 10–24 August 1995 (Fig. 1). The time of these measurements provided a good opportunity for this study since oceanic sulfur recycling, along with polonium, should follow the spring-summer plankton and bacterial blooms very effectively. Also, the continuous measurement at a fixed station reduces possible spatial variation of ^{210}Po from local sources. During the sampling period, there was only one small rain event (~0.5 cm) on the 15th August. The sample collection was discontinued on the 17th when Hurricane Felix threatened the study area, although the storm did not impact the study area.

The activity of ^{222}Rn in air was measured using an automated radon counter based on the design of Larson and Bressan (1978). Daily aerosol samples were collected by filtering nearly 400 m^3 air through glass fiber filters (Saunders et al., 1968), which were used in the radon counter (Andreae et al., 1991). These filters were later digested and analyzed for ^{210}Po, ^{210}Pb, and ^{7}Be following standard procedures (Church et al., 1994). The ^{210}Po sources were counted via their alpha activities on a silicon barrier detector coupled to a multichannel analyzer, whereas the activities of ^{210}Pb and ^{7}Be were measured using an intrinsic germanium co-axial detector (Hussain et al., 1996). The ^{210}Pb activities were corroborated by remilking ^{210}Po after 6 months (alpha counting results are shown in Table 1). The ^{210}Pb activities based on both methods agreed within counting errors. Appropriate reagent blank and background corrections were applied which remained within 20% of the ^{210}Pb signals and 5% of ^{7}Be signals. Results are presented in Table 1. Similar measurements were also carried out on aerosols and air collected on the roof of Cannon Lab, Lewes, Delaware (Fig. 1) from 11 July–6 August 1995.

Table 1. *^{222}Rn, ^{210}Pb, ^{210}Po, and ^{7}Be contents in the air and aerosol of Chesapeake Bay during 10–24 August 1995*

Sample ID	Sampling date	^{222}Rn (dpm/m^3) average	^{222}Rn (dpm/m^3) STD*	^{210}Pb† (dpm/100 m^3)	^{210}Po† (dpm/100 m^3)	^{7}Be† (dpm/100 m^3)	^{7}Be/^{210}Pb† (AR)
C10	10	64	47	2.49 ± 0.05	0.32 ± 0.02	37.7 ± 1.6	15.1 ± 0.7
C11	11	110	63	2.48 ± 0.07	0.18 ± 0.02	35.1 ± 1.9	14.1 ± 0.9
C12	12	122	78	1.61 ± 0.05	0.13 ± 0.01	22.9 ± 1.3	14.2 ± 0.9
C13	13	348	87	4.57 ± 0.16	0.22 ± 0.02	24.8 ± 1.7	5.4 ± 0.4
C14	14	320	169	4.01 ± 0.06	0.32 ± 0.02	30.3 ± 1.7	7.6 ± 0.5
C15	15	395	111	6.79 ± 0.24	0.37 ± 0.02	31.1 ± 1.4	4.6 ± 0.3
C16	16	239	82	4.43 ± 0.07	0.17 ± 0.01	25.8 ± 1.1	5.8 ± 0.3
C18	18	394	131	6.49 ± 0.20	1.48 ± 0.12	57.0 ± 2.8	8.8 ± 0.5
C19	19	155	53	3.67 ± 0.11	0.72 ± 0.02	48.5 ± 2.1	13.2 ± 0.7
C20	20	92	36	3.81 ± 0.25	0.45 ± 0.01	78.0 ± 2.6	20.5 ± 1.5
C21	21	396	122	5.25 ± 0.12	0.49 ± 0.01	55.5 ± 2.9	10.6 ± 0.6
C22	22	547	222	8.47 ± 0.31	0.58 ± 0.01	63.8 ± 2.5	7.5 ± 0.4
C23	23	204	56	2.18 ± 0.09	0.63 ± 0.02	6.0 ± 1.0	2.8 ± 0.5
C24	24	419	149	4.88 ± 0.27	0.69 ± 0.02	22.2 ± 1.7	4.5 ± 0.4

* Standard deviation based on 18 observations per day.
† Errors are based on 1σ counting statistics.

3. Results and discussions

During the sampling period, different air masses displayed characteristically different specific activities of ^{222}Rn on either an hourly or daily basis (Table 1). During 10–12th and 19–20th, relatively lower ^{222}Rn content air (mean 60–150 dpm/m^3) was encountered, which was much lower than the mean ^{222}Rn concentrations observed in the eastern US air with a mean activity of about 440 dpm/m^3 (Genthon and Armengaud, 1995). Isentropic air mass back trajectories of 72 hours showed those air masses to be primarily of marine origin for the preceding 72 hours (J. Keeler, UMI, per. comm.).

The values of ^{7}Be/^{210}Pb ratios range from 2–20 at lower altitudes and reach approximately 5000 in the stratosphere (Graustein and Turekian, 1996). Longitudinal variation of this ratio is much smaller, with the annual mean ratio of 7 and 10 for the eastern US and Bermuda, respectively (Graustein and Turekian, 1996). Since our ^{7}Be/^{210}Pb ratios ranged between 5–20 (Table 1), the stratospheric contribution to the air sampled in this study appears to be negligible. This decreases the possibility of ^{210}Po inputs from the stratosphere.

The residence times of aerosols using a steady state ^{210}Pb/^{222}Rn model can be calculated by assuming a constant supply of ^{210}Pb from ^{222}Rn and constant scavenging of ^{210}Pb by aerosols. In this study, a steady-state assumption seems to be valid considering a good correlation ($r = 0.87$) between ^{210}Pb and ^{222}Rn (Fig. 2a). At steady state, the aerosol residence time (τ) based on the activity ratio (R) can be calculated using equation (1):

$$\tau = \frac{R}{(1-R)\lambda_{Pb}}, \quad (1)$$

where λ_{Pb} is the decay constant of ^{210}Pb. We calculated the aerosol residence times based on the ^{210}Pb/^{222}Rn model to be 1.3–4.5 days. This is lower than the values reported using ^{210}Bi/^{210}Pb (mean 5 days) for variable tropospheric air-masses (Turekian et al., 1977). In contrast, much longer residence times (12–90 days) are calculated using ^{210}Po/^{210}Pb ratios (Fig. 2b), similar to previous results (Hussain et al., 1998). The equation for the ^{210}Po/^{210}Pb model can be found elsewhere (Poet et al., 1972). These apparently high residence times are caused by direct inputs of ^{210}Po from secondary sources (defined to be 'excess') over ^{210}Bi decay products as discussed earlier. The magnitude of such sources is significantly larger than what could be accounted for by the errors in the ^{210}Pb/^{222}Rn based aerosol residence times, since the general values of the aerosol residence times are only on the order of a few days (Turekian et al., 1977; Hussain et al., 1998).

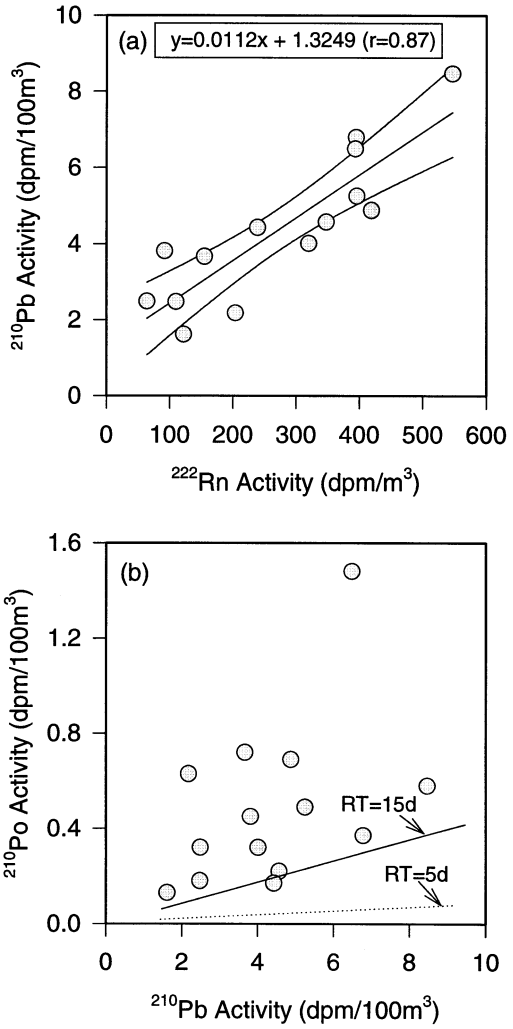

Fig. 2. Plots of (a) ^{210}Pb versus ^{222}Rn and (b) ^{210}Po versus ^{210}Pb activities for aerosol samples collected in Chesapeake Bay during 10–24 August 1995. The measured activities of ^{210}Po show uniformly large excesses with an exceptional residence time of over 5 days, suggesting sources of excess ^{210}Po in the region.

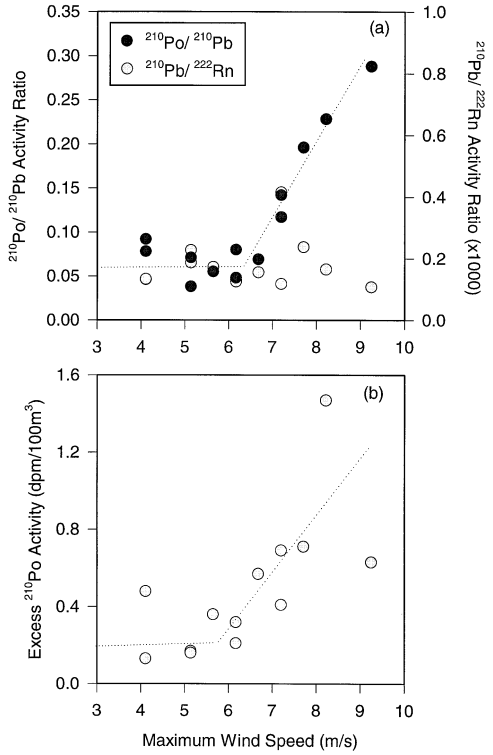

Fig. 3. Plots of maximum wind speed versus (a) ^{210}Po/^{210}Pb activity ratio and (b) excess ^{210}Po activity for aerosol samples collected in Chesapeake Bay during 10–24 August 1995. Wind speeds were measured on board hourly, which are absolute since the ship was at anchor. Excess ^{210}Po activity is calculated by subtracting the decay product ^{210}Po (calculated from the ^{210}Pb/^{222}Rn based residence time of aerosols) from the in-situ measured ^{210}Po. In general, the decay production is negligible relative to excess ^{210}Po.

Assuming that the source of excess ^{210}Po in the Chesapeake Bay environment is from the local waters, one would expect wind speed, thus water turbulence, to affect atmospheric concentrations (Fig. 3). Both, the ^{210}Po/^{210}Pb ratios and excess ^{210}Po show increases with increasing maximum wind speeds, over a threshold of 6 m/s (Fig. 3). This increase of ^{210}Po/^{210}Pb ratios may result from the resuspension of surrounding soils, with relatively high ^{210}Po/^{210}Pb ratios ($\geqslant 1$). However, if soils are the source of the excess ^{210}Po in the atmosphere, one would also expect an increase in ^{210}Pb. Measurements of the ^{210}Pb/^{222}Rn ratios do not show any increase with wind speed (Fig. 3a). In addition, ^{226}Ra in the aerosol samples was not detectable, suggesting no appreciable soil inputs of either ^{210}Po or ^{210}Pb. Based on just this observation, resuspension of the top soil as a source of excess atmospheric ^{210}Po appears unlikely in the Chesapeake Bay environment.

Two possible mechanisms which can continuously supply excess ^{210}Po to the coastal ocean

atmosphere by wind effects can be 1) sea spray of surface micro-layer which has high ^{210}Po enrichment (Bacon and Elzermann, 1980), and 2) bio-volatilization of ^{210}Po from the coastal ocean (Hussain et al., 1995). Excess ^{210}Po versus wind speed shows a characteristic threshold (Fig. 3b) such that excess Po starts to increase at a mean wind speed of 3 m/s (max 6 m/s). This is a unique feature of gas exchange between water and air as the threshold is caused by breaking of capillary waves at the wind speed of 3 m/s (Nguyen et al., 1984; Liss and Merlivat, 1986; Wanninkhof, 1992; Turner et al., 1996). Such a trend of excess ^{210}Po versus wind speed is reminiscent of that observed for other marine biogenic gases (e.g., DMS). Thus it suggests that the source of excess ^{210}Po could be from surrounding coastal seawater by gaseous air-sea exchange of volatile biogenic species (e.g., dimethyl polonide).

The potential formation of bio-volatile species of Po in this region is also supported by the measurements of ^{210}Po and ^{210}Pb in Delaware Bay (Church and Sarin, 1995). Their studies showed concentrations of ^{210}Po and ^{210}Pb ranging between 0.2–2.2 and 0.01–0.11 dpm/100 l, respectively, in the surface waters, over the salinity range of 0–30 ppt. The higher ^{210}Po/^{210}Pb ratios (AR = 1.0–6.5) suggest strong ^{210}Po re-mineralization by bacterial activity following efficient uptake by marine phytoplankton in this area. The biogeochemical conditions in this large estuarine and shelf region of the mid-Atlantic Bight are favorable for the production of volatile species of the other oxygen group elements, such as DMS, DMSe, DMTe, and possibly DMPo (Hussain et al., 1995).

It is instructive to estimate the extent of volatile ^{210}Po in seawater in order to check whether such an amount can be supported by gas exchange. The amounts of volatile ^{210}Po ($C_{Po\text{-}sw}$) in seawater can be calculated using the following equation:

$$C_{Po\text{–}sw} = \frac{C_{Po\text{–}atm} H}{k\tau}, \quad (2)$$

where $C_{Po\text{–}atm}$ is excess ^{210}Po measured in the atmosphere of the Chesapeake Bay, H is the height of the atmospheric mixed layer for ^{210}Po, k is the gas exchange velocity of volatile ^{210}Po, and τ is the residence time of ^{210}Po in the atmosphere. To calculate the most reasonable value of $C_{Po\text{–}sw}$, one can take the excess ^{210}Po from the measured excess ^{210}Po activity in the air during this study

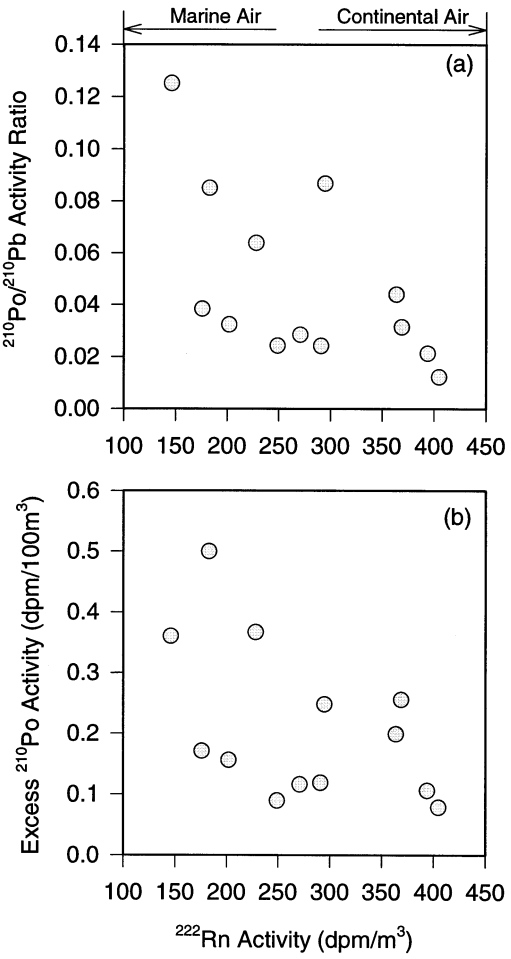

Fig. 4. Plots of ^{222}Rn versus (a) ^{210}Po/^{210}Pb activity ratio and (b) excess ^{210}Po activity for aerosol samples collected from a laboratory roof, at Lewes, DE.

(0.6 dpm/100m^3), lower tropospheric mixed layer as 1000 meters based on published profiles of ^{222}Rn, DMS, and meteorological parameters at 40°N (Brost and Chatfield, 1989; Watanabe et al., 1995), the mean aerosol residence time (3 days) based on the ^{210}Pb/^{222}Rn model, and a value of 8 cm/h for k based on DMS studies at wind speed of 7 m/s (Turner et al., 1996). Based on these values, the volatile ^{210}Po in seawater is calculated to be 0.1 dpm/100 l using eq. (2).

Thus, less than 10% of total ^{210}Po in Chesapeake or Delaware Bay is required to be volatile species to account for the excess ^{210}Po in

the coastal atmosphere. Although this calculation is based on parameters which have some uncertainties (e.g., the k for volatile Po species is assumed to be the same as that for DMS), it still supports the basic concept for the mechanism of Po volatilization and subsequent air-sea exchange. Also, a summer time activity ratio of ^{210}Po/^{210}Pb in the aerosols following the spring phytoplankton bloom in and around the mid-Atlantic region is nearly $3\times$ higher than that in the winter (0.08 ± 0.02). Though the winter time winds are also strong (Hussain et al., 1993), absence of a corresponding high ^{210}Po/^{210}Pb ratio on the aerosol during winter signifies the importance of biological productivity (in addition to wind speed) in contributing to the observed excess ^{210}Po in the atmosphere.

If the main source of the atmospheric ^{210}Po is from the coastal productive ocean, marine air that moves over the continent should show higher excess ^{210}Po relative to air masses that develop primarily over land. In Fig. 4, the measured ^{210}Po/^{210}Pb activity ratios and excess ^{210}Po from on-shore, Lewes, DE, are plotted against ^{222}Rn activities. Since ^{222}Rn emission from the ocean is negligible compared with that from the continents, two arbitrary regions based on ^{222}Rn contents are identified in Fig. 4 as "marine air" and "continental air". Here it is observed that the marine air contains higher ^{210}Po relative to continental air. This study therefore suggests that like DMS, the excess ^{210}Po activity in coastal atmospheres depends upon both, the biological productivity of the surface ocean, as well as wind speeds.

4. Acknowledgments

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REFERENCES

Andreae, T. W., Cutter, G. A., Hussain, N., Radford-Knoery, J. and Andreae, M. O. 1991. Hydrogen sulfide and radon in and over the western North Atlantic Ocean. *J. Geophys. Res.* **96**, 18753–18760.

Bacon, M. P. and Elzerman, A. W. 1980. Enrichment of ^{210}Pb and ^{210}Po in the sea-surface microlayer. *Nature* **284**, 332–334.

Balkanski, Y. J., Jacob, D. J., Gardner, G. M., Graustein, W. C. and Turekian, K. K. 1993. Transport and residence times of troposphere aerosols inferred from a global three-dimensional simulation of ^{210}Pb. *J. Geophys. Res.* **98**, 20573–20586.

Baskaran, M. 1995. A search for the seasonal variability on the depositional fluxes of ^{7}Be and ^{210}Pb. *J. Geophys. Res.* **100**, 2833–2840.

Brost, R. A. and Chatfield, R. B. 1989. Transport of radon in a three-dimensional, subhemispheric model. *J. Geophys. Res.* **94**, 5095–5119.

Church, T. M. and Sarin, M. M. 1995. Natural U–Th series radionuclide studies of esturine processes. In: *Changes in fluxes in estuaries: implications from science to management* (ed. K. R. Dyer and R. J. Orth). Proceedings from ECSA22/ERF Symposium, Plymouth, England, September 13–18, 1992, 91–95.

Church, T. M., Hussain, N., Ferdelman, T. G. and Fowler, S. W. 1994. An efficient quantitative technique for the simultaneous analyses of radon daughters ^{210}Pb, ^{210}Bi and ^{210}Po. *Talanta* **41**, 243–249.

Genthon, C. and Armengaud, A. 1995. Radon 222 as a comparative tracer of transport and mixing in two general circulation models of the atmosphere. *J. Geophys. Res.* **100**, 2849–2866.

Graustein, W. C. and Turekian, K. K. 1996. ^{7}Be and ^{210}Pb indicate an upper troposphere source for elevated ozone in the summertime subtropical free troposphere of the eastern North Atlantic. *Geophys. Res. Lett.* **23**, 539–542.

Hussain, N., Church, T. M. and Burnett, W. C. 1993. Volatile polonium in Floridan groundwater. AGU Abstract H22D-5. Fall Meeting, San Francisco.

Hussain, N., Ferdelman, T. G., Church, T. M. and Luther III, G. W. 1995. Bio-volatilization of Polonium: results from laboratory analyses. *Aquatic Geochem.* **1**, 175–188.

Hussain, N., Kim, G., Church, T. M. and Carey, W. L. 1996. A simplified technique for gamma spectroscopic analysis of ^{210}Pb in sediment samples. *App. Rad. Iso.* **47**, 473–477.

Hussain, N., Church, T. M., Veron, A. J. and Larson, R. E. 1998. Radon daughter disequilibria and lead systematics in the western North Atlantic. *J. Geophys. Res.* **103** (D3), 16059–16071.

Kim, G. 1998. Atmospheric inputs and upper ocean biogeochemistry of trace elements and radionuclide tracers in the Atlantic, PhD dissertation, University of Delaware, USA.

Lambert, G., Ardouin, B. and Polian, G. 1982. Volcanic output of long-lived radon daughter. *J. Geophys. Res.* **87**, 11103-11108.

Larson, R. E. and Bressan, D. J. 1978. Automatic radon counter for continual unattended operation. *Rev. Sci. Instrum.* **49**, 965–969.

Liss, P. S. and Merlivat, L. 1986. Air–sea gas exchange rates: introduction and synthesis. In: *The rôle of air–sea exchange in geochemical cycling* (ed. P. Buat-Menard). D. Reidel, Hingham, Mass, 113–129.

Moore, H. E., Poet, S. E. and Martell, E. A. 1973. ^{222}Rn, ^{210}Pb, ^{210}Bi and ^{210}Po profiles and aerosol residence times versus altitude. *J. Geophys. Res.* **78**, 7065–7075.

Nguyen, B. C., Bergeret, C. and Lambert, G. 1984. Exchange rates of dimethyl sulfide between ocean and atmosphere. In: *Gas transfer at water surfaces* (ed. W. Brutsaert and G. H. Jirka), 539–545.

Poet, S. E., Moore, H. E. and Martell, E. A. 1972. Lead 210, Bismuth 210 and Polonium 210 in the atmosphere: accurate ratio measurement and application to aerosol residence time determination. *J. Geophys. Res.* **77**, 6515–6527.

Saunders, A. W., Patterson, R. L. Jr. and Lockhart, L. B. Jr. 1968. *The interaction of radon decay products with aerosols.* Naval Research Laboratory Report 6802, 21 pp.

Tokieda, T., Yamanaka, K., Harada, K., and Tsunogai, S. 1996. Seasonal variations of residence time and upper atmospheric contribution of aerosols studied with Pb-210, Bi-210, Po-210 and Be-7. *Tellus* **48B**, 690–702.

Tsunogai, S. and Fukuda, K. 1974. ^{210}Pb, ^{210}Bi, and ^{210}Po in meteoric precipitation and the residence time of tropospheric aerosol. *Geochem. J.* **8**, 141–152.

Turekian, K. K., Kharkar, D. P. and Thomson, J. 1974. The fates of ^{210}Pb and ^{210}Po in the ocean surface. *J. Rech. Atm.* **8**, 639–646.

Turekian, K. K., Nozaki, Y. and Benninger, L. K. 1977. Geochemistry of atmospheric radon and radon products. *Ann. Rev. Earth Planet. Sci.* **5**, 227–255.

Turner, S. M., Malin, G., Nightingale, P. D. and Liss, P. S. 1996. Seasonal variation of dimethyl sulphide in the North Sea and an assessment of fluxes to the atmosphere. *Mar. Chem.* **54**, 245–262.

Vilenskii, V. D. 1970. The influence of natural radioactive atmospheric dust in determining the mean stay time of lead-210 in the troposphere. *Izv. Acad. Sci. USSR, Atmos. Oceanic Phys.* **6**, 307–310.

Wanninkhof, R. 1992. Relationship between wind speed and gas exchange over the ocean. *J. Geophys. Res.* **97**, 7373–7382.

Watanabe, S., Yamamoto, H. and Tsunogai, S. 1995. Relation between the concentrations of DMS in surface seawater and air in the temperate North Pacific region. *J. Atmos. Chem.* **22**, 271–283.