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Atmospheric Depositional Fluxes of ^{7}Be and ^{210}Pb at Galveston and College Station, Texas

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The bulk depositional fluxes of ^{210}Pb and ^{7}Be were measured at a coastal (Galveston) and an inland (College Station) station for about 3 years, between 1989 and 1991. The annual depositional fluxes of ^{7}Be and ^{210}Pb at Galveston during this period varied by a factor of about 2.5, between 8.9 and 23.2 disintegrations per minute (dpm) cm^{-2} yr^{-1}, with a mean of 14.7 dpm cm^{-2} yr^{-1} for ^{7}Be, and 0.67 and 1.71 dpm cm^{-2} yr^{-1}, with a mean of 1.03 dpm cm^{-2} yr^{-1} for ^{210}Pb, respectively. The precipitation-normalized ^{7}Be flux increases with increasing amount of precipitation. There is no systematic and consistent seasonal trend in the depositional fluxes for ^{7}Be or for ^{210}Pb. The volume-weighted ^{210}Pb concentrations, when normalized to the amount of precipitation, seem to be constant over the time period of this study. Four to six heavy rain events (> 5 cm) in a single day account for 20-30% of the annual deposition of ^{7}Be and ^{210}Pb. Such events account, however, for only about 4-6% of the total number of rainy days in a year. The dry depositional fluxes of these nuclides appear to be a significant fraction of the bulk depositional flux only during the months when there is very little rain. The fraction of dry to total depositional flux of ^{210}Pb appears to be higher than that of ^{7}Be. The strong positive correlation between ^{7}Be and ^{210}Pb depositional fluxes indicates that the flux of both nuclides is controlled by scavenging processes by local precipitation. This correlation also indicates that a major portion of the air masses that brings precipitation to Galveston and College Station is of continental origin. Our data therefore suggest that ^{7}Be and ^{210}Pb cannot be used as independent atmospheric tracers in our coastal station. This observation is consistent with those observed at many other continental and coastal stations.

INTRODUCTION

Beryllium 7 (half-life = 53.3 days) and lead 210 (half-life = 22.1 years) are two radionuclides which have been widely used as tracers and chronometers in aquatic and atmospheric systems [*Wogman et al.*, 1968; *Perkins et al.*, 1970; *Martell*, 1970; *Poet et al.*, 1972; *Craig et al.*, 1973; *Somayajulu and Craig*, 1976; *Young and Silver*, 1974, 1980; *Bacon et al.*, 1976; *Benninger et al.*, 1979; *Krishnaswami et al.*, 1980; *Aaboe et al.*, 1981; *Turekian et al.*, 1983; *Olsen et al.*, 1985, 1986; *Casey et al.*, 1986; *Dominik et al.*, 1989; *Santschi and Honeyman*, 1989; *Todd et al.*, 1989; *Brost et al.*, 1991; *Feichter et al.*, 1991; *Kritz et al.*, 1991; *Schuler et al.*, 1991; *Wieland et al.*, 1991]. Even though the sources of these two nuclides to the atmosphere are distinctly different, both are highly particle reactive and thus get attached to aerosols in the atmosphere soon after production.

Beryllium 7 is produced throughout the atmosphere as a product of the spallation of oxygen and nitrogen nuclei by energetic cosmic rays [*Lal et al.*, 1958]. Owing to its short mean life (76.9 days) and the longer residence time of stratospheric aerosols (about 1 year [*Kuroda et al.*, 1962]), most of the ^{7}Be nuclei that are produced in the stratosphere do not readily reach the troposphere except during spring when seasonal thinning of the tropopause takes place at midlatitudes, resulting in air exchange between stratosphere and troposphere. Since ^{7}Be is of cosmogenic origin, its flux to the Earth's surface has a latitudinal dependence [*Lal and Peters*, 1967]. Its concentration in the air increases with increasing altitude from the surface of the Earth, and its atmospheric flux to the Earth's surface should be independent of geography at any particular latitude [*Turekian et al.*, 1983]. In other words, the standing crop of ^{7}Be in the atmosphere at a particular latitude should be the same irrespective of whether it is located over the ocean or a continent.

In contrast, ^{210}Pb is produced by radioactive decay from its progenitor, ^{222}Rn. This Rn nuclide, which is a noble gas isotope in the ^{238}U decay chain, emanates primarily from land surfaces. The concentration of ^{210}Pb in the air over the continents decreases with its elevation from the ground due to a decrease in the concentration of ^{222}Rn [*Moore et al.*, 1973]. As a consequence of its continental source, the ^{222}Rn flux from soils is about 100 times higher than the oceanic ^{222}Rn flux [*Wilkening et al.*, 1975]. Therefore the standing crop of ^{210}Pb in the atmosphere strongly depends on the longitude, depending on whether it is above the ocean or a continent.

These two radionuclides with their different source functions are therefore useful in studies designed to better understand the mechanisms and rates of removal of aerosols. The activity ratio in the air, as well as in the precipitation, is expected to vary with location and time. The $^{7}Be/^{210}Pb$ activity ratios in the precipitation are useful for two purposes: (1) the variations in the ratio can be used to predict the physical behavior of some of the chemical species injected into the troposphere and (2) they could be used, in principle, to predict the elevation at which the cloud condensation takes place. For example, aerosols from the lower part of the troposphere will be enriched in ^{210}Pb over ^{7}Be, while those from the upper troposphere will be enriched in ^{7}Be. Thus qualitative information on the elevation at which the precipitation originated can, in principle, be obtained from this ratio. Precise determination of the value of the activity ratio is required to obtain this information.

Our investigation is designed to shed light on the following questions: (1) How do the annual ^{7}Be and ^{210}Pb depositional fluxes vary with varying amounts of precipitation? (2) How do the $^{7}Be/^{210}Pb$ activity ratios from individual rain events vary within the same month? (3) Do these ratios vary from season to season? (4) What are the relationships between activity ratios and amounts of precipitation? (5) Can ^{7}Be and ^{210}Pb be used as independent atmospheric tracers for oceanic and continental air? To answer these questions, we have collected bulk atmospheric precipitation samples for ^{7}Be and ^{210}Pb analysis at two sites, one in Galveston and the other in College Station, Texas. In addition, a compilation of all earlier data has been made and is compared with our data. This is one of the few studies in which the bulk deposition samples were collected and analyzed for ^{7}Be and ^{210}Pb at very close time intervals.

MATERIALS AND METHODS

Two sampling sites were chosen for this study, the roof of the Kirkham Hall (11 m above ground level) at the Mitchell campus of

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Texas A&M University, Pelican Island, Galveston (29°18'N, 94°48'W) and the roof of the Oceanography and Meteorology Building (58 m above ground level) in College Station (30°35'N, 96°22'W), Texas. Polyethylene drums of 200-L capacity and a surface area of 2800 cm^2 were used as rain collectors. Bulk deposition samples were obtained from December 1988 through February 1992 for Galveston and from June 1989 through February 1992 for College Station. Since evaporation rates in the containers were high in some of the samples, the precipitation record for Galveston was obtained from a meteorological station located about 3 km southeast of the sampling site, and for College Station it was obtained from the airport about 3 km away from the sampling site. Although during periods of individual convective storms the amounts of precipitation could be different, the integrated amounts of precipitation over the sampling period for most of our samples are likely to remain constant within a spatial distance of 3 km. A comparison of the meteorological record from the Galveston site with the record obtained from a rain gauge installed with our rain collector during the second half of the collection period indicates that most of the time the amounts of precipitation in individual rain events are essentially the same for the two sites, which are only 3 km apart. Since the rain collector was exposed to the atmosphere continuously, the sum of wet and dry fallout (equal to bulk fallout) was collected. To determine the dry fallout alone, the 200-L drum's lid (2800 cm^2, height of 3.5 cm) was deployed 3 times continuously for a finite period of time (7-14 days) when there was no rain. Since we primarily use these radionuclides as tracers in aquatic systems where deposition rates occur onto a water surface, the lid was filled with distilled water up to 1.5 cm (~1/2 of the total height).

The drums were cleaned by repeated 6 M HCl rinsings. Prior to each deployment, 1 mg of stable Be and Pb in 100 mL of 6 M HCl were added to the drum or the lid. The acid prevents adsorption of Be and Pb isotopes onto the polyethylene drum surfaces. The collectors were retrieved at monthly intervals in College Station and more frequently in Galveston. The more frequent retrieval enabled us to determine the fallout of these nuclides during thunder storms and other short-term rain events. After the collectors were emptied, the walls of the drum were rinsed with 2 L of 6 M HCl, which was then added to the sample. Following this thorough acid cleaning and a distilled water rinsing, the collectors were redeployed after the addition of dilute acid and stable Pb and Be carriers. In the first set of Galveston samples (during the first year), the samples were evaporated to about 50 mL, and the reduced volume was subsequently filtered through a 0.4-μm Nuclepore filter paper. Ferric chloride was added to this filtrate, which had a pH < 1, and then, after 1 hour of equilibration, $Fe(OH)_3$ was precipitated by adding NH_4OH. The precipitate was then filtered through a Whatman 42 filter. In all College Station samples and in the Galveston samples after the first year, known amounts of $FeCl_3$ were added and $Fe(OH)_3$ was precipitated directly from the original rainwater samples, without an evaporation step, by adding NH_4OH. The precipitates were separated by filtering through a Whatman 42 filter paper. The residues retained on the filter were washed with 50 mL deionized water and redissolved in a minimum amount of 6 M HCl. The solution was subsequently dried on a hot plate.

The dried residues were packed into 10-mL gamma counting vials and counted on a high-purity Ge well detector coupled to a Canberra S-100 multichannel analyzer. Most of the recently published reasearch has used gamma counting methods to determine the ^{210}Pb concentrations in soils and rainwater (soil profile [*Graustein and Turekian*, 1986]) [*Olsen et al.*, 1985; *Todd et al.*, 1989; *Monaghan*, 1989; *Dibb*, 1989; *Schuler et al.*, 1991]. This gamma counting method eliminates the problems of processing each sample twice (first, in situ ^{210}Po is separated from ^{210}Pb and then, after waiting for about 3-6 months, the ^{210}Po grown in from ^{210}Pb is measured). Furthermore, it involves relatively little manual labor and analytical time.

Typically, the samples were counted for about 6-24 hours, depending on the activity of ^{210}Pb in the sample, since all the samples had relatively high concentrations of ^{7}Be. The peak analysis of ^{7}Be (I = 10.3%, 477.7 keV) and ^{210}Pb (I = 4.05%, 46.5 keV) was done using SPECTRAN-AT peak analysis software (CANBERRA Company). There is no peak background in the ^{210}Pb energy range, 44.5-48.5 keV. The count rates of rainwater samples were typically between 2 and 10 counts per minute (cpm). In selected samples, the net counts obtained from the peak analysis software were compared with the peak analysis carried out manually in order to convince ourselves that both methods essentially gave the same net counts. In a suite of samples from a sediment core, ^{210}Pb concentrations were measured by alpha and gamma spectrometry. The ^{210}Pb concentrations obtained by these two different methods were in good agreement, always within ± 5% (M. Baskaran, unpublished data, 1992). The peak/Compton ratio for ^{60}Co (1332 keV) was 45.0:1.

One of the ^{7}Be standard solutions was obtained from Swiss Federal Institute of Water Resources and Water Pollution Control (EAWAG), Switzerland, but originated from the Laboratoire de Meteorologie des Rayonnements Ionisants, Gif-sur-Ivette, France, and the other from Brookhaven National Laboratory, New York. Our gamma counting equipment was calibrated with both of these ^{7}Be standards. The ^{210}Pb standard solution was obtained from Amersham Company and was calibrated with respect to a working standard which in turn was calibrated with respect to a National Institute of Standards and Technology standard. Using these standard solutions, the disintegrations per minute (dpm)/cpm conversion factors for various vial geometries were determined. This conversion factor was then used to calculate all ^{7}Be and ^{210}Pb activities. The error associated with the dpm/cpm conversion factor was always less than 1 %. The final ^{7}Be concentrations and deposition fluxes were corrected for radioactive decay from the end of sample collection to midtime of counting, as well as for the ingrowth and decay during the deployment period. The yields of chemical extraction were determined using the Pb and Be recoveries in the final $Fe(OH)_3$ precipitate measured by atomic absorption spectrometry. The overall propagated error (± 1 sigma) in the value of the final concentration and flux is estimated to be less than 5% for ^{7}Be and less than 10% for ^{210}Pb.

RESULTS AND DISCUSSION

The amount of precipitation, periods of rain collector deployment, specific concentrations, and depositional fluxes of ^{7}Be and ^{210}Pb results are given in Tables 1 and 2 for Galveston and College Station, respectively. Between December 16, 1988, and February 24, 1992, 67 bulk deposition samples and three dry-only deposition samples were collected and measured at Galveston. In College Station, between June 1989 and May 1992, 22 bulk deposition samples were collected. One bulk deposition sample was lost from both Galveston (May 17 to June 14) and College Station (March 22 to April 22, 1991) during sample collection and handling. The annual amount of precipitation in 1989, 1990, and 1991 was 103, 97, and 150 cm, respectively, at Galveston and 80, 98, and 146 cm, respectively, at College Station. The rainfall in 1991 for both Galveston and College Station was thus about 50% higher than during the other two years.

The monthly precipitation data for Galveston are plotted in Figure 1 for the years 1989-1991. Generally, the amount of precipitation is higher in the summer months. In 1991 (and also in 1992), January and February months also had relatively high rainfall. The monthly precipitation data for College Station are plotted in Figure 2. In general, the precipitation records for the two stations are similar.

Monthly and Seasonal Variations of ^{7}Be and ^{210}Pb Fluxes

At Galveston, the samples were collected more frequently between May 1991 and February 1992. In those samples which were collected in between two calendar months, the monthly flux was calculated by assuming the specific concentration during the

TABLE 1. Sampling Time Intervals, Annual Depositional Fluxes, and Concentrations of ^{7}Be and ^{210}Pb Measured in Galveston, Texas, From December 1988 Through February 1992

Collection Interval	Days in Collection Interval	Number of Rainy Days	Rainfall cm	^{7}Be dpm L^{-1}	^{210}Pb dpm L^{-1}	$^{7}Be/^{210}Pb$ Activity Ratio	^{7}Be Flux dpm cm^{-2}	^{210}Pb Flux dpm cm^{-2}
Dec. 16, 1988 to Jan. 12, 1989	28	8	12.40	83.5	6.74	12.4	13.50	1.09
Jan. 13 to Feb. 15, 1989	33	8	10.20	114.3	7.45	15.4	12.90	0.84
Feb. 16 to March 9, 1989	22	5	10.46	NM	NM	NM	NM	NM
March 10 to April 10, 1989	32	7	5.90	182.8	16.50	11.1	12.30	1.11
April 11 to May 16, 1989	36	11	8.10	138.8	4.26	32.6	11.40	0.35
May 17 to June 14, 1989	29	3	7.40	NM	NM	NM	NM	NM
June 15 to June 29, 1989	15	7	21.70	40.3	5.40	7.5	21.30	2.85
June 30 - July 19, 1989	20	6	4.90	146.5	11.70	12.5	13.10	1.05
July 20 to Sept. 7, 1989	50	12	21.90	81.3	5.07	16.0	13.00	0.81
Sept. 8 to Oct. 22, 1989	45	4	7.40	30.3	2.50	12.1	1.82	0.15
Oct. 23 to Dec. 4, 1989	43	10	8.10	142.1	14.50	9.8	9.77	1.00
Dec. 5, 1989 to Jan. 7, 1990	34	10	6.07	236.3	20.30	11.7	15.40	1.32
Jan. 8 to Feb. 12, 1990	36	12	6.45	103.7	6.47	16.0	6.78	0.42
Feb. 13 to Feb. 27, 1990	15	4	5.36	63.9	5.18	12.3	8.33	0.68
Feb. 28, 1990	1	1	6.36	46.5	4.35	10.7	108.00	10.10
March 1 to April 2, 1990	33	8	8.74	138.6	6.87	20.2	13.40	0.66
April 3 to May 6, 1990	34	7	12.10	112.8	4.91	23.0	14.70	0.64
May 7 to May 8, 1990	2	1	7.50	44.3	3.51	12.6	60.70	4.81
May 9 to June 5, 1990	28	3	1.50	289.0	18.00	16.1	5.65	0.35
June 6 to July 5, 1990	30	8	8.31	223.5	8.01	27.9	22.60	0.81
July 6 to Aug. 12, 1990	38	8	6.81	96.8	5.95	16.3	6.33	0.39
Aug. 13 to Sept. 6, 1990	25	5	1.88	233.2	14.40	16.2	6.40	0.39
Sept. 7 to Sept. 9, 1990	3	2	6.43	104.2	4.92	21.2	81.6	3.85
Sept. 10 to Sept. 13, 1990	4	4	8.57	34.8	2.03	17.1	27.20	1.59
Sept. 14 to Oct. 11, 1990	28	5	4.55	47.5	3.47	13.7	2.82	0.21
Oct. 12 to Nov. 11, 1990	30	7	5.54	124.6	7.21	17.3	8.40	0.49
Nov. 12 to Dec. 12, 1990	31	3	2.01	226.1	17.80	12.7	5.35	0.42
Dec. 13, 1990 to Jan. 13, 1991	32	11	18.40	190.6	8.48	22.5	40.00	1.78
Jan. 14 to Jan. 15, 1991	2	2	8.57	107.5	14.50	7.4	168.20	22.60
Jan. 16 to Jan. 20, 1991	5	1	6.07	70.6	12.90	5.5	31.30	5.70
Jan. 21 to Feb. 4, 1991	15	2	12.90	82.5	6.47	12.8	25.90	2.03
Feb. 5 to March 19, 1991	43	9	7.29	315.1	19.60	16.1	19.50	1.21
March 20 to April 24, 1991	36	10	6.15	230.9	33.00	7.0	14.40	2.06
April 25 to May 19, 1991	25	13	26.70	213.2	12.00	17.8	83.10	4.66
May 20, 1991	1	1	0.11	528.0	109.30	4.8	21.20	4.39

TABLE 1. (continued)

Collection Interval	Days in Collection Interval	Number of Rainy Days	Rainfall cm	^{7}Be dpm L^{-1}	^{210}Pb dpm L^{-1}	$^{7}Be/^{210}Pb$ Activity Ratio	^{7}Be Flux dpm cm^{-2}	^{210}Pb Flux dpm cm^{-2}
May 21 to May 24, 1991	4	1	4.29	62.3	8.33	7.5	24.40	3.26
May 25 to May 29, 1991	5	2	0.13	1243.0	218.10	5.7	11.80	2.07
May 30 to June 9, 1991	11	5	6.79	98.5	4.88	20.2	22.20	1.10
June 10 to June 11, 1991	2	2	0.07	1104.0	148.70	7.4	14.10	1.90
June 12 to June 19, 1991	8	4	4.93	193.8	12.70	15.2	43.60	2.86
June 20 to June 27, 1991	8	5	0.69	412.9	57.20	7.2	13.00	1.80
June 28 to July 7, 1991	10	5	5.36	158.5	7.00	22.6	31.00	1.37
July 8 to July 15, 1991	8	2	7.14	113.9	8.53	13.3	37.10	2.78
July 16 to July 21, 1991	6	1	1.79	148.8	9.64	15.5	16.20	1.05
July 22 to Aug. 7, 1991	17	4	2.31	157.3	6.13	25.7	7.80	0.30
Aug. 8 to Aug. 21, 1991	14	6	4.24	102.2	6.73	15.2	11.30	0.74
Aug. 22, 1991	1	1	1.45	110.9	16.40	6.8	58.70	8.68
Aug. 23 to Aug. 26, 1991	4	1	NM	NM	NM	13.3	7.85	0.59
Aug. 27, 1991	1	1	0.71	165.9	11.20	14.8	43.00	2.90
Aug. 28 to Sept. 1, 1991	5	3	3.61	70.6	6.64	10.6	18.60	1.75
Sept. 2 to Sept. 3, 1991	2	2	5.54	67.4	3.16	21.3	68.10	3.19
Sept. 4, 1991	1	1	4.43	52.4	2.45	21.4	84.70	3.96
Sept. 5 to Sept. 7, 1991	3	3	4.64	31.7	2.52	12.6	17.90	1.42
Sept. 8, 1991	1	1	1.43	47.7	5.73	8.3	24.90	2.99
Sept. 9 to Sept. 23, 1991	15	1	2.67	67.3	3.59	18.8	4.37	0.23
Sept. 24 to Oct. 29, 1991	36	3	5.72	5.48	2.48	2.2	0.32	0.14
Oct. 30 to Oct. 31, 1991	2	2	2.14	60.4	2.97	20.3	23.60	1.16
Nov. 1 to Nov. 7, 1991	7	1	3.89	61.1	5.67	10.8	12.40	1.15
Nov. 8 to Nov. 18, 1991	11	1	6.27	75.9	4.74	16.0	15.80	0.99
Nov. 19, 1991	1	1	2.50	56.9	6.15	9.3	51.9	5.61
Nov. 20 to Dec. 1, 1991	12	4	0.74	174.6	17.80	9.8	3.93	0.40
Dec. 2 to Dec. 21, 1991	20	8	4.70	112.9	6.00	18.8	9.68	0.52
Dec. 22, 1991 to Jan. 5, 1992	15	5	1.68	170.3	14.00	12.2	6.96	0.57
Jan. 6 to Jan. 8, 1992	3	1	8.36	51.0	3.89	13.1	51.90	3.96
Jan. 9 to Jan. 18, 1992	10	6	8.51	89.8	7.95	11.3	27.90	2.47
Jan. 19 to Feb. 6, 1992	19	6	14.70	47.1	1.83	25.7	13.30	0.52
Feb. 7 to Feb. 11, 1992	5	1	3.40	113.6	10.00	11.3	28.20	2.49
Feb. 12 to Feb. 17, 1992	6	3	1.75	135.3	10.40	13.0	14.40	1.11
Feb. 18 to Feb. 24, 1992	7	3	5.13	92.0	3.46	26.6	24.60	0.93

For most of the samples, the precipitation record was obtained from a meteorological station located at ~ 3 km southeast of sampling site. In individual rain events, the actual volume in the rain collector was used to obtain amount of rainfall. NM means not measured, as the sample was lost in handling and/or processing.

TABLE 2: Sampling Time Intervals, Annual Depositional Fluxes and Concentrations of ^{7}Be and ^{210}Pb Measured in College Station, Texas, From June 1989 Through February 1992

Collection Interval Date	Days in Collection Interval	Rainfall cm	^{7}Be dpm L^{-1}	^{210}Pb dpm L^{-1}	$^{7}Be/^{210}Pb$ Activity Ratio	^{7}Be Flux dpm cm^{-2}	^{210}Pb Flux dpm cm^{-2}
June 16 to Aug. 7, 1989	53	17.10	126.5	10.40	12.2	14.90	1.22
Aug. 8 to Nov. 8, 1989	93	7.14	265.1	17.80	14.9	7.43	0.50
Nov. 9, 1989 to Feb. 6, 1990	90	17.00	181.3	12.00	15.1	12.50	0.83
Feb. 7 to March 4, 1990	26	8.43	45.3	2.71	16.7	5.36	0.32
March 5 to April 16, 1990	43	10.70	186.1	12.70	14.7	16.90	1.15
April 17 to May 29, 1990	43	18.40	121.6	7.56	16.1	19.00	1.18
May 30 to July 4, 1990	36	4.55	108.2	12.90	8.4	4.99	0.60
July 5 to Aug. 22, 1990	49	13.90	188.3	8.21	22.9	19.50	0.85
Aug. 23 to Sept. 17, 1990	26	10.70	77.2	3.73	20.7	11.60	0.56
Sept. 18 to Oct. 4, 1990	17	4.34	46.9	4.99	9.4	4.37	0.47
Oct. 5 to Nov. 8, 1990	35	12.00	96.7	5.52	17.5	12.10	0.69
Nov. 9, 1990 to Jan. 13, 1991	66	27.90	175.0	11.00	16.0	27.00	1.69
Jan. 14 to Feb. 10, 1991	28	20.50	101.0	11.50	7.7	23.50	3.06
Feb. 11 to March 21, 1991	39	8.20	298.4	25.70	11.6	22.90	1.97
March 22 to April 22, 1991	-	14.50	NM	NM	NM	NM	NM
April 23 to May 23, 1991	31	10.10	151.4	13.50	11.2	18.00	1.61
May 24 to July 4, 1991	42	14.40	91.1	7.21	12.6	11.40	0.90
July 5 to Sept. 6, 1991	64	19.30	40.7	4.97	8.2	4.48	0.55
Sept. 7 to Sept. 13, 1991	7	6.22	125.8	11.70	10.8	40.80	3.79
Sept. 14 to Nov. 15, 1991	63	9.02	109.1	8.52	12.8	5.70	0.45
Nov. 16, 1991 to Jan 8, 1992	54	24.60	34.3	2.68	7.5	3.32	0.45
Jan. 9 to Feb. 16, 1992	39	22.30	15.9	2.13	15.0	8.43	0.56

The precipitation record was obtained from a meteorological station located at the College Station Airport, ~ 3 km away from the sampling site. NM means not mesured, as the sample was lost in handling.

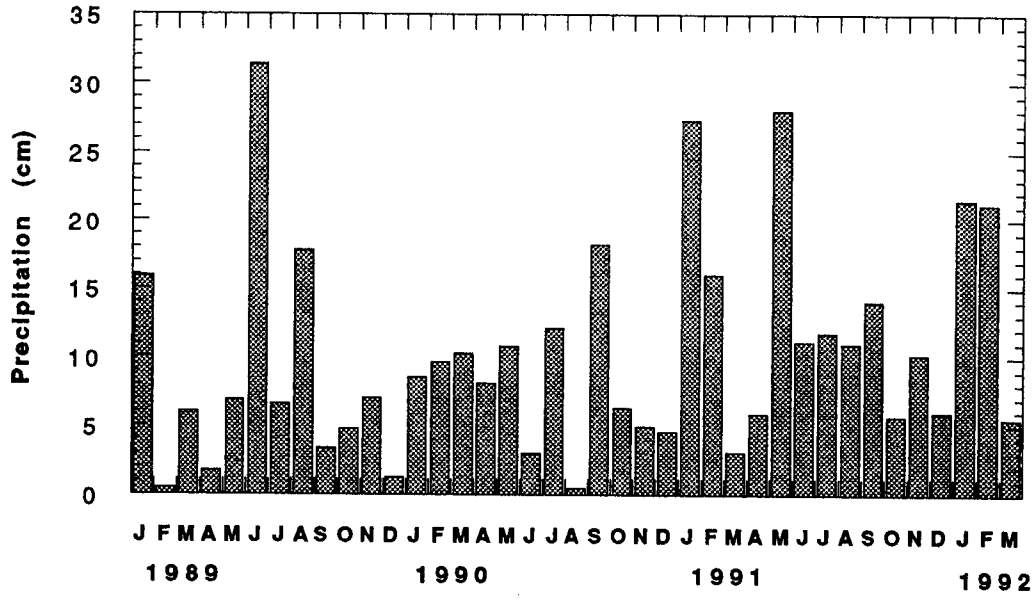

Fig. 1. Total monthly precipitation at Galveston, Texas.

whole deployment period was constant and by using the amount of precipitation during the deployment period. College Station rainwater collectors were sometimes deployed for periods longer than 1 month, and thus reliable estimation of monthly fluxes was not attempted under those circumstances. The frequency distributions of the depositional fluxes of ^{7}Be and ^{210}Pb for Galveston in individual bulk depositional samples are plotted in Figures 3 and 4, respectively. These figures indicate that in >80% of these individual samples, the variations of the depositional fluxes are within a factor of 10. The temporal trend of the monthly bulk depositional fluxes of ^{7}Be and ^{210}Pb at the Galveston station is shown in Figure 5. There are no clear consistent seasonal trends in the distribution of monthly fluxes during the study period. However, the amounts of precipitation seem to be controlling the magnitude of the depositional fluxes. For example, at Galveston, the 38 months for which the depositional fluxes are reported in Table 1, the highest monthly fluxes were during winter of 1991, coinciding with relatively high rainfall during those months. During January and February 1991, the ^{7}Be and ^{210}Pb fallout was equivalent to about 29.0% of the total annual fallout, which is similar to the percentage of the amount of precipitation of 43 cm, which was also 29% of the annual rainfall. During May and June, the ^{7}Be and ^{210}Pb fallout was equivalent to 35 and 32%, respectively, of the total annual fallout, slightly higher than the percentage of the amount of precipitation of 39 cm, which was ~26% of the annual rainfall.

The monthly ^{7}Be depositional fluxes at Galveston varied between 1.2 and 74.5 dpm cm^{-2} yr^{-1} with a mean of 13.9 dpm cm^{-2} yr^{-1} over the 38 months for which the depositional fluxes were measured. The corresponding monthly fluxes of ^{210}Pb varied between 0.072 and 4.75 dpm cm^{-2} yr^{-1} with a mean of 0.97 dpm cm^{-2} yr^{-1}. There was a spring maximum of ^{7}Be or ^{210}Pb fallout in

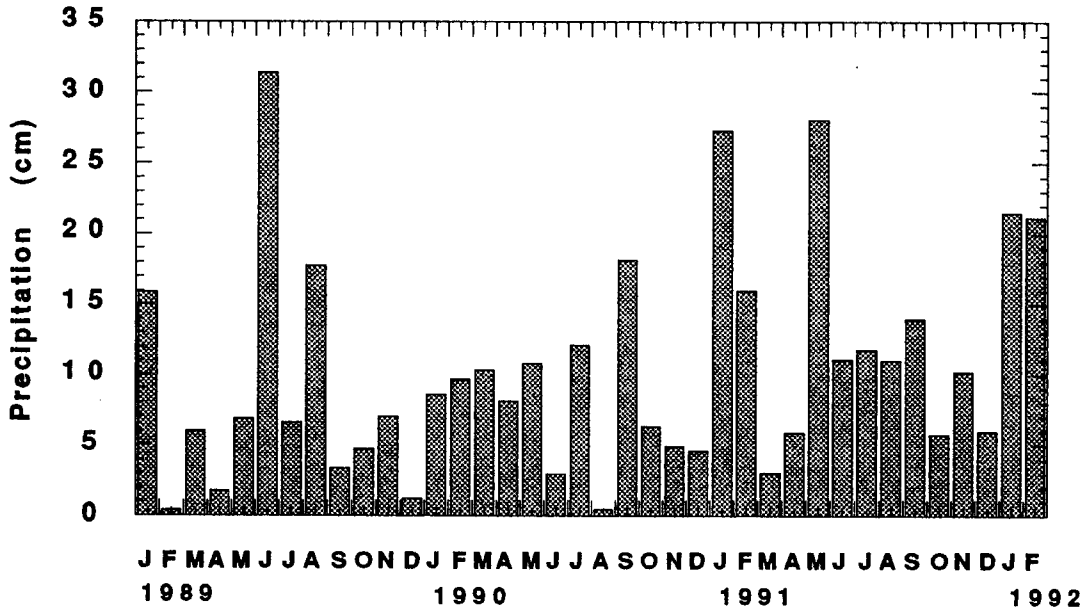

Fig. 2. Total monthly precipitation at College Station, Texas.

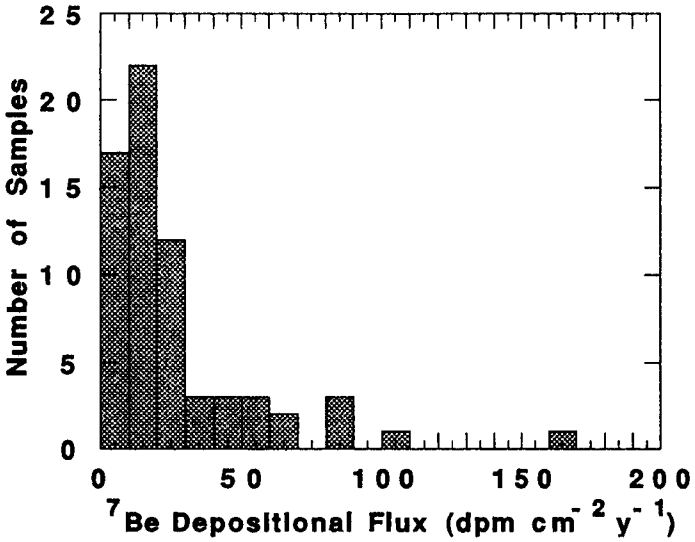

Fig. 3. Frequency distribution of the depositional fluxes of ^{7}Be in the 67 samples collected in Galveston, Texas.

only one of the three years (1991) for which we have a complete data set. The ^{7}Be flux maximum during spring is generally attributed to a significant injection of stratospheric ^{7}Be into the troposphere due to midlatitude (38°-51°N) folding of the tropopause which enhances stratospheric-tropospheric exchange [*Young et al.*, 1970; *Rangarajan and Gopalakrishnan*, 1970; *Dutkiewicz and Husain*, 1979, 1985; *Olsen et al.*, 1985; *Todd et al.*, 1989]. The ^{210}Pb flux maximum is likely related to the spring increase in the amount and frequency of precipitation, but in other places, it could be due to warming of frozen soils, or the drying out of saturated soils during spring or summer [*Olsen et al.*, 1985; *Schuler et al.*, 1991]. These differences in the depositional fluxes of ^{7}Be and ^{210}Pb during spring can also be due to the differences in the relative proportions of scavenging from stratiform and convective clouds. Frequent passage of strong fronts (associated with high rainfall) during all times of the year could also be a reason why we do not see a consistent ^{7}Be fallout maximum during spring.

During the spring months March, April and May, the fractions of ^{7}Be annual fluxes deposited in Galveston were 25, 29, and 37%, respectively, for the years 1989, 1990, and 1991. The corresponding values for ^{210}Pb are 19, 26, and 26% for the years 1989, 1990, and 1991, respectively. While we do not observe a markedly consistent seasonality in the ^{7}Be and ^{210}Pb fluxes at Galveston, possibly due to more complicated atmospheric conditions, such as differences in the relative proportion of the stratiform and convective cloud scavenging, seasonal variations on the depositional fluxes have been reported for other continental and coastal stations. About 40-45% of the total annual deposition is reported to occur in three spring months (March, April, and May) at several places, such as Oak Ridge, Tennessee [*Olsen et al.*, 1985], and Norfolk, Virginia [*Todd et al.*, 1989]. *Dibb* [1989] and *Canuel et al.* [1990] found higher fluxes during the months May and June over the entire 1-year study period, and *Schuler et al.* [1991] reported consistent flux maxima during the summer months. *Tsunogai et al.* [1985] reported that deposition rates of ^{210}Pb were higher in the winter at stations along the Japan Sea coasts, in contrast to the higher rates observed in the summer at stations along the Pacific coast.

Variations of the Specific Concentrations of ^{7}Be

Specific concentrations of ^{7}Be in all the samples collected are given in Tables 1 (Galveston) and 2 (College Station). More than 95% of both Galveston and College Station samples have activity concentrations of less than 250 dpm L^{-1}. The ^{7}Be specific concentration in bulk deposition samples from Galveston ranged between 5.5 dpm L^{-1} and 1243 dpm L^{-1}, with a geometric mean of 130 dpm L^{-1} (Table 1; since two of the 67 samples have an order of magnitude higher concentrations than the mean value, the geometric mean is reported). In College Station, ^{7}Be concentrations varied between 15.9 and 298 dpm L^{-1} (Table 2), with a geometric mean of 112 dpm L^{-1}. This difference between sites, somewhat greater nuclide deposition at Galveston, may be related to its coastal meteorology and availability of condensation nuclei such as marine sulfate aerosols. The rain collectors were deployed in College Station for relatively longer time intervals, and therefore, the spread in radionuclide concentrations in the collected rainwater was considerably smaller. This range is comparable to the range of 29 to 191 dpm L^{-1} for the southeastern Virginia coast [*Todd et al.*, 1989]. The higher end of the range in Galveston is due to very close interval sampling. Whenever there was a light shower (or drizzle) after a long dry spell, the samples contained very high specific concentrations of ^{7}Be. During drizzles, smaller-size cloud drops likely have higher radionuclide concentrations. Growth of smaller water droplets (the smaller the size, the higher the affinity to aerosols) through coalescence will tend to keep this concentration constant, while further water moisture condensation would dilute the radionuclide concentration. Furthermore, smaller amounts of rainfall are always associated with higher evaporation rates of droplets owing to the lower humidity below cloud cover. This is true both during their in-cloud cycling and during their fall to the surface of the Earth. In such cases, these droplets will be enriched in radionuclide concentration [*Wogman et al.*, 1968].

Relationship Between ^{7}Be Specific Concentration and Precipitation

The ^{7}Be specific concentration (in disintegrations per minute per liter) measured for Galveston is plotted against the amount of precipitation in Figure 6. The linear correlation between ^{7}Be concentration and rainfall is poor ($r = 0.27$, $P < 0.05$). However, if the parameters are plotted semi-logarithmically, an inverse dependency of the concentration on the amount of precipitation, x, is observed (^{7}Be (dpm L^{-1}) = 334 - 288.8 log(x); $r = 0.70$; $n = 67$, $P < 0.001$; Figure 6). For the rainwater samples from College Station, there is a significant correlation neither between ^{7}Be concentration and rainfall ($r = 0.23$, $n = 21$, Figure 7) nor between ^{7}Be flux and rainfall ($r = 0.072$, $n = 21$). However, other workers reported significant correlations between ^{7}Be specific concentration and the amount of precipitation (Connecticut, $r = 0.74$, $P < 0.001$; Bermuda, $r = 0.66$, $P < 0.05$ [*Turekian et al.*, 1983]; Norfolk, Virginia, r =

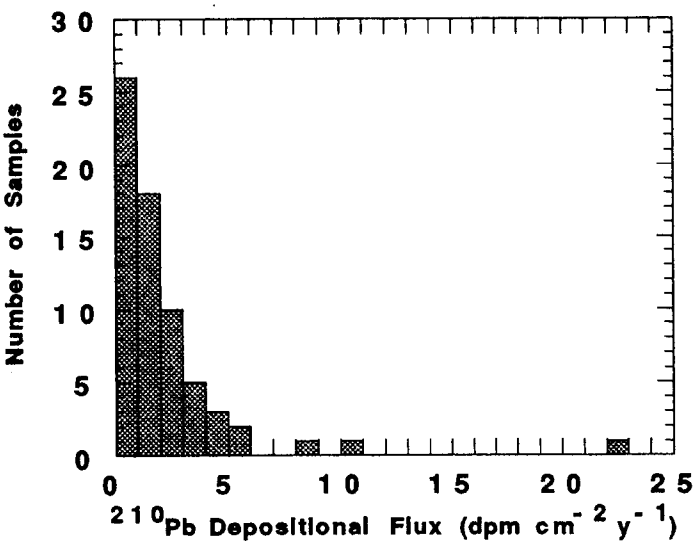

Fig. 4. Frequency distribution of the depositional fluxes of ^{210}Pb in the 67 samples collected in Galveston, Texas.

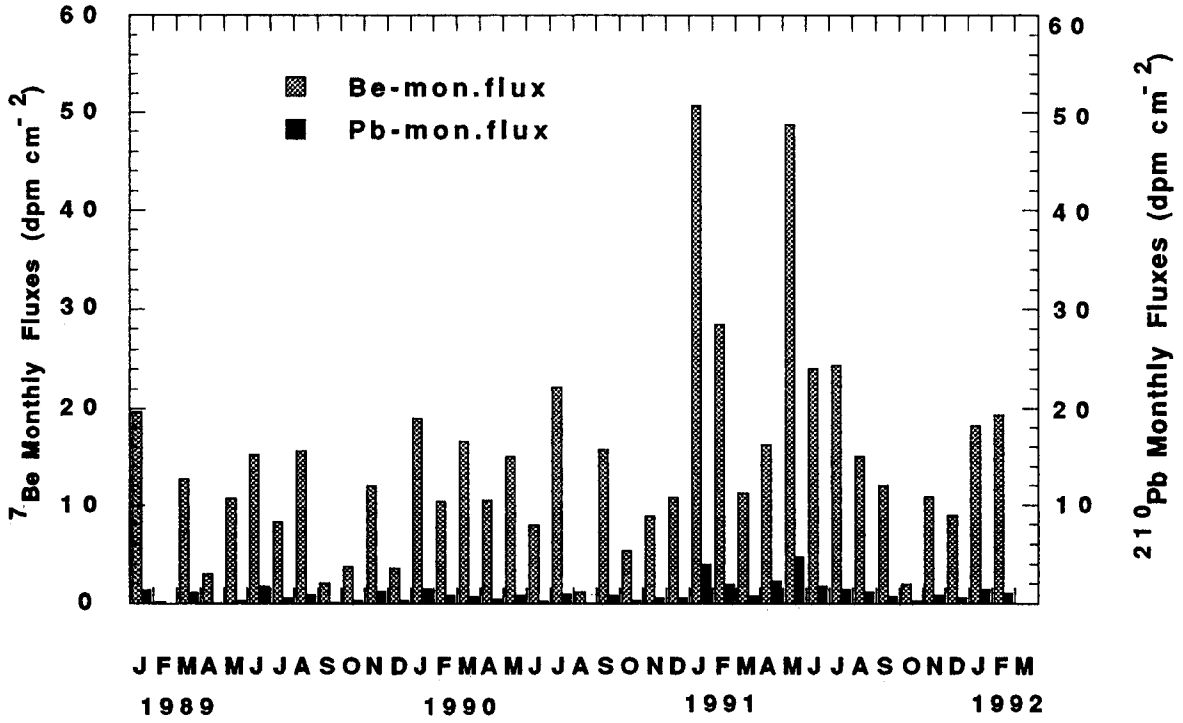

Fig. 5. Bar diagram illustrating monthly depositional fluxes of ^{7}Be and ^{210}Pb at Galveston, Texas during the years 1989, 1990, and 1991.

-0.73 $P < 0.001$; Oak Ridge, Tennessee, $r = -0.79$, $P < 0.001$ [*Olsen et al.*, 1985]). It has been shown that the concentration of ^{7}Be in rain decreased by a factor of 3 during a storm event which deposited 6 cm of rain [*Olsen et al.*, 1985]. Similarly, *Dibb* [1989] found that the specific concentration in samples taken serially during a single precipitation event decreased over time. *Canuel et al.* [1990] suggested that the concentration of ^{7}Be in rain may be higher during drier months and periods characterized by short-duration precipitation events. A very weak correlation or lack thereof in the Texas data set suggests that the ^{7}Be concentration is not mainly controlled by the amount of precipitation. A poor correlation has also been reported by *Brown et al.* [1989] and *Todd et al.* [1989], who similarly suggested that dilution is not the only process that controls the concentration of ^{7}Be in rainwater.

Volume-Weighted Concentration of ^{7}Be

Volume-weighted annual average ^{7}Be concentrations were calculated as total amount of ^{7}Be deposited in a year in the rain collector divided by total volume of water in the rain collector, taken either from the precipitation record (annual precipitation times surface area of the collector) or the actual volume of the water sample. At Galveston during 1989, 1990, and 1991, the average ^{7}Be concentration was 87, 124, and 155 dpm L^{-1}, respectively. In 1990, this average ^{7}Be concentration is identical at Galveston and College Station, whereas the 1991 year values are distinctly different (104 dpm L^{-1} at College Station versus 155 dpm L^{-1} at Galveston) even though the amounts of precipitation are comparable (Table 3). In Table 4, the volume-weighted ^{7}Be

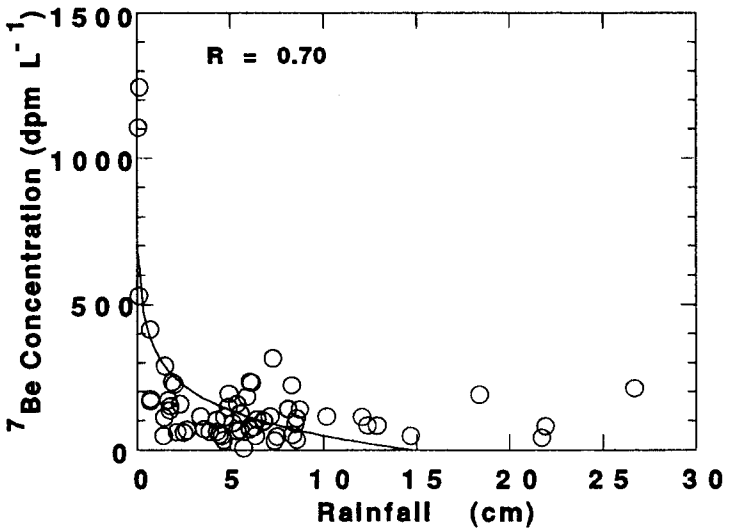

Fig. 6. Specific concentrations of ^{7}Be (disintegrations per minute per liter) versus amount of precipitation at Galveston, Texas.

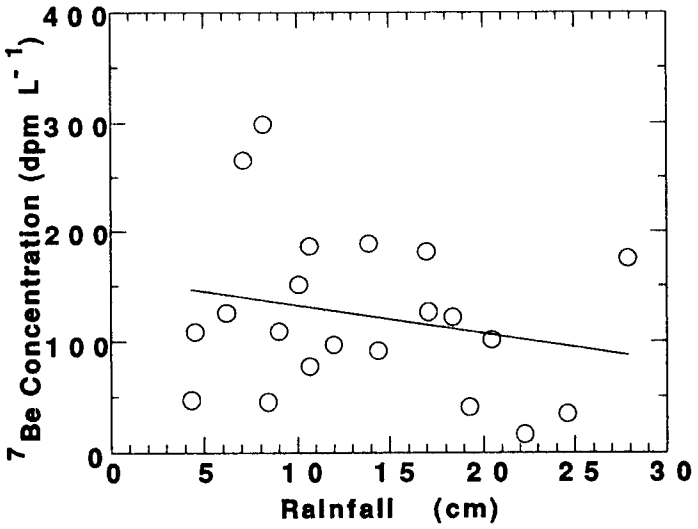

Fig. 7. Specific concentrations of ^{7}Be (disintegrations per minute per liter) versus amount of precipitation at College Station, Texas.

concentrations are compared with values reported in the literature. The value at Galveston in 1991 is much higher than the values reported in many other places; for example, for the years 1983 and 1984, at Norfolk, Virginia (36°53'N), *Todd et al.* [1989] reported 90 and 100 dpm L^{-1} respectively. Earlier results from Bombay (19°N, 33 dpm L^{-1} [*Lal et al.*, 1979]) and Quillayute, Washington (49°N, 30 dpm L^{-1} [*Crecelius*, 1981]) are much lower than the values we are reporting here, and this difference can be partly attributed to experimental artifacts in sample collection (the water in the rain collectors was not acidified, as reported by *Turekian et al.* [1983], and thus a significant portion of ^{7}Be could have been adsorbed onto the container walls).

When these volume-weighted concentrations are normalized to a constant precipitation of 100 cm (by correcting the volume-weighted concentration by the ratios of 100 cm yr^{-1} to the actual amount of rainfall), one obtains 84-127 dpm L^{-1} for Galveston and 71-126 dpm L^{-1} for College Station. This variation from year to year in the average ^{7}Be concentration can be attributed to differences in frequencies of atmospheric turbulence and to sequences and altitude of washout events.

Variations of the Specific Concentration of ^{210}Pb

The specific concentrations of ^{210}Pb in bulk deposition samples from Galveston varied between 2.03 and 218.1 dpm L^{-1} (geometric mean of 10.8 dpm L^{-1}; Table 1). In College Station, they ranged between 2.13 and 25.7 dpm L^{-1} (geometric mean of 8.6 dpm L^{-1}; Table 2). Most of the ^{210}Pb specific concentrations fall within a narrow range of 2-20 (only three samples have more than 100 dpm L^{-1}) for Galveston, whereas in College Station, they fall within an even narrower range of 2-18 dpm L^{-1} (with the exception of one sample). These variations in the specific ^{210}Pb concentrations can be attributed to factors such as scavenging efficiency, the altitude from which the precipitation occurred, and the sources of the air masses (as the continental and oceanic air masses will have very different atmospheric inventories of ^{210}Pb [*Turekian et al.*, 1977]). Since the deployment times of the rain collectors are longer for College Station samples, the specific concentration there is a result of integration of many rain events; thus the range of values becomes narrower than for the Galveston samples.

Relationship Between ^{210}Pb Concentrations and Amounts of Precipitation

The ^{210}Pb specific concentrations in individual samples are plotted against amount of precipitation in Figure 8 for Galveston and in Figure 9 for College Station. There is only a very weak correlation between the specific concentration and amount of precipitation (for Galveston, $r = 0.29$ and $P < 0.05$; for College Station, $r = 0.31$ and $n = 21$). *Benninger* [1976, 1978] found a correlation of 0.67 ($P < 0.001$) for the four years when precipitation samples were collected monthly. A relatively weak positive or negative correlation between ^{210}Pb concentration and amount of precipitation has been observed by other workers in Oak Ridge, Tennessee ($r = 0.65$, $P < 0.001$ [*Olsen et al.*, 1985]), Norfolk, Virginia ($r = -0.49$, $P < 0.05$ [*Todd et al.*, 1989]), and Texel, De Bilt, and Groningen, in the Netherlands ($r = 0.74$-0.94, $P < 0.001$ [*Zuo*, 1992]). *Nevissi* [1985] observed a positive correlation between monthly rainfall and the ^{210}Pb deposition at Seattle, Washington; however, for the same amount of rainfall the ^{210}Pb deposition rate also varied from year to year. *Hussain et al.* [1990] found no significant correlation between monthly atmospheric ^{210}Pb fallout and rainfall. This lack of correlation can be attributed to several factors that control the depositional fluxes of ^{210}Pb such as sources, pathways, altitude of the clouds, intensity of fronts,

TABLE 3. Annual Precipitation, Total Concentrations, and Depositional Fluxes of ^{7}Be and ^{210}Pb During 1989-1991 for Galveston and College Station

Parameter	1989		1990		1991	
	GVS	CLL	GVS	CLL	GVS	CLL*
Total Annual Precipitation (cm)	103	80	97	98	150	146
Total ^{7}Be (dpm)+	24,954	NM	33,557	35,483	65,006	38,355#
Total ^{210}Pb (dpm)+	1,946	NM	1,887	2,179	4,797	3,397#
$^{7}Be/^{210}Pb$ ratio	12.8	NM	17.8	16.3	13.6	11.3#
^{7}Be Flux (dpm cm^{-2} yr^{-1})	8.91	NM	12.0	12.7	23.2	15.0§
^{210}Pb Flux (dpm cm^{-2} yr^{-1})	0.70	NM	0.67	0.78	1.71	1.33§
^{7}Be (dpm L^{-1})¶	86.6	NM	123.6	123.9	154.8	104.2§
^{210}Pb (dpm L^{-1})¶	6.75	NM	6.95	7.94	11.4	9.23§

GVS is Galveston, and CLL is College Station. NM means not measured for the whole year; samples collected from June 1989 onward.
* One rain sample from College Station was lost during collection between March 22 and April 22 (14.5 cm rain).
+ Total concentrations collected in container with 2800 cm^2 area.
# These are the values from rainwater samples collected during 333 days in 1991 (except March 22 to April 22).
§ The fluxes (dpm cm^{-2} yr^{-1}) and specific concentrations (dpm L^{-1}) were calculated for 333 days and then prorated for 365 days.
¶ Concentration (dpm L^{-1}) = annual flux (dpm cm^{-2} yr^{-1})* 1000 / annual precipitation in cm = total amount (disintegration per minute)/total volume (L).

TABLE 4. Comparison of Volume-Weighted Concentrations of ^{7}Be and ^{210}Pb Measured in Galveston and College Station, Texas in 1989 through 1991 With Those Measured From Other Regions of the World after 1980

Location and Time of Collection	Rainfall cm	^{7}Be Weighted Concentration dpm L^{-1}	^{210}Pb Weighted Concentration dpm L^{-1}	$^{7}Be/^{210}Pb$ Activity Ratio	Source
New Haven, Connecticut (41°N)					
March 1977-Feb. 1978	148	158	7.98	19.7	*Turekian et al.* [1983]
Bermuda (33°N)					
Sept. 1977- Aug. 1978*	170	94	4.07	23.2	*Turekian et al.* [1983]
Oak Ridge, Tennessee (35°58'N, 84°17'W)					
Sept. 1982-Aug. 1983	110	93	8.78	10.6	*Olsen et al.* [1985]
Sept. 1983-Aug. 1984	143	99	8.25	12.0	
Switzerland					
(46.5°N)	120	138	7.58	18.1	*Dominik et al.* [1987]
(47°N)	110	146	7.55	19.3	*Schuler et al.* [1991]
Norfolk, Virginia (36°53'N; 76°18'W)					
1983	134	90	5.90	15.3	*Todd et al.* [1989]
1984	129	100	6.60	15.2	
Bordeaux, France (44.8°N)	103	71	3.33	21.3	*Thomas* [1988]
Paris (48.8°N)	68	45	9.60	4.6	*Thomas* [1988]
College Station, Texas (30°35'N; 96°22'W)					
1990	98	124	7.94	16.3	this paper
1991	146	104	9.23	11.3	
Galveston, Texas (29°18'N; 94°48'W)					
1989	103	87	6.75	12.8	this paper
1990	97	124	6.95	17.8	
1991	150	155	11.4	13.6	

Volume-weighted average concentrations were calculated as in Table 3.
* Based on 8 months of precipitation data

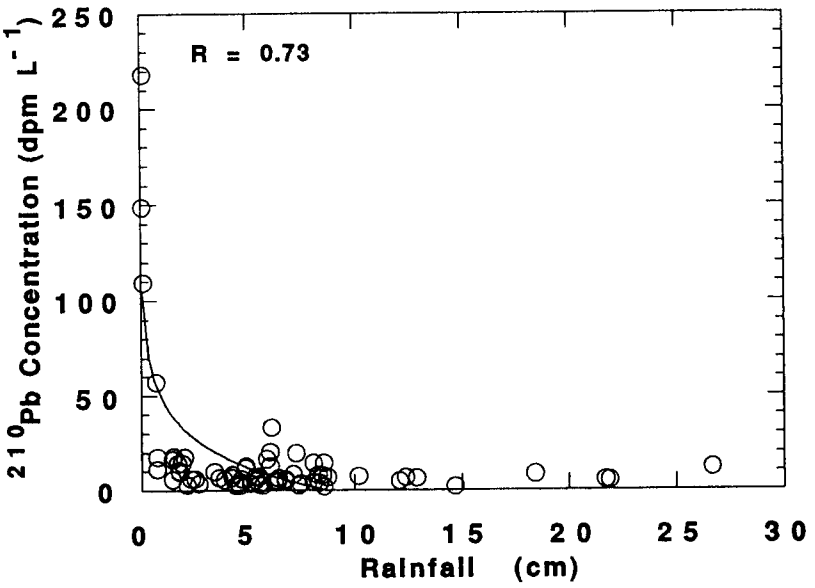

Fig. 8. Specific concentrations of ^{210}Pb (disintegrations per minute per liter) versus amount of precipitation at Galveston, Texas.

relative humidity, and the transit time of aerosols that contain ^{210}Pb from their source to the sink in rain collectors.

Volume-Weighted Concentration of ^{210}Pb

The volume-weighted ^{210}Pb concentrations for Galveston and College Station, calculated as the total amount of ^{210}Pb deposited divided by total rain volume, are given in Table 3. During the years 1989-1991, the concentrations were 6.75, 6.95, and 11.4 dpm L^{-1}, respectively, for Galveston and 7.94 dpm L^{-1} (1990) and 9.23 dpm L^{-1} (1991) for College Station. When the ^{210}Pb specific concentrations were normalized to a constant amount of precipitation, by taking the product of average concentration and the ratio of 100 cm yr^{-1} to the actual amount of rainfall, a relatively constant value resulted for both Galveston (7.10 dpm L^{-1}) and College Station (7.21 dpm L^{-1}). Thus it seems that the annual atmospheric depositional flux of ^{210}Pb is controlled by the amount of precipitation for the years we have studied, as may well be the case in other places.

In Table 4, our volume-weighted ^{210}Pb concentrations are compared with values reported in literature from other places. The value at Galveston in 1991 is much higher than the values reported in many other places; for example, for the years 1983 and 1984, at Norfolk, Virginia (36°53'N) *Todd et al.* [1989] reported only 5.9 and 6.6 dpm L^{-1} respectively. For New Haven, Connecticut (41°N) and Bermuda (33°N) *Turekian et al.*. [1983] reported 8.0 dpm L^{-1} and 4.1 dpm L^{-1}, respectively.

Variations of Specific Concentrations and Fluxes ^{7}Be of and ^{210}Pb Sampled During Individual Rain Events Within One Month

In 1991, some of the Galveston rainwater samples were collected right after individual rain events. Information that can be obtained from these samples will be useful in studying the variations of specific concentrations and fluxes between one rainout event and another, as well in determining the variability for a single month from year to year. For example, depositional fluxes in September 10-13, 1990 (all 4 days were rainy days) can be compared to the depositional flux during September 5-7, 1991 (all 3 days were rainy). The depositional flux of ^{7}Be differed by a factor of almost 2 between the two collection periods (27.2 and 17.9 dpm cm^{-2} yr^{-1} for the first and second periods, respectively) whereas the ^{210}Pb fluxes (1.59 and 1.42 dpm cm^{-2} yr^{-1}) remained essentially the same.

Another example is the four rain events that were collected continuously between September 2 and 8, 1991. If all these rain events were derived from the same water vapor, then one would expect a dilution of the concentration with time. However, the specific concentrations of ^{7}Be decreased from 67.4 dpm L^{-1} in the first rain to 31.7 dpm L^{-1} in the third rain and then increased again to 47.7 dpm L^{-1} in the fourth rain event. Similarly, the specific concentration of ^{210}Pb decreased from 3.16 dpm L^{-1} in the first rain to 2.52 dpm L^{-1} in the third rain and again increased to 5.73 dpm L^{-1} in the fourth rain event. The nuclide fluxes varied by a factor of 3 for ^{210}Pb and a factor of 5 for ^{7}Be. The $^{7}Be/^{210}Pb$ activity ratios varied by a factor of 2.6 within these 8 days. Thus it appears that a significant amount of ^{210}Pb-rich aerosols in the fourth rain is derived from entrained continental air. A record of the wind directions during these 8 days indicates that the fourth rain event was brought by southern winds, different from the others.

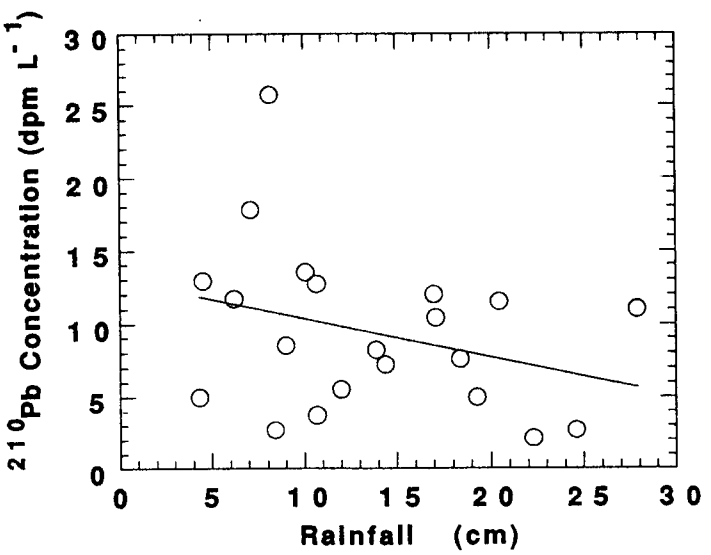

Fig. 9. Specific concentrations of ^{210}Pb (disintegrations per minute per liter) versus amount of precipitation at College Station, Texas.

Relationship Between Wind Direction and Depositional Fluxes of ^{7}Be and ^{210}Pb

The depositional fluxes of ^{7}Be and ^{210}Pb are plotted against wind direction in Figures 10 and 11, respectively. The common wind direction is from the southeast. On only one day (January 15, 1991) did the wind originate from the northwest, and this wind appears to have brought significant amounts of ^{210}Pb and ^{7}Be, resulting in the highest ^{7}Be and ^{210}Pb fluxes. These higher levels could be due to longer transit time of aerosols which brought large amounts of ^{7}Be and ^{210}Pb nuclides or stronger interactions between higher-altitude jet stream and near-surface air masses. However, there is no significant relationship between the depositional fluxes of these nuclides and wind direction. Also, there is no correlation between wind speed and depositional fluxes of ^{7}Be or ^{210}Pb at any of the stations. Thus it appears that strong fronts passing through Texas entrain variable amounts of oceanic and continent-derived aerosols.

Fraction of the Annual Depositional Fuxes of ^{7}Be and ^{210}Pb From Pulse Rain Events

Even though the rainy days are spread over the whole year, during a few days, the rainfall was heavy due to thunderstorms, hurricanes, or tornadoes. The contribution of ^{7}Be and ^{210}Pb to the annual bulk depositional fluxes during these heavy rainout events is not very well known. Since the input of these nuclides during heavy rains into the coastal water is useful for studying the particle dynamics in coastal and bay waters [*Olsen et al.*, 1989; *Baskaran and Santschi*, 1993], it is important to know the relative contribution during these events when using the nuclides as tracers for particle cycling.

In Galveston, there were 82, 88, and 119 rainy days during 1989, 1990, and 1991, respectively (Table 5). Of these, 4, 6, and 5 rainy days had greater than 5 cm rainfall during the years 1989, 1990, and 1991, respectively. Even though the fractional amount of precipitation and the depositional fluxes of radionuclides are comparable in these pulse events, a higher amount of depositional flux into the coastal waters can be used as a tracer to investigate the pollutant transport and particle cycling. For example, in Galveston Bay, with a mean depth of 2 m, one of these individual rainout events (January 6-8, 1992, Table 1) typically would have resulted in total concentrations (dissolved and particulate) of 2.2 dpm L^{-1} for ^{7}Be and 0.15 dpm L^{-1} for ^{210}Pb in the bay water. These concentration levels are easily measurable, and thus the fate of particles as well as the dissolved nuclides can be studied [*Baskaran and Santschi*, 1993].

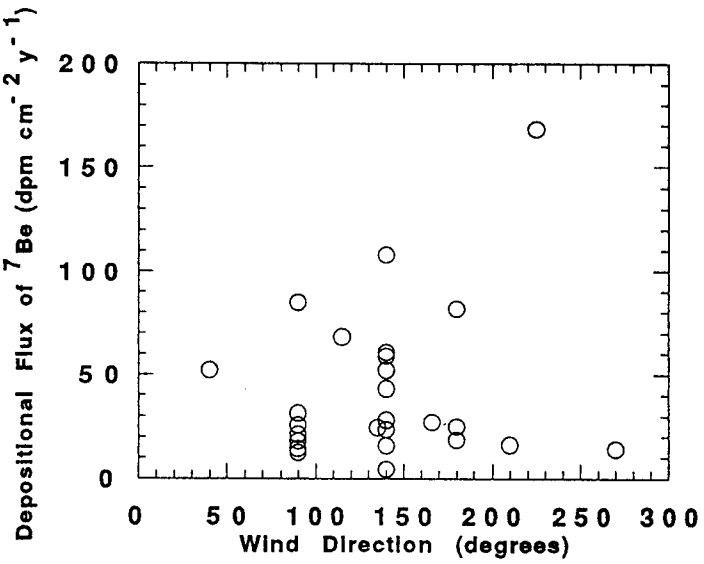

Fig. 10. The depositional flux of ^{7}Be at Galveston, Texas is plotted against wind direction.

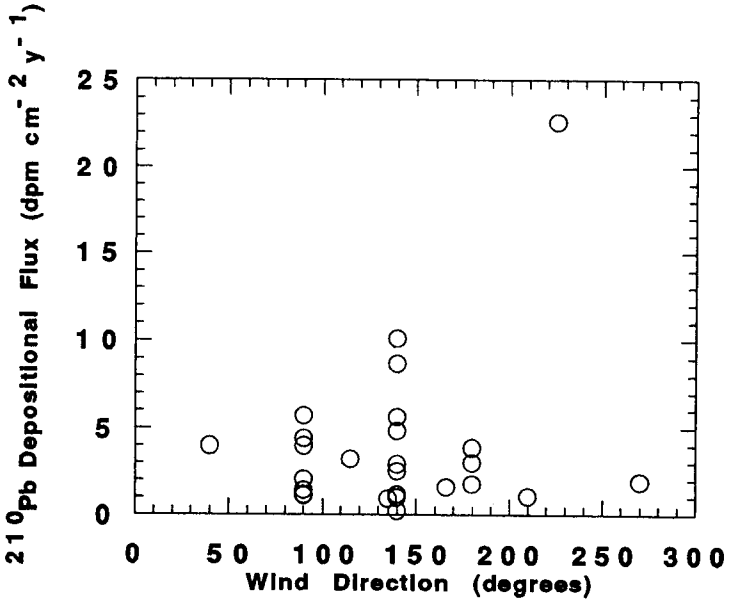

Fig. 11. The depositional flux of ^{210}Pb at Galveston, Texas is plotted against wind direction.

In 1990, six heavy rainfalls with more than 5 cm precipitation deposited 22% of the total annual ^{7}Be flux and 21% of the annual flux of ^{210}Pb (Table 5). During 1991, the ^{7}Be and ^{210}Pb deposited during these 5 days of heavy rainfall amounted to 26 and 20%, respectively, of the annual flux (Table 5). The fraction of the depositional flux of ^{7}Be and ^{210}Pb for 1989 was not calculated as one of the four rain samples with more than 5 cm rain was lost. It appears that 4-6 heavy rain events per year (typically 4-6% of the total number of rainy days) account for about 20-30% of the total annual deposition rate of ^{7}Be and ^{210}Pb. This is likely to be true for other atmospherically delivered stable elements as well.

Dry Depositional Fluxes of ^{7}Be and ^{210}Pb

The time during which the dry depositional flux study was carried out was limited to about a week each in October and November 1990 and to 2 weeks in October 1991. The dry depositional fluxes of radionuclides are presented in Table 6. These dry depositional fluxes were obtained by exposing the rain collector's lid (surface area of 2800 cm^2, depth of 3.5 cm) for 7-14 days with 1.5 cm distilled water. Such a method was advisable, since we were primarily interested in estimating the depositional fluxes of ^{7}Be and ^{210}Pb into coastal waters [*Dasch*, 1985]. Another method of determining the dry fallout is from the measurement of bulk (wet and dry) fallout of these nuclides and extrapolation to zero precipitation. Soil profiles have been mentioned too, as they provide bulk atmospheric depositional fluxes over the mean life of the radionuclide [*Graustein and Turekian*, 1986]. However, it will be difficult to get information on the annual dry fallout for ^{210}Pb and ^{7}Be from such studies.

The dry depositional flux for ^{7}Be varied between 0.249 and 0.394 dpm cm^{-2} yr^{-1}. This dry depositional flux was 3.0 to 7.4% of the bulk depositional flux during October 1990, but it amounted to about 87% during October 1991. This high value is likely due to the fact that the bulk deposition rate during that period was low, as there was no rainfall for a period of about a month (September 25 to October 27), thus giving rise to high concentrations of ^{7}Be and

TABLE 5. Percentage of Bulk ^{7}Be and ^{210}Pb Depositional Fluxes During Heavy Rain at Galveston, Texas, During 1990 and 1991

Collection Interval	Total amount of precipitation, cm	Number of Days With > 5 cm Rain	Percentage of Annual ^{7}Be Flux	Percentage of Annual ^{210}Pb Flux
1989	103	4	IC	IC
1990	97	6	22.2 (2.66)	20.5(0.137)
1991	150	5	25.8(5.99)	20.0(0.342)

Numbers in parentheses denote depositional fluxes (disintegrations cm^{-2} yr^{-1}) during the heavy rain events. IC means ^{7}Be and ^{210}Pb annual flux for the year 1989 is incomplete and hence was not calculated.

^{210}Pb in surface air aerosols. *Todd et al.* [1989] observed a weak positive correlation between dry ^{7}Be depositional flux and the amount of monthly precipitation and attributed this to higher relative humidity during dry fallout periods. However, our present data do not allow us to address this question to any great extent. It is sufficient to say that humidity is high in Galveston year around. Furthermore, *Santschi et al.* [1988] reported higher dry fallout values during humid, foggy days for Chernobyl-derived radionuclides. Also, traditionally, the linear relationship between amount of precipitation and bulk depositional flux can enable one to estimate the dry fallout at zero precipitation. Since we do not find any relationship between amount of precipitation and depositional fluxes of ^{7}Be and ^{210}Pb it is not possible to estimate dry depositional fluxes from such an approach.

The dry depositional flux for ^{210}Pb varied between 0.032 and 0.092 dpm cm^{-2} yr^{-1} during 7- to 14-day periods, which appears to be a higher fraction of the total fallout than was measured for ^{7}Be for the same period (6 and 22% in October/November 1990 and 41% in October 1991; Table 6). This higher fraction of dry fallout for ^{210}Pb relative to ^{7}Be could likely be caused by ^{210}Pb leaching from fine resuspended dust from ground in the acidified dry fallout collector. Alternatively, dry deposition could be more important for ^{210}Pb than for ^{7}Be. A similar observation has been reported by *Olsen et al.* [1985], based on the relatively weaker correlation between total monthly ^{210}Pb deposition and the amount of precipitation. The average fraction of dry deposition flux of ^{210}Pb, 14% in 1990, appears to be closer to the value reported by other workers. For example, *Brown et al.* [1989] measured dry ^{7}Be depositional fluxes and estimated that less than 10% of the bulk fallout is dry fallout. The ^{210}Pb dry depositional flux in 1991 is higher than in 1990, as was also the case for ^{7}Be.

Annual Bulk Depositional Fluxes of ^{7}Be and ^{210}Pb

The annual bulk depositional fluxes of ^{7}Be and ^{210}Pb at Galveston and College Station are given in Table 3. During the years 1989 to 1991, depositional fluxes of ^{7}Be in Galveston varied between 8.9 and 23.2 dpm cm^{-2} yr^{-1}, with an average of 14.7 dpm cm^{-2} yr^{-1}. The highest radionuclide fallout in 1991 corresponds to a maximum annual rainfall. Even though the annual depositional flux of ^{7}Be varied by a factor of 2.6, the ^{7}Be flux normalized to 100 cm yr^{-1} rainfall varies only by a factor of 1.8. The College Station annual depositional values of ^{7}Be in 1990 and 1991 were 12.7 and 15.0 dpm cm^{-2} yr^{-1}. This range of values can be compared to the values reported from other places, including New Haven, Connecticut (22.7 dpm cm^{-2} yr^{-1} [*Turekian et al.*, 1983]), Bermuda (17.1 dpm cm^{-2} yr^{-1} [*Turekian et al.*, 1983]), Oak Ridge, Tennessee (12.0 dpm cm^{-2} yr^{-1} [*Olsen et al.*, 1985]), Norfolk, Virginia (12.0-12.9 dpm cm^{-2} yr^{-1} [*Todd et al.*, 1989]), Chesapeake Bay, Maryland (13.6 dpm dpm cm^{-2} yr^{-1} [*Dibb*, 1989]), and Dubendorf, Switzerland (16.0-16.5 dpm cm^{-2} yr^{-1} [*Schuler et al.*, 1991]).

The annual ^{210}Pb depositional fluxes in Galveston range between 0.67 and 1.71 dpm cm^{-2} yr^{-1}, with a mean of 1.03 dpm cm^{-2} yr^{-1}. Depositional fluxes in College Station during 1990 were 0.78 dpm cm^{-2} yr^{-1} and in 1991 1.33 dpm cm^{-2} yr^{-1}. The ^{210}Pb fluxes normalized to 100 cm yr^{-1} rainfall for College Station are, however, about the same in both years, 0.80 dpm cm^{-2} yr^{-1} in 1990 and 0.77 dpm cm^{-2} yr^{-1} in 1991. The corresponding values for Galveston range between 0.68 and 1.14 dpm cm^{-2} yr^{-1}, with a mean of 0.84 dpm cm^{-2} yr^{-1} for 100 cm yr^{-1} rainfall. These values can be compared to the values reported from other regions. *Benninger* [1976, 1978] reported for the 4-year period between 1973 and 1977, annual ^{210}Pb fluxes in New Haven, Connecticut, of 0.86 to 1.04

TABLE 6. Dry Depositional Fluxes of ^{7}Be and ^{210}Pb Measured in Galveston, Texas

Collection Interval	Time for Which Sample Was Collected days	^{7}Be Flux dpm cm^{-2} yr^{-1}	^{210}Pb Flux dpm cm^{-2} yr^{-1}	$^{7}Be/^{210}Pb$ Activity Ratio
Oct. 26 to Nov. 2, 1990	7	0.249 (8.40)	0.031 (0.486)	8.11 (17.3)
Nov. 12 to Nov. 19, 1990	7	0.394 (5.35)	0.092 (0.420)	4.28 (12.7)
Oct. 7 to Oct. 21, 1991	14	0.277 (0.318)	0.059 (0.144)	4.72 (2.21)

Numbers in parentheses denote bulk depositional fluxes (or activity ratio) during this sampling period.

dpm cm^{-2} yr^{-1}. *Nevissi* [1985] measured the monthly depositional fluxes of ^{210}Pb in Seattle, Washington, over a period of 7 years, and the annual depositional flux varied between 0.18 and 0.81 dpm cm^{-2} yr^{-1}. Average fluxes were 0.44 dpm cm^{-2} yr^{-1} for Seattle, Washington [*Nevissi*, 1985], 1.2 dpm cm^{-2} yr^{-1} for New Haven, Connecticut [*Turekian et al.*, 1983], 0.69 dpm cm^{-2} yr^{-1} for Bermuda [*Turekian et al.*, 1983], 1.04 dpm cm^{-2} yr^{-1} for Oak Ridge, Tennessee [*Olsen et al.*, 1985], 0.79-0.85 dpm cm^{-2} yr^{-1} for Norfolk, Virginia [*Todd et al.*, 1989], 0.83-0.91 dpm cm^{-2} yr^{-1} for Dubendorf and Geneva, Switzerland [*Dominik et al.*, 1987; *Schuler et al.*, 1991], and 0.32-0.43 dpm cm^{-2} yr^{-1} for Texel, De Bilt, and Groningen, the Netherlands [*Zuo*, 1992]. *Moore and Poet* [1976] estimated the local ^{210}Pb flux from a correlation of ^{210}Pb concentration profiles versus latitude. For the western central United States, they reported an average value of 0.8 dpm cm^{-2} yr^{-1}.

Our measured ^{210}Pb fallout of 0.67-1.71 dpm cm^{-2} yr^{-1} corresponds to a ^{222}Rn emanation rate from continental soils of 0.36-0.91 atoms cm^{-2} s^{-1}, with a mean of 0.55 atoms cm^{-2} s^{-1} for Galveston and College Station. This is in reasonable agreement with 0.40 atoms cm^{-2} s^{-1} for the central and coastal Texas and 0.7 atoms cm^{-2} s^{-1} for Texas plains [*Wilkening et al.*, 1975]. However, the ^{210}Pb containing aerosols may have been derived from continental sites, which were farther away and had different radon exhalation rates.

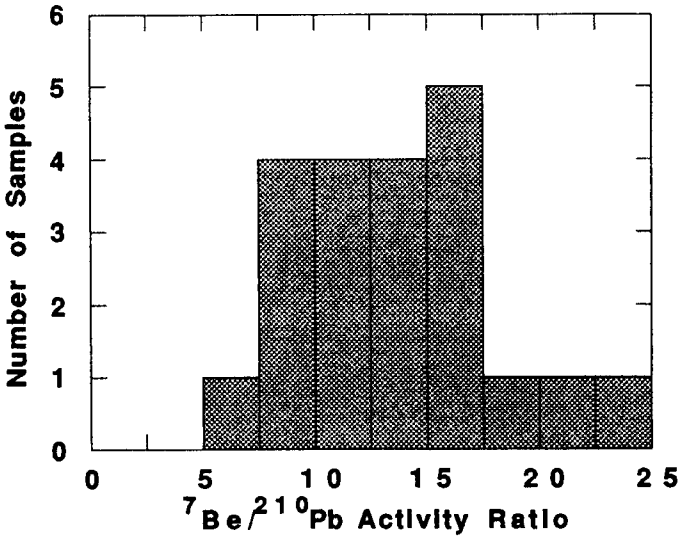

Fig. 13. Frequency distribution of $^{7}Be/^{210}Pb$ activity ratio in College Station, Texas samples.

Can ^{7}Be and ^{210}Pb Depositional Fluxes be Used as Two Independent Atmospheric Tracers?

Since their modes of production are different, ^{7}Be and ^{210}Pb have two distinct sources; however, they are removed from the atmosphere by very similar mechanisms. If the depositional fluxes of these two nuclides were found to be linearly related to each other, then they could not be used as two independent atmospheric tracers for continental and oceanic air.

The $^{7}Be/^{210}Pb$ ratios in all the rainwater samples, from both Galveston and College Station, are given in Tables 1 and 2. The $^{7}Be/^{210}Pb$ activity ratio histograms are plotted in Figures 12 (Galveston) and 13 (College Station). The ratios varied over an order of magnitude, between 2.21 and 32.6. The annual $^{7}Be/^{210}Pb$ ratios varied between 11.3 and 17.8, with a mean of 14.4 (Table 3). Over the 7 consecutive days during which rainfall was collected (four rains, September 2-8, 1991), this ratio varied between 8.33 and 21.4. Thus, when it was raining every day for several days, the ratio gradually decreased and this decrease can be attributed to the variable mixing of continental aerosols that are enriched in ^{210}Pb compared to oceanic aerosols. For a given latitude, however, aerosols are expected to have more or less the same ^{7}Be concentration.

The bulk depositional fluxes of ^{7}Be are plotted against ^{210}Pb depositional fluxes in Figure 14 for Galveston and in Figure 15 for College Station. The strong correlation between the two nuclides, for both Galveston ($r = 0.94$, significant at $P < 0.001$) and College Station ($r = 0.91$, significant at $P < 0.001$), indicates dependence rather than independence of these two tracers.

There is another reason for exploring the possibility of using ^{7}Be and ^{210}Pb nuclides as two independent tracers. In the years 1989 and 1990, with about the same rainfall, the depositional fluxes of ^{210}Pb remained the same at Galveston (0.70 and 0.67 dpm cm^{-2} yr^{-1}, respectively), while the annual ^{7}Be flux in 1990 was 35% higher than that of 1989. However, the good correlation between the two (as seen in Figures 14 and 15) suggests that they were generally removed by the same atmospheric process(es) with only minor variations in the annual flux ratios. Theoretically, one would expect a good correlation between the two mainly for continental stations,

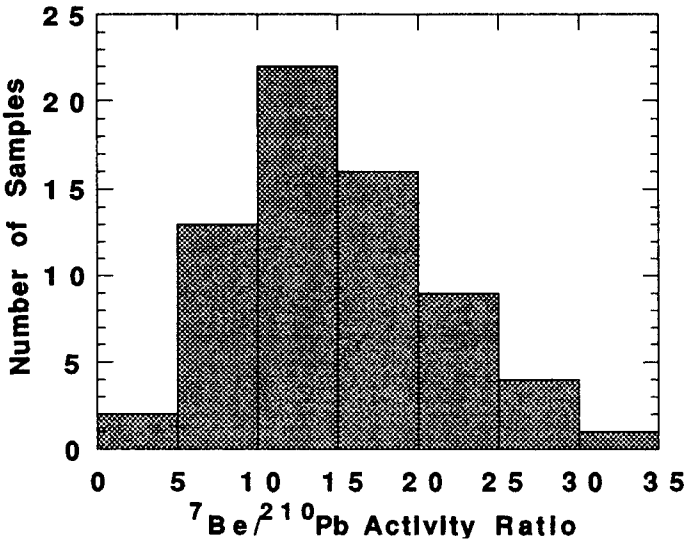

Fig. 12. Frequency distribution of $^{7}Be/^{210}Pb$ activity ratio in Galveston, Texas samples.

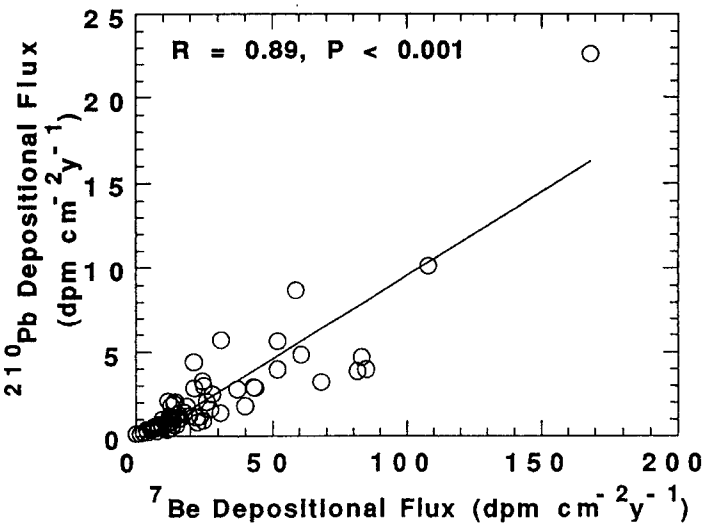

Fig. 14. Bulk depositional fluxes of ^{7}Be are plotted against ^{210}Pb fluxes for Galveston, Texas.

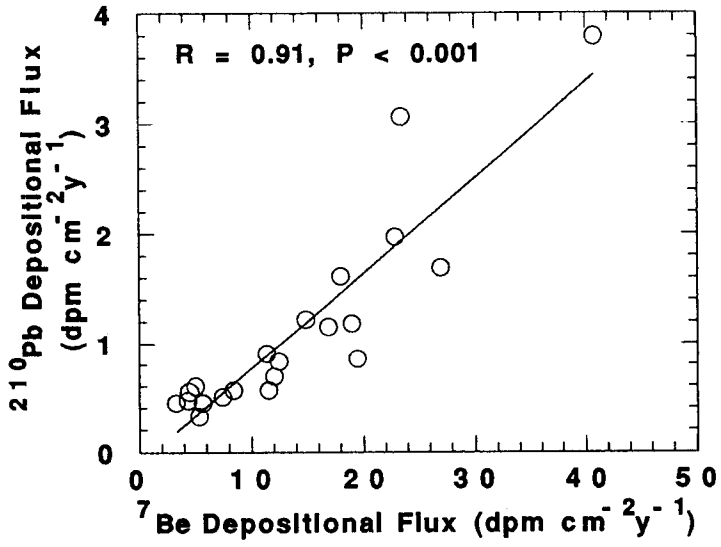

Fig. 15. Bulk depositional fluxes of ^{7}Be are plotted against ^{210}Pb fluxes for College Station, Texas.

such as Dubendorf, Switzerland, since most of the "^{210}Pb-tagged" aerosols are land derived and are primarily removed by precipitation. In a purely oceanic station, like Bermuda, parts of the air mass are likely derived from continents and so are enriched in ^{210}Pb, whereas other parts are ocean derived, and so are depleted in ^{210}Pb. Thus we do not expect any correlation between the two nuclides in an oceanic station even if they are removed by the same kinds of atmospheric processes. In coastal stations, we might expect a mixture of the two end-member cases and thus a relatively weak correlation.

The depositional flux of ^{210}Pb is plotted against depositional flux of ^{7}Be in Figure 16 for continental stations Dubendorf, Switzerland ($r = 0.90$, $P < 0.001$) and Oak Ridge, Tennessee, ($r = 0.86$, $P < 0.001$), coastal stations Galveston, Texas ($r = 0.94$, $P < 0.001$], New Haven, Connecticut ($r = 0.59$, $P < 0.05$), and Norfolk, Virginia ($r = 0.77$, $P < 0.001$), and oceanic station Bermuda ($r = 0.43$, $n = 8$). The least squares best fit line for the oceanic station is distinctly different from all stations, even from most coastal stations. The regression line for New Haven, Connecticut, appears to be that expected for a coastal station. Galveston, Texas, and Norfolk, Virginia ($r = 0.65$, $P < 0.001$) appear to behave more like continental stations in terms of the relationship between ^{7}Be and ^{210}Pb depositional fluxes (data taken from *Olsen et al.* [1985], *Todd et al.* [1989] and *Schuler et al.* [1991]). It appears that in most of the continental and coastal stations, ^{7}Be and ^{210}Pb cannot be used as two independent atmospheric tracers. Only in oceanic and a few coastal stations do ^{7}Be and ^{210}Pb fluxes seem to vary independently, and in such places these nuclides can be used as independent air mass tracers.

Implications of the Variations of Annual ^{210}Pb Depositional Fluxes for the ^{210}Pb-Based Sediment Chronologies in Lakes and Coastal Waters

The ^{210}Pb sediment chronology in aquatic systems is based on the assumption that the annual flux of ^{210}Pb to the sediments remains constant [*Robbins*, 1978; *Robbins and Edgington*, 1975]. When the residence time of ^{210}Pb in the water column is much shorter than the half-life of ^{210}Pb, that is, 22 years, then any variability in the annual atmospheric delivery of ^{210}Pb will cause a similar variability in the annual delivery to the sediments. For example, in coastal and estuarine waters, the residence time of dissolved ^{210}Pb is of the order of a month or less [*Baskaran and Santschi*, 1993, and references therein]. Under such circumstances, all of the atmospheric ^{210}Pb will be delivered to the sediments, and therefore the annual variability of the depositional flux of ^{210}Pb will result in variable flux to the sediment-water interface. Annual depositional fluxes of ^{210}Pb in Galveston varied between 0.67 and 1.71 dpm cm^{-2} yr^{-1} over the 3 years this study was carried out. Similar depositional flux variations have been previously reported by *Nevissi* [1985] for Seattle, Washington (a 4.4-fold range in the ^{210}Pb annual flux during the periods 1973 and 1980). Thus it is likely that a vertical profile of ^{210}Pb concentrations in coastal (as well as lake) sediments will be affected not only by sediment mixing (physical and biological) but also by the variability of the

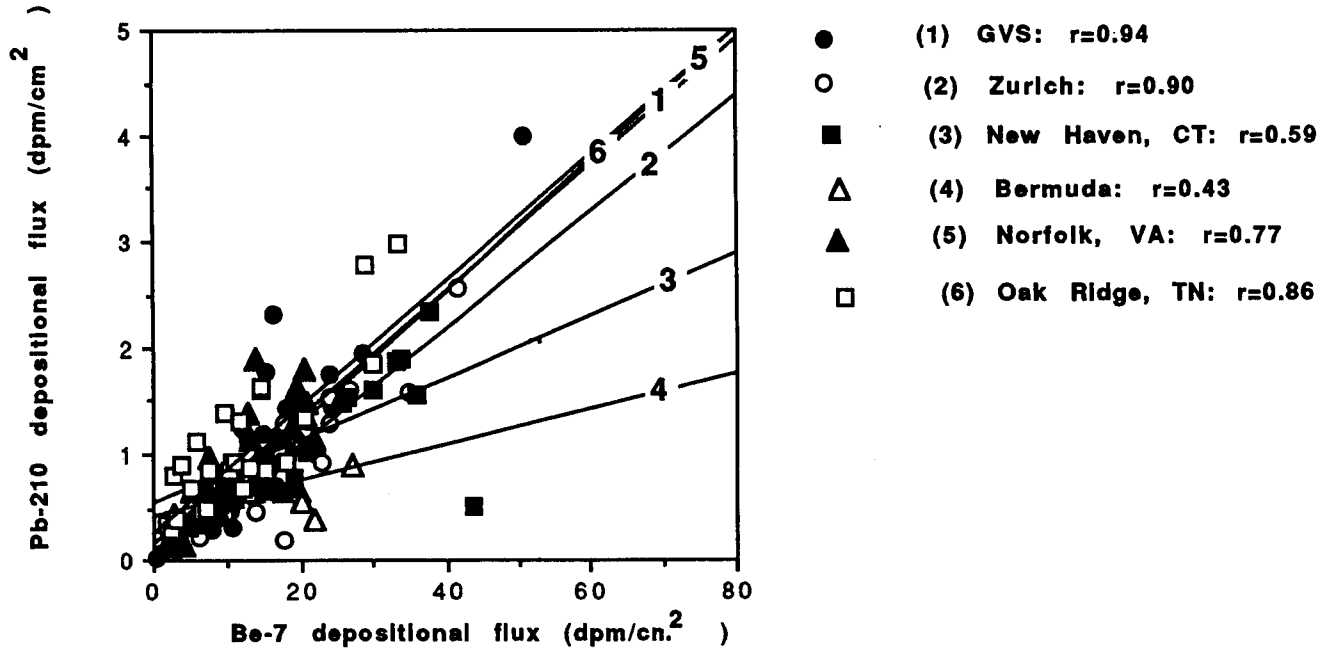

Fig. 16. Bulk depositional fluxes of ^{7}Be are plotted against ^{210}Pb fluxes for data collected from continental, coastal, and oceanic stations.

atmospheric depositional flux of ^{210}Pb. In open ocean waters, on the other hand, such as in the North Atlantic and North Pacific oceans, the dissolved ^{210}Pb residence time is of the order of 10-100 years [*Craig et al.*, 1973; *Somayajulu and Craig*, 1976]; thus the annual variability of the atmospheric ^{210}Pb flux will be smoothed out, and the variations of the annual depositional flux of ^{210}Pb to the sediments will be much more smaller when sediment delivery rates are constant.

CONCLUSIONS

Simultaneous measurements of bulk atmospheric depositional fluxes of ^{7}Be and ^{210}Pb were made at a coastal station (Galveston, Texas) and an inland site (College Station, Texas), for about 3 years. From this extensive database, we draw the following conclusions.

1. The annual depositional flux of ^{7}Be, measured at Galveston during 1989 to 1991, does not remain constant but varies between 8.9 and 23.2 dpm cm^{-2} yr^{-1}, with a mean of 14.7 dpm cm^{-2} yr^{-1}. The flux varies with the amount of precipitation. The precipitation-normalized ^{7}Be flux is not constant either but varies within a factor of 1.8. The ^{210}Pb depositional fluxes range between 0.67 and 1.71 dpm cm^{-2} yr^{-1}, with a mean of 1.03 dpm cm^{-2} yr^{-1}. The highest values are in the year with the maximum rainfall. The precipitation-normalized ^{210}Pb fluxes are constant for College Station, while they vary by a factor of 1.7 for Galveston.
2. The bulk depositional fluxes of ^{7}Be and ^{210}Pb do not show any consistent seasonal trends. In one year there was a spring maximum, and in another year there were flux maxima in both winter and summer. However, the annual depositional fluxes are most likely controlled by the amount of precipitation during that year.
3. It appears that 4-6 heavy rain events in a year (typically 4-6% of the total number of rainy days) account for about 20-30% of the total annual deposition rate of ^{7}Be and ^{210}Pb. This is likely to be true for other atmospherically delivered stable elements also.
4. The dry depositional fluxes were generally less than 10% for ^{7}Be. However, in the months when there was scanty rain, dry depositional fluxes of ^{7}Be and ^{210}Pb became a major portion of the bulk depositional fluxes. The fraction of dry depositional flux to total flux appears to be higher for ^{210}Pb than ^{7}Be. This higher ^{210}Pb dry depositional flux relative to ^{7}Be could also be due to leaching of resuspended dust by acid solution in the dry fallout collector.
5. On the basis of comparison with other measurements reported in the literature, it appears that in most of the continental and coastal stations, ^{7}Be and ^{210}Pb fallout pattern is tightly correlated, as is observed in Galveston and College Station, and thus the two nuclides cannot be used as two independent atmospheric air mass tracers; only in oceanic and a few coastal stations do ^{7}Be and ^{210}Pb behave as two independent atmospheric tracers.

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