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A search for the seasonal variability on the depositional fluxes of ^{7}Be and ^{210}Pb

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Abstract. Investigations of the atmospheric fallout of ^{7}Be and ^{210}Pb in many places around the globe indicate that there are seasonal variations on the depositional fluxes of these nuclides: high in winter in a few places, while in most places the high occurs in spring. However, these earlier studies did not address quantitatively the corresponding seasonal variations of the amount of precipitation during these periods. Data on the depositional fluxes of ^{7}Be and ^{210}Pb in Galveston, Texas, during the years 1989-1991 indicate that the seasonal variation is not uniform from year to year, and the amount of precipitation primarily controls the depositional fluxes. All the recently published data on the depositional fluxes of these radionuclides to date are synthesized. Also, the seasonal variations of the depositional fluxes of ^{7}Be and ^{210}Pb due to the variations in the amounts of precipitation during these seasons are discussed. This study clearly shows that the depositional fluxes of ^{7}Be and ^{210}Pb during fall were minimum, and most of the time the fallout maxima were confined to either spring and/or summer. Interseasonal variations of the depositional fluxes of these nuclides do not remain the same. The data presented in this paper in conjunction with earlier published data seem to indicate that the observed seasonal increase in the depositional fluxes of ^{7}Be during spring is not due to the troposphere-stratosphere exchange of air masses during that season. The $^{7}Be/^{210}Pb$ activity ratio values remain the highest in summer and this is attributed to the increased rate of vertical transport of ^{7}Be (from the upper troposphere to the middle and lower troposphere) and ^{210}Pb (from the lower troposphere to upper troposphere) due to decreased stability of the troposphere during summer months.

Introduction

It has been shown that there are seasonal variations on the depositional fluxes of ^{7}Be and ^{210}Pb [*Matsunami et al.*, 1979; *Olsen et al.*, 1985; *Todd et al.*, 1989] (for other references, see work by *Baskaran et al.* [1993]). The seasonal increases in the depositional fluxes of these nuclides are commonly attributed to the injection of ^{7}Be-enriched stratospheric air to the troposphere [*Olsen et al.*, 1985; *Todd et al.*, 1989]. However, *Feely et al.* [1981, 1989] have addressed the seasonal variability of ^{7}Be at several locations around the globe from ^{7}Be measurements of air samples and have observed that several additional meteorological variables, such as seasonal variability of the amounts of precipitation and vertical mixing of lower and upper troposphere, could explain the seasonal variability of ^{7}Be concentration in surface air at many locations.

Even though a general linear relationship between the amount of precipitation and depositional fluxes of ^{7}Be and ^{210}Pb has been observed [*Olsen et al.*, 1985; *Dibb*, 1989; *Todd et al.*, 1989; *Schuler et al.*, 1991; *Baskaran et al.*, 1993] in many places, a quantitative evaluation on the precipitation-normalized seasonal variations of the depositional fluxes of ^{7}Be and ^{210}Pb is lacking.

The amount of precipitation in a particular location varies from season to season and from year to year. Since >90% of the depositional fluxes of atmospherically delivered nuclides are derived through wet precipitation, the amount of precipitation has a major influence on the depositional fluxes of ^{7}Be and ^{210}Pb [*Olsen et al.*, 1985; *Dibb*, 1989; *Todd et al.*, 1989; *Baskaran et al.*, 1993] even though production rate and transport are important even if they do not vary in time as precipitation does. Recently, it was found that pulse rain events, with ≥ 5 cm of rain in a single day, brought significant amounts of ^{7}Be and ^{210}Pb. For example, recent study on the depositional fluxes of ^{7}Be and ^{210}Pb at Galveston, Texas, indicated that 4-6% of the total number of rainy days, in the form of 4-6 heavy rain events, bring about 20-30% of the total annual deposition of ^{7}Be and ^{210}Pb [*Baskaran et al.*, 1993]. It has also been observed that the annual depositional fluxes of ^{7}Be and ^{210}Pb increase with the increase in the amount of precipitation [*Nevissi*, 1985; *Baskaran et al.*, 1993].

For a constant annual flux of ^{210}Pb or ^{7}Be, any increase in the depositional fluxes of ^{7}Be and ^{210}Pb during one season in a year should theoretically lead to a minimum depositional flux during another season in the same year. However, the season(s) where the depositional fluxes of these nuclides are consistently minima at different geographical location is not known. In the present paper, the seasonal variations of precipitation-normalized ^{7}Be and ^{210}Pb depositional fluxes at Galveston and College Station, Texas, are discussed. A fairly large database from different regions of the globe is available for the depositional fluxes of ^{7}Be or ^{210}Pb, but simultaneous depositional flux data of both nuclides are limited. Since ^{7}Be concentration in the air could be affected by stratosphere-troposphere exchange of air masses but not ^{210}Pb, both nuclides together only can provide information on the specific processes that control the seasonal variations of the atmospheric depositional fluxes of these nuclides. Also, prior to 1980 many of the rain collectors were not acidified (as discussed in work by *Turekian et al.* [1983]). As a result, the reported depositional fluxes were lower than actual values and thus these

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data also were not included. Available data on the depositional fluxes of ^{7}Be and ^{210}Pb measured simultaneously since 1980 are limited and are utilized in this paper to investigate the seasonal variability of ^{7}Be and ^{210}Pb when normalized to the amounts of precipitation during these seasons. Combining the fallout data at Galveston and College Station with the published literature data, the following four questions are addressed:

1. How do the precipitation-normalized depositional fluxes of ^{7}Be and ^{210}Pb vary from season to season?
2. Is there any clearly identifiable season in a year during which the depositional fluxes of ^{7}Be and ^{210}Pb are minimal?
3. Is there any significant increase in the depositional fluxes of ^{7}Be due to stratosphere-troposphere exchange of air masses during spring?
4. How do $^{7}Be/^{210}Pb$ activity ratios vary from season to season within a year as well as from year to year for the same seasons?

This is the first attempt to quantify seasonal variations of the precipitation-normalized depositional fluxes of ^{7}Be and ^{210}Pb.

Results and Discussion

Seasonal Variations of the Depositional Fluxes at Galveston and College Station

The depositional fluxes (activity per unit of area per unit of time) of ^{7}Be and ^{210}Pb during all 8 seasons of 1990 and 1991 for Galveston and only 1990 for College Station are plotted in Figures 1 and 2, respectively, and are presented in Table 1. Even though 12 seasons' worth of data were collected for Galveston and 8 seasons' worth for College Station, one sample in each place was lost during collection, and thus the seasonal fluxes were not calculated for that year. This resulted in 8 seasons for Galveston and 4 seasons for College Station. The monthly fluxes and the seasonal fluxes were calculated by assuming that the specific concentrations during the deployment period were constant and using the amounts of precipitation during the deployment period. The depositional fluxes of ^{7}Be during those 8 seasons at Galveston vary by more than a factor of 4, between 2.07 and 8.86 disintegrations per minute (dpm) cm^{-2} $season^{-1}$. In the case of ^{210}Pb, the seasonal depositional fluxes during that period varied almost by a factor of 6, between 0.115 and 0.653 dpm cm^{-2} $season^{-1}$.

The fractional amount of precipitation (equal to rainfall during a particular season divided by total annual rainfall) and the

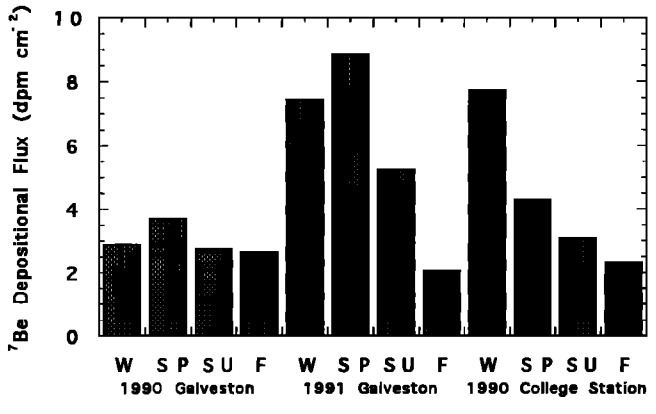

Figure 1. Seasonal depositional fluxes of ^{7}Be at Galveston in 1990 and 1991 and at College Station in 1990.

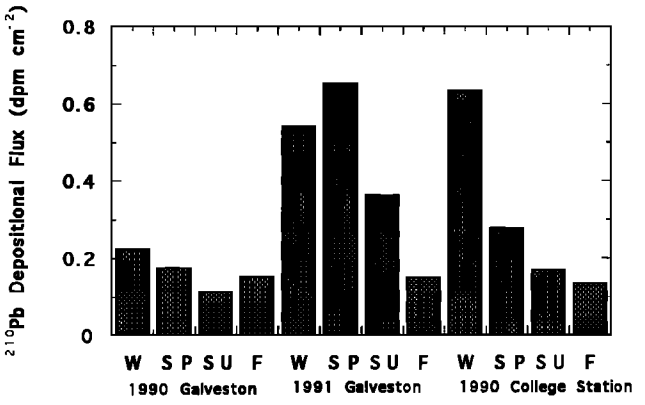

Figure 2. Seasonal depositional fluxes of ^{210}Pb at Galveston in 1990 and 1991 and at College Station in 1990.

fractional depositional fluxes (equal to depositional flux during a season divided by annual flux) of ^{7}Be and ^{210}Pb during all four seasons are plotted Figures 3 (^{7}Be) and 4 (^{210}Pb). As can be seen in Figures 3 and 4, the fractional depositional fluxes and fractional amount of rainfall for each season are higher in 1991 than in 1990, except for fall. These relatively higher depositional fluxes during winter, spring, and summer of 1991 are attributed to the increase in the amount of precipitation (annual rain = 150 cm) compared to 1990 (annual rain = 97 cm). The following observations are evident from Table 1 and Figures 3 and 4: (1) During fall, the fractional depositional fluxes of these nuclides are minimum compared to the fractional amount of precipitation at both Galveston and College Station. (2) Generally, the fractional depositional fluxes of ^{7}Be and ^{210}Pb during summer are not high and are comparable to the fractional amount of rainfall both for Galveston and College Station. (3) The fractional depositional fluxes of ^{7}Be and ^{210}Pb during winter were generally comparable to the fractional amount of rainfall during this season and in some cases slightly higher than the fractional amount of precipitation both for Galveston and College Station. (4) In spring 1990, the fractional depositional fluxes of ^{7}Be and fractional amounts of precipitation for Galveston and College Station were comparable, while for spring 1991 a seasonal maximum of ^{7}Be and ^{210}Pb fallout at Galveston is observed. The fractional depositional flux of ^{210}Pb at Galveston during spring 1990 was ~15% lower than the fractional amount of precipitation. For College Station, it was ~4% higher than the fractional amount of precipitation. Thus it appears that the seasonal increase in the depositional fluxes during spring is not the same from year to year for Galveston.

Atmospheric Processes That Cause Seasonal Variations in the Depositional Fluxes of ^{7}Be and ^{210}Pb

Since ^{7}Be is of cosmogenic origin and its production rate is high in the upper troposphere, its concentration in the air increases with increasing altitude from the surface of the Earth. Lead 210, on the other hand, has a higher production rate in the lower troposphere and relatively lower production rates in the upper troposphere and hence its concentration in air is expected to decrease with elevation from ground due to decrease in concentration of ^{222}Rn [*Moore and Poet*, 1976]. However, recent model prediction by *Balkanski et al.* [1993] suggests that there is a weak gradient of ^{210}Pb between the continental mixed layer and the troposphere. The longer residence times of ^{210}Pb aerosols in the tropics are attributed to the frequent convective updraft of

Table 1. Amount of Precipitation, Seasonal Depositional Fluxes of ^{7}Be and ^{210}Pb, Precipitation-Normalized Enrichment factors (α) for ^{7}Be and ^{210}Pb, and $^{7}Be/^{210}Pb$ Activity Ratios

Parameter	Winter (Dec., Jan., Feb.)	Spring (March, April, May)	Summer (June, July, Aug.)	Fall (Sept., Oct., Nov.)
		Galveston, 1990		
Precipitation (cm)	20.2	30.3	16.1	30.4
^{7}Be Fallout (dpm cm^{-2})	2.89	3.71	2.75	2.65
^{7}Be (α)	1.16	0.99	1.38	0.70
^{210}Pb Fallout (dpm cm^{-2})	0.225	0.177	0.115	0.153
^{210}Pb (α)	1.62	0.85	1.04	0.73
$^{7}Be/^{210}Pb$ Ratio	12.8	21.0	23.9	17.3
		Galveston, 1991		
Precipitation (cm)	48.3	37.2	34.2	30.3
^{7}Be Fallout (dpm cm^{-2})	7.45	8.86	5.24	2.07
^{7}Be (α)	1.00	1.47	0.99	0.44
^{210}Pb Fallout (dpm cm^{-2})	0.542	0.653	0.364	0.150
^{210}Pb (α)	0.98	1.54	0.93	0.44
$^{7}Be/^{210}Pb$ Ratio	13.7	13.6	14.4	13.8
		*College Station, 1990**		
Precipitation (cm)	40.5	23.7	14.3	21.5
^{7}Be Fallout (dpm cm^{-2})	7.74	4.29	3.09	2.32
^{7}Be (α)	1.28	0.97	0.98	0.52
^{210}Pb Fallout (dpm cm^{-2})	0.634	0.280	0.170	0.136
^{210}Pb (α)	1.10	1.04	1.24	0.62
$^{7}Be/^{210}Pb$ Ratio	12.2	15.3	18.2	17.1

dpm, disintegrations per minute.
*The collection period is between March 1990 and February 1991.

^{222}Rn to higher altitudes where precipitation is infrequent. There seems to be no published data to confirm this model prediction. Owing to the distinctly different sources of ^{210}Pb and ^{7}Be, steep concentration gradients between the upper and lower troposphere are commonly maintained. Seasonal variations in the surface air concentrations of ^{7}Be as well as in the depositional fluxes of ^{7}Be and ^{210}Pb have been observed in many places. It has been shown that the surface air concentration of ^{7}Be depends on wet scavenging, stratosphere-troposphere exchange of air masses, downward transfer of aerosols in the troposphere, and horizontal movement of aerosols from subtropical and lower and higher latitudes [*Feely et al.*, 1981, 1989; *Brost et al.*, 1991]. The seasonal variation in the depositional fluxes of ^{7}Be as well as in the surface air is a complicated function of all four processes since

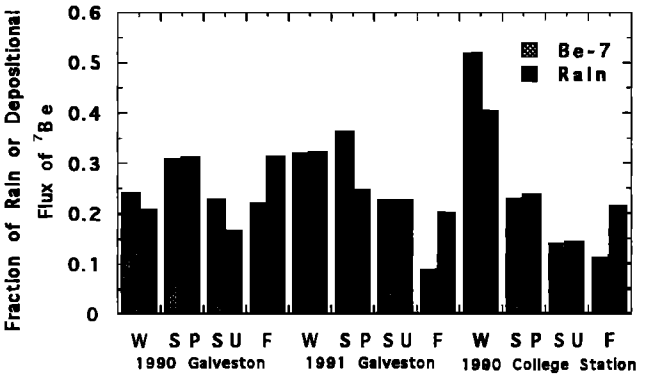

Figure 3. Fractional amounts of rainfall and depositional fluxes of ^{7}Be in 1990 and 1991 at Galveston and in 1990 at College Station.

each of these processes can have its own seasonal cycle [*Brost et al.*, 1991]. For example, stratosphere-troposphere exchange results in a peak concentration of surface ^{7}Be in spring and summer near midlatitude regions especially at high elevations. During the summer months, the troposphere is less stable than in the winter due to the heating of the air in contact with the surface resulting in convective circulation. The consequence of this convective circulation is in more downward transfer of aerosols into the lower troposphere in summer. Certain midlatitude continental stations might have concentration maxima in summer produced by this process [*Brost et al.*, 1991]). *Feely et al.* [1981, 1989] have carried out the most detailed study on the seasonal variations in surface air concentrations of ^{7}Be using data sets from 28 sites in different regions around the world. The seasonal variations in the depositional flux, which are related to the surface air concentrations, are caused by at least three factors: (1) seasonal variations in the amount of precipitation which causes the changes in depositional fluxes as well as in air concentrations; (2) increased stratosphere-troposphere exchange during the late winter and early spring seasons (this process should result in a higher depositional flux of ^{7}Be but not ^{210}Pb during late winter and early spring); and (3) increased vertical transport rate of ^{7}Be from the upper troposphere to the middle and lower troposphere (for ^{210}Pb, from the lower troposphere to upper troposphere) due to decreased stability of the troposphere during the summer months. The causes for the seasonal variations of the depositional fluxes of ^{7}Be and ^{210}Pb are discussed below.

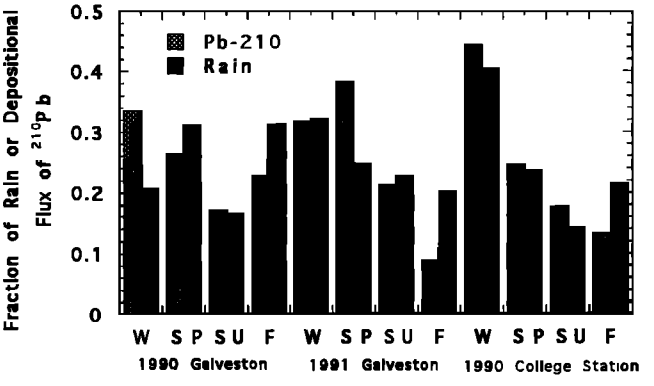

Figure 4. Fractional amounts of rainfall and depositional fluxes of ^{210}Pb in 1990 and 1991 at Galveston and in 1990 at College Station.

Precipitation-Normalized Depositional Fluxes of ^{7}Be and ^{210}Pb During Different Seasons

From the published data of ^{7}Be and ^{210}Pb depositional fluxes, it is possible to quantify the seasonal variations of the precipitation-normalized depositional fluxes. In addition to the data presented in this paper, the following published data also were utilized in the data analysis: New Haven, Connecticut (41°N [*Turekian et al.*, 1983]), Bermuda (33°N [*Turekian et al.*, 1983]), Oak Ridge, Tennessee (35°58'N [*Olsen et al.*, 1985]), Norfolk, Virginia (36°53'N [*Todd et al.*, 1989]), and Paris, France (48.8°N [*Thomas*, 1988]). The precipitation-normalized enrichment factor α is defined as

$$\alpha = (F_S/F_t)/(R_S/R_t)$$

where R_S and R_t are the amount of rainfall during a particular season and in one year, respectively, and F_S and F_t are the corresponding depositional fluxes in that particular season and year, respectively, of the radionuclide, ^{7}Be or ^{210}Pb. It is pertinent to point out that the seasonal variations in α reflects the seasonal variations in the washout ratio. Values greater than 1.0 indicate that the depositional fluxes were higher than expected from the amount of rainfall, and values less than 1.0 indicate the depletion of radionuclide fluxes. Table 1 contains the values of α for all 4 seasons in 1 year. The error in the values of α should be less than 10%, since it is the ratio of the seasonal depositional fluxes to the annual depositional fluxes. Assuming an upper limit of 10% error on the measurement of the depositional fluxes of ^{7}Be and ^{210}Pb, values below 0.9 and above 1.1 are considered significant.

Atmospheric Processes That Influence the Seasonal Variability of the Depositional Fluxes of ^{7}Be and ^{210}Pb From Galveston, College Station, and Other Published Data

As mentioned earlier, of the three atmospheric processes that could cause the seasonal variations in the depositional fluxes of ^{7}Be and ^{210}Pb, the relative importance of each one can be evaluated using the data presented in Table 1. During fall 1990 and 1991, both at Galveston and College Station the α values for ^{7}Be and ^{210}Pb are considerably less than 1. There appears to be no simultaneous seasonal increase in the precipitation-normalized depositional fluxes of ^{7}Be or ^{210}Pb in 1990 at either Galveston or College Station. The α values for ^{7}Be and ^{210}Pb during spring 1991 are 1.47 and 1.54, respectively (Table 1). The higher α values of ^{7}Be (1.38) relative to ^{210}Pb (1.04) during summer 1990 at Galveston can be attributed to the increased vertical rate of transport of ^{7}Be from the upper troposphere to the middle and low troposphere. In College Station, during summer 1990, the fractional depositional flux of ^{210}Pb (α=1.24, for ^{7}Be, α=0.98) appears to be enriched, and this is likely due to the continental influence. Also, there is a clear increase in depositional fluxes of both ^{7}Be and ^{210}Pb during winter 1990 at both Galveston and College Station, while there is no such increase in the depositional fluxes of ^{7}Be or ^{210}Pb observed for Galveston in 1991.

The fractional depositional fluxes of ^{7}Be and ^{210}Pb for all the stations listed in Table 2 are plotted against fractional rainfall in Figures 5a (winter), 5b (spring), 5c (summer), and 5d (fall). All the data points with $\alpha > 1$ (slope = α) correspond to higher precipitation-normalized depositional fluxes and those with $\alpha < 1$ to lower values. For ^{210}Pb during winter, seven data points correspond to samples with values having $\alpha > 1$ and three with $\alpha < 1$, while one point corresponds to a sample with an α value of 1. On the other hand, for ^{7}Be, four data points have a slope of $\alpha > 1$, five points $\alpha < 1$, and one point corresponds to a sample with an α value of 1. Thus, it appears that the fraction of depositional fluxes of ^{210}Pb are slightly higher during winter than are those of ^{7}Be. In spring (Figure 5b), most of the data points for both ^{7}Be (eight points >1, two ~1, and one <1) and ^{210}Pb (seven points >1, one ~1, and three <1) have α values >1 and thus indicate that in most places the depositional fluxes were higher relative to the amount of precipitation. Combining this observation with what was observed at Galveston, it appears that the depositional fluxes of ^{7}Be and ^{210}Pb are generally higher during spring, but it need not be a rule for any particular site. Most of the α values during summer (Figure 5c) are >1, similar to the spring values. In other words, during the spring and summer seasons, generally, the depositional fluxes of ^{7}Be and ^{210}Pb relative to the amount of precipitation are higher than during winter. A plot of the fractional depositional fluxes of ^{7}Be and ^{210}Pb plotted against the fractional amounts of precipitation in fall clearly shows that for almost all samples (except one data point for ^{7}Be) the depositional fluxes of ^{7}Be and ^{210}Pb are lower in fall than in the other three seasons (Figure 5d).

Is There Any Increase in the Depositional Flux of ^{7}Be During Spring due to Troposphere and Stratosphere Exchange of Air Masses?

Even though most of the ^{7}Be production, like the other cosmogenic radionuclides, takes place in the stratosphere, only a fraction of this ^{7}Be will reach the Earth's surface when advection of stratospheric air into the troposphere takes place. It has been shown that the stratospheric component was not delivered uniformly throughout the year; it accounted for only ~25% of the ^{7}Be in surface air on an annual basis and accounted for ~40% of the observed ^{7}Be depositional flux in spring, but only for ~10% of the depositional flux during fall [*Dutkiewicz and Husain*, 1985]. This low contribution from stratosphere to the troposphere is due to the 4-5 times longer residence times of stratospheric aerosols (~1 year) compared to ^{7}Be mean life (77 days) [*Dutkiewicz and Husain*, 1985]. However, the stratospheric source becomes significant during the spring when midlatitude folding of the tropopause enhances the stratospheric-tropospheric exchange. Maximum depositional flux of ^{7}Be is expected at midlatitude, high-elevation stations [*Brost et al.*, 1991]. If this stratospheric source is a significant one for the observed increase in the depositional flux of ^{7}Be during spring, then the ratio of the fractional depositional flux of ^{7}Be to the amount of precipitation should increase from lower latitudes to midlatitudes. This ratio (α, as given in equation (1)), is given in Table 2 and plotted in Figure 6. From this figure, there are evidently only two stations (New Haven and Paris) which fall within this midlatitude belt (38°-51°N). There were no significant maxima observed during spring for these two stations. Consequently, the input to the depositional flux from stratosphere-troposphere exchange during spring at all these stations should be negligible. It is also possible that infrequent precipitation events at any given site may result in the differences in the seasonal air concentration and depositional fluxes of these nuclides in that site. On the contrary, the precipitation normalized enrichment factors for ^{7}Be in spring are 1.04 and 1.06 for New Haven and Paris, respectively. This value, α, for ^{210}Pb (=1.12) is even higher for New Haven.

In order to investigate if the precipitation-normalized depositional fluxes of ^{7}Be and ^{210}Pb are related in spring, the precipitation-normalized enrichment factor (α) for ^{210}Pb is

Table 2. Amounts of Precipitation and Precipitation Normalized Enrichment Factor for ^{7}Be and ^{210}Pb During all Four Seasons

Time of Collection	Rainfall (cm)	α, ^{7}Be				α, ^{210}Pb				Source
		Winter	Spring	Summer	Fall	Winter	Spring	Summer	Fall	
					New Haven, Conn., 41°N; 72.2°W					
March 1977 - Feb. 1978	148.1	0.61	1.04	1.37	1.05	0.90	1.12	1.30	0.85	*Turekian et al.* [1983]
					Bermuda, 33°N; 65°W					
Sept 1977- Aug. 1978	169.9	NM	0.78	NM	0.84	1.48	1.23	1.05	0.54	*Turekian et al.* [1983]
					Oak Ridge, Tenn. 35°58'N, 84°17'W					
Sept. 1982 - Aug. 1983	110	0.73	1.18	1.88	0.66	1.06	1.01	1.92	0.63	*Olsen et al.* [1985]
Sept. 1983 - Aug. 1984	143	0.84	1.31	1.13	0.61	1.10	1.17	1.20	0.56	
					Norfolk, Va., 36°53'N; 76°18'W					
Dec. 1982 - Nov. 1983	132	1.12	1.25	1.36	0.51	1.37	1.16	1.15	0.50	*Todd et al.* [1989]
Dec. 1983 - Nov. 1984	141	0.94	1.18	0.91	0.92	0.75	1.20	1.03	0.99	
					Paris, France, 48.8°N; 2.3°E					
Jan. 1982 - Dec. 1982	64.6	1.22	1.06	1.23	0.65	1.30	0.90	1.22	0.75	*Thomas* [1988]
Jan. 1983 - Dec. 1983	65.1	0.51	1.04	1.47	0.98	0.55	0.66	1.95	0.91	*Thomas* [1988]
					College Staion, Tex., 30°35' N; 96°22'W					
Mar. 1990 - Feb. 1991	128	1.28	0.966	0.98	0.52	1.10	1.04	1.24	0.62	This paper
					Galveston, Tex., 29°18' N; 94°48'W					
Dec. 1989 - Nov. 1990	97	1.16	0.99	1.38	0.70	1.62	0.85	1.04	0.73	This paper
Dec. 1990 - Nov. 1991	150	1.00	1.47	0.99	0.44	0.98	1.54	0.93	0.44	

Enrichment factor, α, annual rainfall times seasonal flux of a radionuclide/(amount of rainfall in a season times annual flux of radionuclide); NM, not measured.
*Based on 8 months of precipitation data.

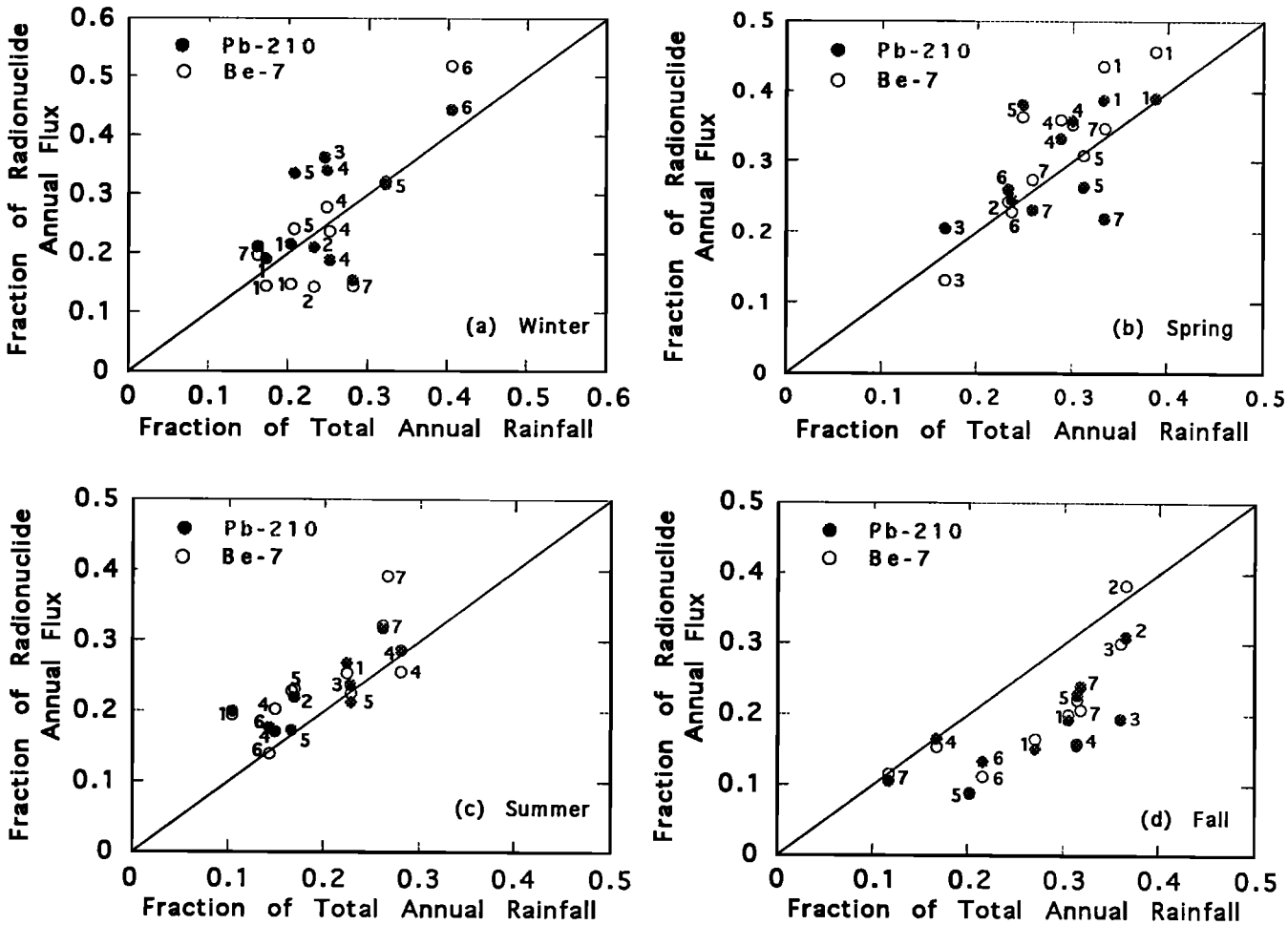

Figure 5. Fractional amounts of ^{7}Be and ^{210}Pb fluxes (depositional flux in a season/annual depositional flux) plotted against fractional amount of rainfall (rainfall in a season/annual rain fall) for (a) winter, (b) spring, (c) summer, and (d) fall. Those data points which fall above the 45° diagonal line (slope=1) have seasonal increase in the depositional flux of that nuclide (^{7}Be or ^{210}Pb). The key for the numbers given are as follows: 1, Oak Ridge, Tennesse [*Olsen et al.*, 1985]; 2, New Haven, Connecticut [*Turekian et al.*, 1983]; 3, Bermuda [*Turekian et al.*, 1983]; 4, Norfolk, Virginia [*Todd et al.*, 1989]; 5, Galveston, Texas [*Baskaran et al.*, 1993]; 6, College Station, TX [*Baskaran et al.*, 1993], and 7, Paris, France [*Thomas*, 1988].

plotted against the α values of ^{7}Be in Figure 7. When the data points lie on the 45° diagonal line (slope = 1), this would indicate that there are no enrichments of one nuclide over the other for those stations. In cases where the data points fall above the slope = 1 line, there is a seasonal increase of ^{210}Pb over ^{7}Be. There are about four points lying above and five points lying below the line. This observation appears to suggest that in some spring seasons, there is a seasonal increase of ^{210}Pb over ^{7}Be. There are two alternate explanations possible for Figures 6 and 7:

1. There is an increased transfer of ^{7}Be from the stratosphere to the troposphere in the spring, as observed by *Dutkiewicz and Husain* [1985], but this increase does not result in a measurable increase in the ^{7}Be deposition. Or,

2. The contribution of stratosphere-derived depositional flux of ^{7}Be to all the stations reported in Table 2 is insignificant and can be attributed to the changes in the atmospheric processes (described below) during spring as compared to fall, for example.

Taking the α values of both ^{7}Be and ^{210}Pb during different seasons (Tables 1 and 2) at various places into consideration, I

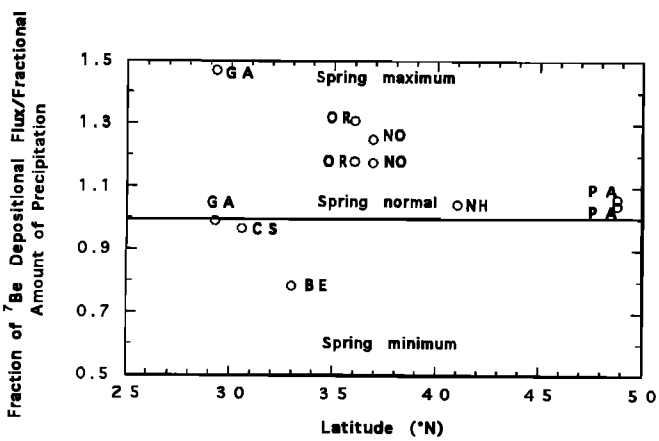

Figure 6. The precipitation-normalized enrichment factor [α = annual rainfall times seasonal flux of a radionuclide/amount of rainfall in a season times annual flux of radionuclide] is plotted against latitude. The central line with y value = 1 indicates normal fallout; samples with data points above this line have higher fallout than expected from the amount of precipitation, and samples with data points below this line have lower fallout. BE: Bermuda; CS: College Station; GA: Galveston; NH: New Haven; NO: Norfolk; OR: Oak Ridge; PA: Paris, France.

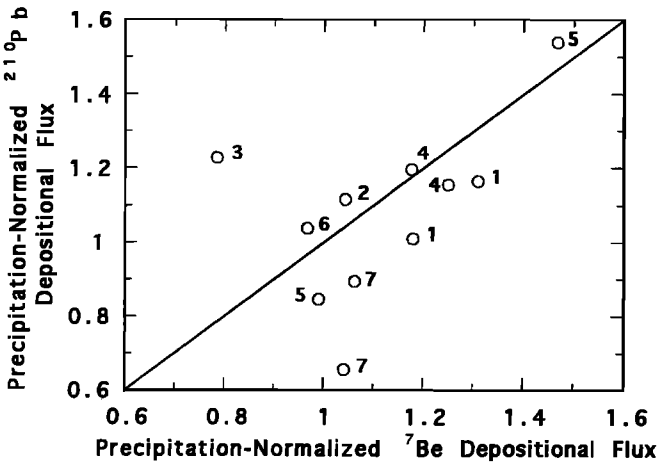

Figure 7. The precipitation-normalized ^{210}Pb flux data in spring [α = annual rainfall times seasonal flux of ^{210}Pb/amount of rainfall in spring times annual flux of ^{210}Pb] is plotted against precipitation-normalized ^{7}Be flux data in spring [α = annual rainfall times seasonal flux of ^{7}Be/amount of rainfall in spring times annual flux of ^{7}Be]. Data points below 45° diagonal line have lower depositional flux for ^{7}Be while those above the line have higher ^{210}Pb flux. The key for the numbers given is as follows: 1, Oak Ridge, Tennessee [*Olsen et al.*, 1985]; 2, New Haven, Connecticut [*Turekian et al.*, 1983]; 3, Bermuda [*Turekian et al.*, 1983]; 4, Norfolk, Virginia [*Todd et al.*, 1989]; 5, Galveston, Texas [*Baskaran et al.*, 1993]; 6, College Station, Texas [*Baskaran et al.*, 1993], and 7, Paris, France [*Thomas*, 1988].

hypothesize that the slight increase in seasonal depositional fluxes during spring is due to changes in the atmospheric processes rather than the contribution from stratosphere-troposphere exchanges of air masses. This hypothesis is based on the observation that the α values of both ^{7}Be and ^{210}Pb generally increase simultaneously for a particular season (Table 2 and Figure 7). However, *Balkanski et al.* [1993] have predicted that the ^{210}Pb concentrations in the free troposphere in summer are much higher than expected and this prediction is attributed to the frequent convective pumping of ^{222}Rn to high altitudes in the tropics and at northern midlatitudes. In latitudes between the tropics and midlatitudes (23°-38°N), the convective updrafts are not that dominant, and thus one would not see this effect in other seasons. But Table 2 clearly shows that α values for ^{210}Pb for spring are 1.23 (33°N), 1.17 (35°58'N), 1.16 and 1.20 (36°53'N), 1.04 (30°35'N), and 0.85 and 1.54 (29°18'N). Thus, even if there is no convective updraft of ^{222}Rn in the latitude belt of 23°-38°, there is still a seasonal increase of ^{210}Pb in this latitude belt. The other factors that could play an important role in the seasonal variations of the depositional fluxes of radionuclides are history of air masses reaching the sampling sites and seasonal variation in cloud altitude (W. C. Graustein, personal communication, 1994).

Seasonal Variations on the $^{7}Be/^{210}Pb$ Activity Ratio

Although the production source functions of these two nuclides are distinctly different, a relatively high correlation between the ^{7}Be fluxes and ^{210}Pb fluxes has been observed in different places (Japan [*Matsunami et al.*, 1979], Oak Ridge [*Olsen et al.*, 1985], Norfolk [*Todd et al.*, 1989], and College Station and Galveston [*Baskaran et al.*, 1993]). This high correlation between the fluxes of ^{7}Be and ^{210}Pb appears to suggest that the atmospheric removal behavior of ^{7}Be and ^{210}Pb are relatively similar, and that these two nuclides cannot be used as two independent atmospheric tracers [*Todd et al.*, 1989; *Baskaran et al.*, 1993]. Since the seasonal trend on the depositional flux of ^{7}Be is stronger than that of ^{210}Pb, as has been observed by *Todd et al.* [1989], there ought to be a seasonal variation of the activity ratios of $^{7}Be/^{210}Pb$.

The $^{7}Be/^{210}Pb$ activity ratio for the years 1990 and 1991 for Galveston and 1990 for College Station are given in Table 1 and are plotted in Figure 8. During 1990, the ratios varied between 12.8 and 23.9 for the four seasons at Galveston, with the highest value in summer. In College Station also, the ratio is the highest in summer (18.2), with values ranging between 12.2 and 18.2 for the four seasons in 1990. This consistently higher ratio in summer can be attributed to dynamic atmospheric conditions such as internal heating and subsequent mixing of the troposphere that could bring more ocean-derived air masses than in other seasons. During the summer months, solar heating of the Earth's surface leads to heating of air masses in contact with the surface. This less dense, warm air, which is enriched in ^{210}Pb and depleted in ^{7}Be, is forced upward and replaced by denser, colder air that is enriched in ^{7}Be and depleted in ^{210}Pb. The convection circulation that is produced by this heating will result in more ^{7}Be and possibly less ^{210}Pb in the air and thus the $^{7}Be/^{210}Pb$ activity ratio in the air as well as in the precipitation samples is expected to be the highest in summer.

The lower ratios of $^{7}Be/^{210}Pb$ during spring and summer in 1990 at College Station compared to the corresponding values at Galveston can be attributed to the relatively higher oceanic influence (oceanic air masses bring lower amounts of ^{210}Pb compared to the continental air masses) at Galveston compared to College Station. In 1990, at both Galveston and College Station, the ratio varied from season to season. This is also clearly seen from the seasonal depositional fluxes of ^{210}Pb presented in Table 1. In winter and fall, the values in Galveston (12.8 in winter and 17.3 in fall) and College Station (12.2 in winter and 17.3 in fall) are about the same. This can be attributed to the relatively calm atmospheric conditions during these seasons. On the contrary, during spring and summer, the Galveston values are higher than the corresponding values at College Station. Galveston, being a coastal station, will receive more oceanic air mass than College Station. Also, the oceanic air masses could pick up some continent-derived aerosols which are enriched in ^{210}Pb during their transits inland. Since data on the concentrations of ^{7}Be and ^{210}Pb in the air samples at different altitudes are not available, it is not possible to draw a firm conclusion on this aspect.

The seasonal variations of $^{7}Be/^{210}Pb$ activity ratio during 1990 (12.8-23.9) at Galveston are much larger than those of 1991 (13.6-

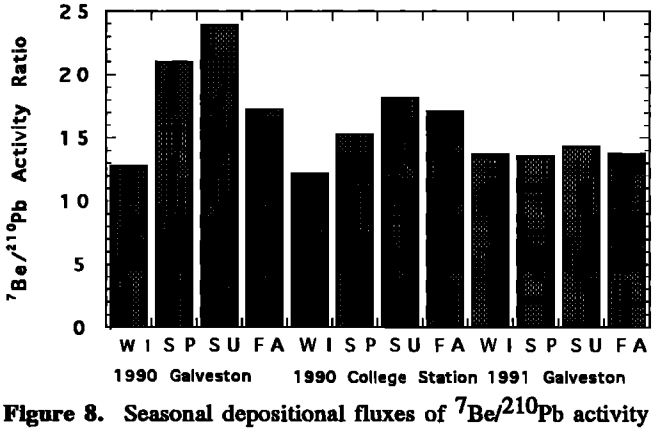

Figure 8. Seasonal depositional fluxes of $^{7}Be/^{210}Pb$ activity ratio at Galveston in 1990 and 1991 and at College Station in 1990.

14.4). The year 1990 had the highest rainfall. In 1991, the seasonal rainfall during spring, summer, and fall were comparable, 37, 34, and 30 cm, respectively, and thus it appears that the nature of air masses that bring precipitation control the seasonal activity ratio of the precipitation.

Conclusions

The seasonal variations in the depositional fluxes for the naturally occurring radionuclides ^{7}Be and ^{210}Pb have been determined at Galveston and College Station, Texas. Data from a total number of seven stations for which the depositional fluxes of ^{7}Be and ^{210}Pb are available for a period of 1-2 years were analyzed for the seasonal variations of the precipitation-normalized depositional fluxes of these nuclides. Even though the data set represents sampling stations from an oceanic station (Bermuda), coastal stations (Galveston, Texas, New Haven, Connecticut and Norfolk, Virginia) and a continental station (Paris, France), the data is limited. Information obtained from this present study will be useful in understanding the seasonal variations on the atmospheric delivery of other pollutants that geochemically resemble these nuclides [*Graustein and Turekian*, 1983; *Turekian et al.*, 1983]. This present investigation leads me to draw the following conclusions:

1. The depositional fluxes of ^{7}Be and ^{210}Pb at Galveston and College Station during the years 1990 and 1991 are observed to be minimum during fall. Published data on the depositional fluxes from other places do indicate clearly that the depositional fluxes of these nuclides in most places are minimum during fall. There is a general increase in the fraction of depositional fluxes during spring and/or summer.

2. The precipitation-normalized depositional fluxes of ^{7}Be and ^{210}Pb are generally higher during spring and summer, but it need not be a rule for any particular site.

3. There is no significant increase on the depositional fluxes of ^{7}Be during spring at Galveston or College Station; also, the data analysis of the published data on the depositional flux of ^{7}Be and ^{210}Pb for the seasonal variations of the precipitation-normalized depositional fluxes of these nuclides seems to indicate that the slight increase in the depositional flux of ^{7}Be (as well as of ^{210}Pb) is not due to the stratosphere-troposphere exchange, contrary to the suggestion by earlier work. I hypothesize that the increase in the depositional flux of ^{7}Be (and ^{210}Pb) is attributed to the vertical mixing of the troposphere due to heating in the late spring and summer months. The seasonal increase in the depositional fluxes during spring is not the same from year to year for Galveston.

4. In the years when the annual rainfall is much higher than the long-term annual average rainfall, the interseason variability of $^{7}Be/^{210}Pb$ activity ratio seems to be rather narrow. This ratio is the highest in the summer and this is attributed to the internal heating of the troposphere and subsequent deep vertical mixing of the troposphere, even though other factors such as altitude of origination of precipitation could also affect the $^{7}Be/^{210}Pb$ activity ratios.

Future work needs to address the relationship between the variability of $^{7}Be/^{210}Pb$ activity ratio and the atmospheric processes that cause such variability in individual rain events. The atmospheric processes that cause the distinctly lower depositional fluxes during certain seasons, such as fall, need to be quantified.

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