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Depositional characteristics of ^7Be and ^{210}Pb in southeastern Michigan

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[1] The depositional fluxes in the bulk and dry fallout as well as the concentrations of ^7Be and ^{210}Pb in aerosols were measured for a period of 17 months at Detroit, Michigan. The bulk depositional fluxes of ^7Be and ^{210}Pb varied between 3.11 and 63.0 $\text{dpm cm}^{-2} \text{yr}^{-1}$ (mean of 11.4 $\text{dpm cm}^{-2} \text{yr}^{-1}$) and 0.35 and 10.3 $\text{dpm cm}^{-2} \text{yr}^{-1}$ (mean of 1.41 $\text{dpm cm}^{-2} \text{yr}^{-1}$), respectively, and this variability in the depositional fluxes is attributed to the frequency and amount of precipitation and seasonal variations in the depositional fluxes. The dry depositional fluxes of ^7Be and ^{210}Pb contributed 2.1–19.8% and 3.6–48.6% of the bulk depositional fluxes, respectively. The higher precipitation-normalized enrichment factor for ^7Be during spring is attributed to the stratosphere-troposphere exchange of air masses during spring and large-scale atmospheric mixing during summer. We report that snow is more efficient than rain in removing ^{210}Pb from the atmosphere, and this is likely due to higher surface area of snow flurries compared to the rain droplets. The volume-weighted concentrations of ^7Be and ^{210}Pb are the highest reported in literature so far. The concentration of ^7Be in the air showed a seasonal increase during the spring and summer months, and no seasonal variation was observed for ^{210}Pb . The mean deposition velocities of aerosols calculated using concentrations of ^7Be and ^{210}Pb in air and precipitation are 1.6 and 1.1 cm s^{-1} , respectively, and the corresponding washout ratios are 948 and 637. There is no significant correlation between the aerosol mass and depositional velocities of ^7Be and ^{210}Pb in the air masses. We propose that a relatively minor portion of the aerosols actively participate in the removal of these nuclides from the air

masses. **INDEX TERMS:** 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0330 Atmospheric Composition and Structure: Geochemical cycles; 4860 Oceanography: Biological and Chemical: Radioactivity and radioisotopes; **KEYWORDS:** deposition velocity, washout ratio, depositional fluxes of ^7Be and ^{210}Pb , enrichment factor, concentration of ^{210}Pb and ^7Be in air

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1. Introduction

[2] Naturally occurring beryllium-7 and the daughter products of ^{222}Rn , such as ^{210}Pb and ^{210}Po , have been proven useful as tracers and have been utilized to gain insight into rates of sediment mixing, sedimentation, dynamics of particle transport, fate of particle-reactive contaminants as well as meteorological information especially on the origin of air masses and residence times of aerosols [e.g., Lal, 1968; Wogman *et al.*, 1968; Poet *et al.*, 1972; Young and Silker, 1974; Robbins, 1978; Benninger *et al.*, 1979; Krishnaswami *et al.*, 1980; Dominik *et al.*, 1989; Kritz *et al.*, 1991; Baskaran *et al.*, 1997; Sommerfield *et al.*, 1999; Steinmann *et al.*, 1999; Fitzgerald *et al.*, 2001]. In addition, these tracers have been utilized as aerosol tracers to study the vertical transport and residence time of aerosols in the troposphere using modified versions of the general

circulation models [Brost *et al.*, 1991; Balkanski *et al.*, 1993; Koch *et al.*, 1996]. The source terms of these radionuclides are relatively well known, and are removed from the atmosphere by precipitation, dry fallout, and radioactive decay, as well as removal by coagulation of smaller radionuclide-laden particles followed by gravitational settling. Knowledge of the behavior of ^7Be and ^{210}Pb in the atmosphere will yield insight on the behavior of other similar chemical species in the atmosphere. For example, chemical species that undergoes gas-to-particle conversion similar to ^{222}Rn decay will behave like ^{210}Pb . If the chemical species is supplied from the stratosphere by settling (such as debris from nuclear testing) or produced in the upper troposphere it will behave like ^7Be .

[3] Beryllium-7 ($T_{1/2} = 53.3$ days) is a cosmogenic radionuclide produced in the stratosphere when cosmic rays bombard the nucleus of oxygen and nitrogen atoms in the atmosphere [Lal *et al.*, 1958] and its flux to the Earth's surface has a latitudinal dependence [Lal and Peters, 1967], similar to other cosmogenic nuclides. The concentration of

^7Be in the atmosphere increases with altitude from the surface of the Earth and its production rate for a given latitude (as well as the flux) on the Earth's surface is independent of longitude. Due to its short half-life, most of the ^7Be that are produced in the stratosphere do not readily reach the troposphere except during spring when seasonal thinning of the tropopause takes place at mid-latitudes, resulting in air exchange between the stratosphere and troposphere. In most atmospheric studies, the ^7Be concentration in aerosols does not remain constant with time and show seasonal variations in surface air. A high concentration of ^7Be in surface air aerosols would indicate significant input from the upper troposphere-lower stratosphere regions [Feely *et al.*, 1989; Dibb *et al.*, 1994]. At mid-latitudes, seasonal variations of air concentration of ^7Be in the spring have been observed because air masses with higher concentrations of ^7Be in the stratosphere are injected into the troposphere during spring. Radon-222 ($T_{1/2} = 3.8$ days), one of the daughter-products in the ^{238}U decay chain, predominantly emanates from the Earth's continental crust. The ^{222}Rn flux from soils ranges from 0.7 to 4.45 dpm $\text{cm}^{-2} \text{yr}^{-1}$, with a global average value 0.9 dpm $\text{cm}^{-2} \text{yr}^{-1}$ and is about 100 times higher than the oceanic flux [Turekian *et al.*, 1977]. Radon-222 undergoes radioactive decay in the atmosphere producing several daughter products, including ^{210}Pb . These nuclides are scavenged by aerosols and subsequently are removed from the atmosphere, primarily by precipitation. In areas where the amount of rainfall is low, it is likely that the dry fallout could be a significant component of the bulk depositional flux while in areas where there is a relatively higher amount of rainfall, the wet fallout is likely the major component (>90%) of the bulk depositional flux.

[4] In general, the amount of rainfall controls the seasonal variations of the depositional fluxes of ^7Be [Baskaran, 1995], while precipitation-normalized depositional fluxes during spring are higher in the middle latitudes [Baskaran, 1995; Dueñas *et al.*, 2001]. However, how the changes in the amount of precipitation and the depositional fluxes affect the concentrations of ^7Be and ^{210}Pb in air is not fully addressed. Rainout and washout events will reduce the residence times of these nuclides in the lower atmosphere considerably. Information on the ^{210}Pb - and ^7Be -derived deposition velocity and washout ratios of aerosols is useful to determine the fate of other atmospheric contaminants that behave similar to these nuclides.

[5] A considerable body of literature exists on the depositional fluxes of ^7Be and ^{210}Pb as well as the concentrations of these nuclides in the aerosols. However, combined measurements of these nuclides in the dry and bulk fallout as well as in aerosols are very limited. In addition, the data on the concentrations in aerosols and bulk depositional fluxes of ^7Be and ^{210}Pb in the midwestern region of the United States are limited. The specific objectives of this present investigation are as follows: (1) to determine the depositional fluxes of ^7Be and ^{210}Pb in the bulk and dry fallout and investigate if there are any seasonal variations in the precipitation-normalized depositional fluxes of these nuclides; (2) to determine the concentrations of ^7Be and ^{210}Pb in aerosols and investigate if there is any relationship between the concentration in air and the depositional flux during that time; (3) to determine the deposition velocity of

aerosols using the concentrations of ^{210}Pb and ^7Be in aerosols and precipitation and determine if there is any relationship between the deposition velocities obtained from ^7Be and ^{210}Pb ; (4) to determine the washout ratios for ^7Be and ^{210}Pb and compare the washout ratios obtained using these two nuclides; and (5) to determine whether ^7Be and ^{210}Pb can be used as independent atmospheric tracers.

2. Methods and Materials

2.1. Collection of Bulk and Dry Fallout and Aerosol Samples

[6] The sampling site is one of the air monitoring network stations operated by the Wayne County Air Quality Management Division and jointly operated by the Wayne County and the Michigan Department of Environmental Quality (MDEQ), under cooperative agreement with the U. S. Environmental Protection Agency (EPA). A bulk rain collector (200-L polyethylene drum with surface area of 2800 cm^2) was deployed in September 1999 at a site in the southwest area of Detroit, Michigan ($42^\circ 25' \text{N}$; $83^\circ 1' \text{W}$; 175 m above mean sea level) at $\sim 1\text{m}$ above the ground to prevent the resuspension of dust particles getting into the collector. The lid of the bulk collector was deployed as the dry collector in October 1999 on the roof of a building at the same site at ~ 4 m above ground. To prevent any potential adsorption of ^7Be and ^{210}Pb onto the walls of the collector, both dry and wet collectors were acidified prior to deployment with concentrated HCl and chemical yield spikes [1-ml of stable Pb ($\equiv 1$ mg, Aldrich 05618DR) and Be ($\equiv 1$ mg, Baker 6921-01)] were added. The bulk rain samples were collected after each major precipitation event or once in about a month and after ~ 10 days of dry weather for the dry collector. Immediately after collection, the drum and/or the lid was cleaned with repeated rinses of 6M HCl to remove adsorbed Be and Pb isotopes from the surface of the drum and/or the lid. The rinsings were combined with the rain sample and subsequently the solutions were further processed.

[7] A high-volume sampler, operated by the Wayne County Air Quality Management Division, loaded with a pre-weighed high purity Glass Microfibre filter (GFF) was used to collect aerosol samples at the ground level. The high-volume sampler was continuously run for 1–3 days weekly. This ordinarily allows total suspended particulates (TSP) with diameters of 0.1 to 100 μm to collect on the filter surface. The mass concentration of suspended load collected on the GFF filter was determined gravimetrically.

2.2. Radiochemical Processing of Bulk and Dry Fallout and Aerosol Samples

[8] The chemical processing of the collected samples is similar to that described by Baskaran *et al.* [1993]. Briefly, the collected (bulk and dry) samples were transferred to a pre-cleaned 3-liter Pyrex glass beaker in the lab and evaporated to $\sim 100\text{-ml}$. In many of the samples, there were a few dead bugs present and hence filtration was necessary. This solution was filtered through a Whatman filter paper (#42) in a glass vacuum filtration flask assembly and was rinsed twice with 6M HCl. The filter paper containing the residue was routinely gamma counted and no measurable ^7Be or ^{210}Pb activity was found. The flask was rinsed twice

with 6M HCl and the rinse solution was combined with the solution in the beaker. The solution was further reduced to 50-ml by evaporation for ^{210}Po plating (details and data are not given). The plated solution was transferred into a 100-ml volumetric flask and the volume was made to 100 ml with 2N HCl. A 5-ml aliquot was taken for the atomic absorption spectrometry (Varian AA-1275 AA Spectrophotometer) to determine the chemical yield of Be and Pb. The remaining sample was transferred into a Teflon beaker and the solution was dried completely. The residue was quantitatively transferred into a 10-ml counting vial. A few drops of 2 N HCl were added to the beaker and the rinse was added to the vial. The final volume of the solution in the vial varied between 3 and 5ml. The chemical yield varied between 70 and 100%, for ^7Be and ^{210}Pb . The sample was counted by gamma-ray spectrometry (details below).

[9] The aerosol-laden filter paper was weighed, cut into two halves and one piece was used for chemical analysis and the other half was archived. The cut filter paper was weighed and digested by adding 20-ml of HF acid. The paper was brought into solution by repeated digestion with 10-ml HNO_3 followed by 10-ml conc. HCl. Finally, the solution was taken in 6M-HCl and transferred into a 10-ml gamma counting vial. Most of the time, there was no residue when the solution was taken in 6M HCl. In a few samples, there was some residue and the solution was centrifuged and the supernatant was taken in the counting vial. The residue as well as the solution was assayed for gamma-emitting radionuclides. In the residue, no detectable ^7Be or ^{210}Pb was found.

2.3. Counting Procedures for ^7Be , and ^{210}Pb in Bulk, and Dry Depositional Samples and Aerosols

[10] The radiochemically-processed samples were counted for approximately 24 hours in 10-ml gamma counting vials in a high-purity Ge-Well detector (Canberra) coupled to a Canberra InSpector multi-channel analyzer. The active volume of the crystal was 179 cm^3 . There was no peak background in the any of the regions of interest (46.5 keV and 477.6 keV). The Peak/Compton ratio at 1.332 MeV was 56.6:1. The Full-Width at Half Maxima at 0.122 MeV and 1.332 MeV were 1.26 and 1.97 keV, respectively. The matrix densities of the standard and sample were quite comparable, and hence the self-absorption corrections were not applied. The gamma ray detector was calibrated with solid standards (RGU-1) obtained from the International Atomic Energy Agency (IAEA) for ^{210}Pb and other daughter products of U-series isotopes (such as ^{234}Th , ^{226}Ra). For ^7Be (477.6 keV), the counting efficiency was obtained from the linear extrapolation of the counting efficiencies obtained for ^{214}Pb at 351.6 keV and ^{214}Bi at 609.4 keV. This value was compared to the value obtained from the efficiency ratio of ^{214}Pb (351.6 keV)/ ^7Be obtained about 2 years ago using the National Institute of Standards and Technology (NIST)-traceable standards of ^7Be and the agreement was found to be very good. The ratios of the disintegrations per minute (dpm) to counts per minute (cpm) and counting efficiencies for various geometries were obtained. This conversion factor (dpm/cpm) and counting efficiency were then used to calculate all ^7Be and ^{210}Pb activities. The errors associated with the dpm/cpm conversion factors were

always less than 1%. The propagated errors (errors arising from the calibration of the detector and errors associated with the sample counting) on the activities (both aerosols and bulk and dry depositional fluxes) were less than 5%. The final ^7Be concentrations and deposition fluxes were corrected for radioactive decay from the end of sample collection to mid-counting time as well as for the decay during the deployment period as follows:

$$A_{\text{Be}} = [N_7 D / (\eta T)]_x e^{\lambda t_1} * \lambda t_2 / [1 - e^{-\lambda t_2}] \quad (1)$$

where A_{Be} is the activity of ^7Be (dpm), N_7 is the background subtracted net ^7Be (477.6 keV) counts, D is the (dpm/cpm) factor for the geometry of the sample, T is the counting time (in minutes), η is the chemical efficiency, λ is the decay constant of ^7Be [$= \ln 2 / T_{1/2}$ of ^7Be , 0.0130 day^{-1}], t_1 is the time between end of collection and mid-counting in days, and t_2 is the time between deployment in days.

[11] In the case of ^7Be , the dpm/cpm factor was calculated for each geometry using the counting efficiency for each geometry and the branching ratio [(dpm/cpm) = $1 / (\text{counting system efficiency} \times \text{branching ratio})$].

[12] The total depositional fluxes (F) for ^7Be and ^{210}Pb were calculated as follows:

$$F_{\text{Be/Pb}} = A_{\text{Be/Pb}} / (T \times S) \quad (2)$$

where $A_{\text{Be/Pb}}$ is the total activity (in dpm) deposited in the rain collector, T is the duration of deployment of the rain collector (in year) and S is the total surface area of the collector (in cm^2).

3. Results and Discussion

3.1. Monthly and Seasonal Variations of Rainfall and Bulk Depositional Fluxes of ^7Be and ^{210}Pb

[13] The amount of precipitation, collection interval, specific concentrations, and depositional fluxes of ^7Be and ^{210}Pb for the 27 bulk and 14 dry deposition, and 30 aerosol samples collected between September 1999 and February 2001 are given in Tables 1, 2, and 3. The deployment periods were not strictly from the beginning to end of the months. In order to discuss the monthly and seasonal variations of the amount of rainfall and depositional fluxes of ^7Be and ^{210}Pb , the monthly depositional fluxes were calculated as follows:

[14] Calculated Monthly flux = [Total activity (dpm) during the deployment period + activity collected in the previous collection period that overlaps with the month of interest (days included in the previous sample) - activity collected after the retrieval that overlaps with the next month (days included in the next month sample)]/rain collector area ($2,800\text{ cm}^2$). The activity was prorated based on the amount of rainfall during these periods under the assumption that the activity during a particular collection period is proportional to the amount of rainfall. Although there could be considerable variations within a month, the errors introduced by this assumption are assumed to be small (less than 5%), since the fractional correction is rather small in most of the samples. The monthly precipitation and

Table 1. Sampling Time Intervals, Total Depositional Fluxes, and Concentrations of ^7Be and ^{210}Pb Measured in Detroit, Michigan, From September 1999 Through February 2001

Collection Interval	Sample Code	Days in Collection	Number of Rainy Days	Rainfall, cm	^7Be , dpm L ⁻¹	^{210}Pb , dpm L ⁻¹	$^7\text{Be}/^{210}\text{Pb}$ Activity Ratio	^7Be Flux, dpm cm ⁻² yr ⁻¹	^{210}Pb Flux, dpm cm ⁻² yr ⁻¹
Sept. 24–Oct. 1, 1999	RW-B1	7	3	4.4	89.8	6.31	14.2	20.5	1.44
Oct. 1–Oct. 5, 1999	RW-B2	4	2	1.2	135	24.1	5.60	15.3	2.73
Oct. 5–Oct. 14, 1999	RW-B3	9	3	2.3	163	30.0	5.62	15.0	2.67
Oct. 14–Nov. 4, 1999	RW-B4	21	2	1.8	109	11.7	8.94	3.13	0.35
Nov. 4–Dec. 9, 1999	RW-B5	35	10	3.6	235	27.9	8.50	8.76	1.03
Dec. 9–Jan. 7, 2000 ^a	RW-B6	29	7	4.4	153	22.5	6.80	8.50	1.25
Jan. 7–Jan. 27, 2000 ^a	RW-B7	20	7	1.6	195	41.1	4.91	5.70	1.16
Jan. 27–Feb. 25, 2000 ^a	RW-B8	29	6	1.9	239	41.1	6.39	5.62	0.88
Feb. 25–Mar. 27, 2000 ^a	RW-B9	31	10	3.3	300	25.1	12.0	11.6	0.97
Mar. 27–April 22, 2000	RW-B10	26	11	7.5	177	11.9	15.0	16.5	1.10
April 22–May 12, 2000	RW-B11	20	5	5.4	131	12.8	10.2	12.9	1.27
May 12–May 22, 2000	RW-B12	10	6	4.6	270	17.3	15.6	38.9	2.50
May 22–May 29, 2000	RW-B13	7	2	1.4	453	34.4	13.2	26.0	1.97
May 29–June 1, 2000	RW-B14	3	1	0.56	656	68.7	9.56	21.9	2.29
June 1–June 22, 2000	RW-B15	21	8	2.7	249	20.6	12.1	8.69	0.72
June 22–June 30, 2000	RW-B16	8	5	5.1	160	14.8	10.8	23.7	2.19
June 30–July 29, 2000	RW-B17	29	9	3.8	458	46.1	9.95	20.0	2.01
July 29–Aug. 5, 2000	RW-B18	7	5	4.8	223	34.2	6.51	39.3	6.04
Aug. 5–Aug. 21, 2000	RW-B19	16	5	1.6	268	29.5	9.09	9.54	1.05
Aug. 21–Aug. 23, 2000	RW-B20	2	1	2.6	198	32.4	6.11	63.0	10.31
Aug. 23–Sept. 17, 2000	RW-B21	25	8	9.5	90.3	12.5	7.23	12.5	1.73
Sept. 17–Oct. 9, 2000	RW-B22	22	10	5.6	116	17.4	6.67	10.8	1.62
Oct. 9–Oct. 26, 2000	RW-B23	17	7	2.6	189	44.0	4.29	6.44	1.50
Oct. 26–Nov. 26, 2000	RW-B24	31	11	3.5	76.4	24.6	3.11	3.11	1.00
Nov. 26–Jan. 1, 2001 ^a	RW-B25	36	14	5.1	75.4	21.1	3.57	3.86	1.08
Jan. 1–Feb. 1, 2001 ^a	RW-B26	31	8	1.2	410	74.0	4.78	4.45	0.93
Feb. 1–Mar. 1, 2001 ^a	RW-B27	29	3	3.4	159	18.7	8.49	6.79	0.80

^aPrecipitation in the form of snowflakes or flurries.

the depositional fluxes of ^7Be and ^{210}Pb are plotted in Figures 1 and 2.

[15] The temporal variations of the monthly depositional fluxes of ^7Be and ^{210}Pb and the amount of precipitation indicate that generally the amount of precipitation seems to be controlling the magnitude of the depositional fluxes of ^7Be and ^{210}Pb (Figures 1 and 2). The deposition of ^7Be and ^{210}Pb to the Earth's surface varies with season, latitude (for ^7Be), longitude (for ^{210}Pb), and local meteorological con-

ditions [e.g., *Preiss et al.*, 1996]. The monthly depositional flux of ^7Be between 0.31 and 2.67 dpm cm⁻² (Figure 1), with a mean value (over the 17 months; 'mean' always represent arithmetic mean unless otherwise specified) of 0.97 dpm cm⁻²; this value is comparable to that given by *Todd et al.* [1989], who reported an average monthly ^7Be flux of 1.01 dpm cm⁻². There appears to be seasonality in the ^7Be flux with about 35.2% of the total annual deposition (December 1999 to November 2000) occurring during the

Table 2. Dry Depositional Fluxes of ^7Be and ^{210}Pb Measured in Detroit, Michigan^a

Sample Code	Collection Interval	Deployment Time, days	^7Be Flux, dpm cm ⁻² yr ⁻¹	^{210}Pb Flux, dpm cm ⁻² yr ⁻¹	^7Be Flux, dpm cm ⁻² yr ⁻¹ Percentage	^{210}Pb Flux, dpm cm ⁻² yr ⁻¹ Percentage	$^7\text{Be}/^{210}\text{Pb}$ Activity Ratio
RW-D1	Oct. 14–Oct. 21, 1999	4	0.62 (3.13)	0.39 (0.35)	19.8	>100 ^b	1.59
RW-D2	Oct. 24–Oct. 31, 1999	7	0.36 (3.13)	0.17 (0.35)	11.5	48.6	2.12
RW-D3 ^c	Oct. 31–Nov. 13, 1999	13	6.08	1.53	–	–	3.97
RW-D4 ^c	Nov. 13–Nov. 28, 1999	15	4.56	0.47	–	–	9.70
RW-D6	Jan. 26–Feb. 25, 2000	6	0.96 (5.62)	0.40 (0.88)	17.1	45.5	2.40
RW-D7 ^c	Feb. 25–April 22, 2000	6	11.8	1.12	–	–	10.5
RW-D8	April 22–May 24, 2000	10	0.80 (23.3)	0.14 (1.79)	3.4	7.8	5.71
RW-D10	June 8–June 30, 2000	5	1.37 (15.0)	0.32 (1.32)	9.1	24.2	4.28
RW-D11	June 30–July 19, 2000	4	1.68 (20.0)	0.28 (2.01)	8.4	13.9	6.00
RW-D12	July 19–Aug. 23, 2000	10	0.67 (31.3)	0.22 (6.04)	2.1	3.6	3.05
RW-D13	Aug. 23–Sept. 22, 2000	6	NM ^d	0.33	–	–	–
RW-D15	Oct. 22–Nov. 3, 2000	6	0.61 (4.93)	0.12 (1.33)	12.4	9.0	5.08
RW-D16	Nov. 3–Nov. 26, 2000	4	0.66 (4.93)	0.39 (1.33)	13.4	29.3	1.69
RW-D17 ^c	Nov. 26–Jan. 13, 2001	7	4.52	1.73	–	–	2.61

^aNumbers in parentheses denote bulk depositional fluxes during the deployment period of the dry collector (collected from Table 1).

^bDue to limited time exposure of the dry collector, the fractional dry depositional flux exceeds the bulk depositional flux.

^cLikely exposure to precipitation.

^dNM, not measured in time.

Table 3. Sampling Time Intervals and Specific Concentrations in Aerosols of ^7Be and ^{210}Pb Measured in Detroit, Michigan

Collection Interval	Deployment Time, days	Volume of Air Filtered, m^3	Particulate Concentration, $\mu\text{g m}^{-3}$	^7Be , dpm m^{-3}	^{210}Pb , dpm m^{-3}	$^7\text{Be}/^{210}\text{Pb}$ Activity Ratio
Oct. 28–Oct. 29, 1999	1	1873	183	0.588	0.253	2.32
Oct. 31–Nov. 1, 1999	1	2236	92	0.292	0.147	1.99
Jan. 10–Jan. 11, 2000	1	2377	45	0.170	0.077	2.21
Jan. 22–Jan. 24, 2000	2	3932	56	0.216	0.126	1.71
Jan 26–Jan. 27, 2000	1	2987	129	0.274	0.063	4.35
Feb. 3–Feb. 6, 2000	3	6601	61	0.264	0.080	3.30
Feb. 7–Feb. 9, 2000	2	4941	94	0.336	0.080	4.20
Mar. 8–Mar. 11, 2000	2	5004	123	0.281	0.027	10.4
Mar. 21–Mar. 23, 2000	2	3326	147	0.301	0.035	8.60
April 14–April 16, 2000	2	4899	94	0.284	0.043	6.60
April 30–May 2, 2000	2	4279	73	0.341	0.035	9.74
May 2–May 4, 2000	2	3615	57	0.293	0.029	10.1
May 12–May 14, 2000	2	3472	77	0.220	0.031	7.10
May 14–May 16, 2000	2	4506	102	0.433	0.033	13.1
May 22–May 24, 2000	2	3703	96	0.396	0.055	7.20
May 24–May 26, 2000	2	3387	111	0.340	0.018	18.9
June 30–July 2, 2000	2	4239	97	0.392	0.049	8.00
July 8–July 9, 2000	1	2086	84	0.266	0.023	11.6
July 13–July 14, 2000	1	1525	127	0.464	0.052	8.92
July 24–July 26, 2000	2	3387	80	0.241	0.043	5.60
Aug. 12–Aug. 14, 2000	2	4459	77	0.448	0.045	9.96
Aug. 24–Aug. 26, 2000	2	4426	106	0.330	0.056	5.89
Aug. 29–Sept. 1, 2000	2	5548	75	0.265	0.055	4.82
Sept. 20–Sept. 22, 2000	2	4382	68	0.093	0.038	2.45
Sept. 29–Oct. 1, 2000	2	3670	82	0.218	0.084	2.60
Oct. 11–Oct. 13, 2000	2	4253	118	0.247	0.120	2.06
Oct. 26–Oct. 29, 2000	3	6636	74	0.233	0.071	3.28
Nov. 29–Dec. 1, 2000	2	4331	54	0.105	0.077	1.36
Jan. 21–Jan. 23, 2001	2	4155	81	0.263	0.146	1.80
Feb. 2–Feb. 4, 2001	2	5055	67	0.173	0.082	2.11

spring (March–May, 2000) when 29.6% of the total annual precipitation occurred. This seasonal trend is probably related to the mid-latitude folding of the tropopause and the increasing frequency of precipitation that generally occur during this season.

[16] The ^{210}Pb depositional monthly flux varied between 0.03 and 0.27 dpm cm^{-2} , with a mean value of 0.12 dpm cm^{-2} and was higher in the summer months of 2000 (Figure 2, August 2000 had the highest flux during this study period); however the months of May (spring) and October

(fall) 2000 had the second and third highest fluxes throughout the study (Figure 2).

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

[17] Although seasonal variations in the depositional fluxes of ^7Be and sometimes ^{210}Pb have been reported, the importance of the amount of precipitation on the depositional fluxes need to be elucidated before one can attribute the increase/decrease to seasonal variations. In order to quantify the importance of the amount of precipitation on the depositional fluxes of these nuclides, we calculated the fractional amounts of precipitation and depositional fluxes of ^7Be and ^{210}Pb for the twelve-month

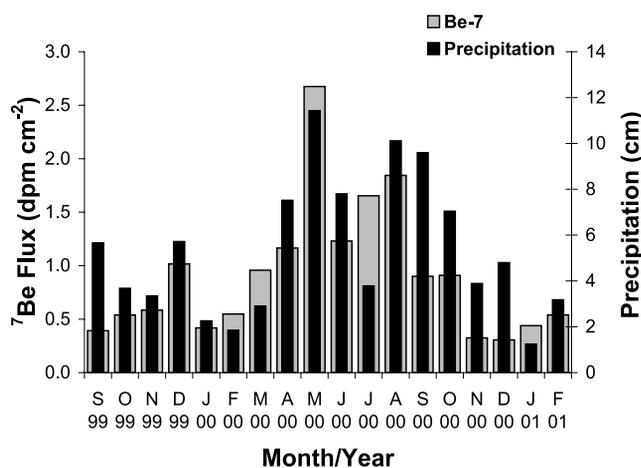
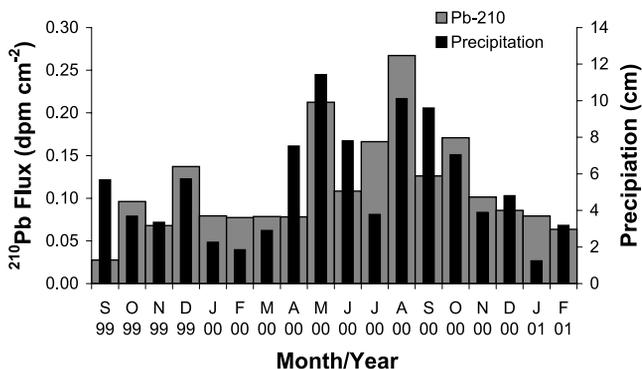
**Figure 1.** Monthly ^7Be depositional flux and precipitation amount with time in Detroit, Michigan.**Figure 2.** Monthly ^{210}Pb depositional flux and precipitation amount with time in Detroit, Michigan.

Table 4. Amount of Precipitation, Seasonal Depositional Fluxes of ^7Be and ^{210}Pb , Normalized Enrichment Factors (α) for ^7Be and ^{210}Pb , and $^7\text{Be}/^{210}\text{Pb}$ Activity Ratios for 1 year (December 1999 to November 2000)^a

Parameter	Winter (December, January, February)	Spring (March, April, May)	Summer (June, July, August)	Fall (September, October, November)
Precipitation, cm	9.8 (13.3)	21.9 (29.6)	21.7 (29.4)	20.5 (27.7)
^7Be Fallout, dpm cm ⁻²	1.98 (14.5)	4.80 (35.2)	4.73 (34.7)	2.13 (15.6)
^7Be (α)	1.09	1.19	1.18	0.56
^{210}Pb Fallout, dpm cm ⁻²	0.293 (18.3)	0.369 (23.0)	0.541 (33.8)	0.398 (24.9)
^{210}Pb (α)	1.38	0.78	1.15	0.90
$^7\text{Be}/^{210}\text{Pb}$ Ratio	6.76	13.0	8.74	5.35

^aNumbers in parenthesis denote percentage of the yearly total amount of precipitation or bulk depositional flux.

period (December 1999–November 2000) for which we have data for all four seasons. The fractional amount of precipitation and depositional fluxes of ^7Be and ^{210}Pb for the four seasons during 2000 are given in Table 4. The precipitation-normalized enrichment factor (α) is defined as [Baskaran, 1995]

$$\alpha = (F_s/F_t)/(R_s/R_t) \quad (3)$$

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 of ^7Be and ^{210}Pb in that particular season and year, respectively. Alpha 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. The error in the values of α should be less than 10% (errors in the depositional fluxes is <3%), since it is the ratio of the seasonal depositional fluxes to the annual depositional fluxes. Assuming an upper limit of 10%, values below 0.9 and above 1.1 are considered to be significant.

[18] Table 4 clearly shows the seasonal increase of ^7Be during spring and summer seasons while there is a significant depletion during fall. There is a depletion of ^{210}Pb during spring and enrichment during winter and summer months. During spring due to troposphere-stratosphere exchange of air masses in mid-latitudes, a certain amount of air is exchanged between the troposphere and stratosphere. Since the production of ^7Be in the stratosphere is significantly higher than that in the troposphere [Lal and Peters, 1967], this exchange will result in a significant increase in the air concentration and increased inventory of ^7Be in the troposphere. In the case of ^7Be , there are two factors that control the depositional fluxes of ^7Be at any given site. They are as follows: (1) The amount of precipitation and, (2) the amount of ^7Be derived from the stratosphere (including the ^7Be derived from air masses above the cloud cover) due to vertical mixing. During summer months, the atmosphere is quite dynamic and hence large scale atmospheric mixing takes place. During vertical mixing, the ^{210}Pb is mixed upward to the middle and upper troposphere where precipitation scavenging is much less, leading to a decrease in the depositional flux of ^{210}Pb . However, increased fractional depositional fluxes of ^{210}Pb during spring have been reported and were attributed to the increases in the amounts and frequency of precipitation, the thawing of frozen soils, or the drying out of saturated soils [Olsen et al., 1985; Schuler et al., 1991]. The fractional depositional flux of ^7Be is distinctly lower than the fractional amount of precipitation during fall and both are

comparable to each other for winter. In most of the previous study areas, the fractional depositional fluxes of ^7Be and ^{210}Pb are higher than the fractional amount of precipitation during summer months, and in some cases in the spring [Baskaran, 1995] and always the opposite is true for fall. The fractional depositional fluxes of both ^7Be and ^{210}Pb during the fall are the lowest based on the analysis of the published literature on the depositional fluxes of ^7Be and ^{210}Pb [Baskaran, 1995]. In addition, the fractional depositional fluxes of ^7Be and ^{210}Pb during winter is higher than the fractional amount of precipitation, possibly suggesting that the snow flurries are more efficient in removing ^{210}Pb and ^7Be from the air than the rain. Since the surface area of snow flurries are significantly higher (flurry average density $\sim 0.1 \text{ g cm}^{-3}$, and hence surface area will be ~ 4.6 times higher, assuming that the flurries are spherical in shape) than those of raindrops for a given mass, it is likely that the snow flurries are more efficient scavengers of particle-reactive nuclides and other pollutants. In addition, due to low density, the flurries can easily flow around due to the wind, and hence the volume of air swept by snow flurries will be significantly higher than those due to water droplets. Our observation is in contrast with the suggestion by Kim et al. [2000], who suggested that ^7Be relative to ^{210}Pb are less efficiently scavenged by the snowfall.

3.3. Relationship Between Specific Concentrations of ^7Be and ^{210}Pb and Amount of Precipitation

[19] A number of factors including cloud height, the amount and duration of rainfall, the time elapsed between successive rain events and the vertical mixing of air masses at the sampling site (which could add additional ^7Be to the air) have been attributed to the variations on the specific concentrations of ^7Be and ^{210}Pb in the bulk deposition samples. The range of values found in this study on the specific concentrations of ^7Be (75.4–656 dpm L⁻¹ with a geometric mean of 190 dpm L⁻¹) and ^{210}Pb (6.31–74.0 dpm L⁻¹ with a geometric mean of 24.4 dpm L⁻¹) is comparable to the ranges reported in other places [Todd et al., 1989; Papastefanou and Ioannidou, 1991; Baskaran et al., 1993; Benitez-Nelson and Buesseler, 1999; Kim et al., 1998].

[20] Generally, samples with low precipitation are likely to have higher specific concentrations of ^7Be and ^{210}Pb because very small raindrops that occur during drizzles have a much larger surface area that will result in higher removal of ^7Be and ^{210}Pb on each rain droplet. The ^7Be and ^{210}Pb nuclides on the smaller size rain droplets have greater affinity to aerosols. The small rain droplets will coalesce with one another and grow in size and keep the nuclide

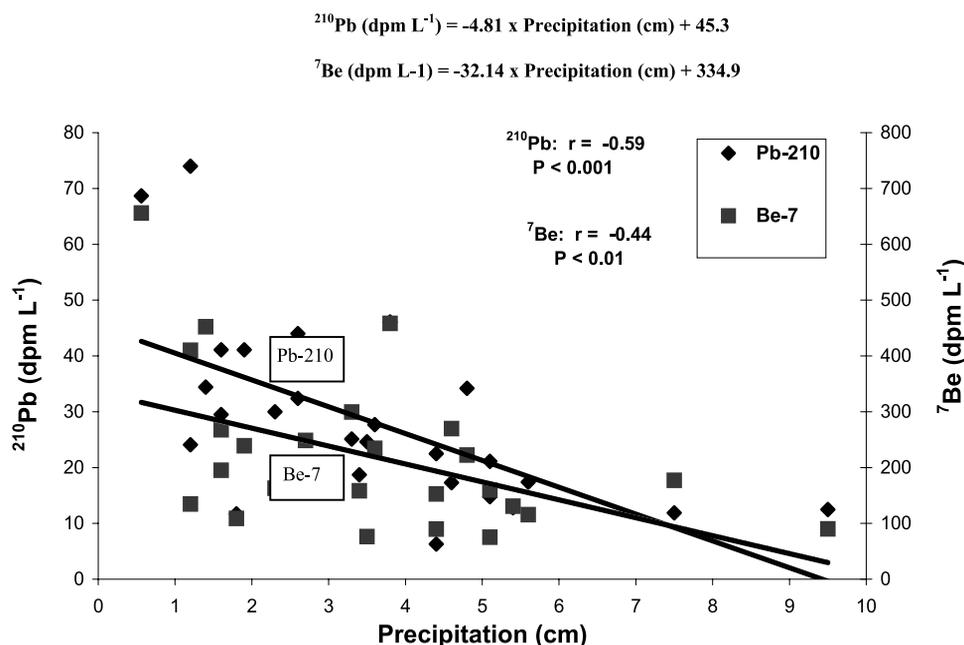


Figure 3. The specific concentrations of ^7Be and ^{210}Pb (dpm L^{-1}) in precipitation versus amount of precipitation in Detroit, Michigan, from September 1999 through February 2001.

concentration constant while continued water condensation will dilute the concentrations of these nuclides. Since smaller amounts of precipitation are associated with very high evaporation rates (surface area effect) due to lower humidity below the cloud cover, these rain droplets are likely enriched in ^{210}Pb and ^7Be [Wogman *et al.*, 1968].

[21] The specific concentrations of ^7Be and ^{210}Pb are plotted against the amount of rainfall and the mathematical relationship between the amount of rain and the specific concentrations of these nuclides are given in Figure 3. Although there is considerable scatter in the data, there is a significant correlation between the specific concentration of ^7Be ($r = -0.44$, $P < 0.01$) and ^{210}Pb ($r = -0.59$, $P < 0.001$) and the amount of rainfall. Similar observations have been reported from other places for ^7Be [Turekian *et al.*, 1983; Olsen *et al.*, 1985; Kim *et al.*, 1998, 2000] and ^{210}Pb [Olsen *et al.*, 1985; Nevissi, 1985]. However, in areas where large portions of precipitation are derived from heavy rain events (such as several places along the Gulf coast), no correlation was found [Brown *et al.*, 1989; Baskaran *et al.*, 1993; Benitez-Nelson and Buesseler, 1999; Kim *et al.*, 1998, 2000]. Significant correlation between the specific concentrations of ^7Be and amount of precipitation could be a measure of the variability in the tropospheric inventory and efficient removal of ^7Be by precipitation below the cloud cover. Lack of correlation could be due to insufficient time for the build-up of ^7Be below the cloud-cover due to slower replenishment of ^7Be from air masses above the cloud-cover. In addition, the specific concentrations of ^7Be and ^{210}Pb are expected to be higher during drier months, as the build-up of these nuclides in the air due to lower removal by precipitation will result in higher specific concentrations of ^7Be and ^{210}Pb . Although no correlation was found between specific radionuclide activities and rainfall, significant correlations were found between radionuclide fluxes and rainfall, indicating importance of rainfall

in the removal of these nuclides from the troposphere [Benitez-Nelson and Buesseler, 1999; Dueñas *et al.*, 2001].

3.4. Volume-Weighted Concentrations of ^7Be and ^{210}Pb

[22] The volume-weighted concentrations of ^7Be and ^{210}Pb are calculated by the total annual ^7Be (or ^{210}Pb) deposited in the bulk collector divided by the annual precipitation amount in the collector. During this study period, volume-weighted concentration for ^7Be was found to be 172 dpm L^{-1} and is significantly higher than the values reported for New Haven (158 dpm L^{-1} [Turekian *et al.*, 1983]), Bermuda (94 dpm L^{-1} [Turekian *et al.*, 1983]), Virginia (90 and 100 dpm L^{-1} for the years 1983 and 1984 [Todd *et al.*, 1989]), Tennessee (93 and 99 dpm L^{-1} for the years 1983 and 1984, respectively [Olsen *et al.*, 1985]), Texas (87 , 124 and 155 dpm L^{-1} for the years 1989, 1990 and 1991, respectively for Galveston [Baskaran *et al.*, 1993]; 124 and 104 dpm L^{-1} for the years 1990 and 1991, respectively for College Station), and Switzerland (138 dpm L^{-1} [Dominik *et al.*, 1987]; 146 dpm L^{-1} [Schuler *et al.*, 1991]) and Ansan, South Korea (90 dpm L^{-1} [Kim *et al.*, 1998]). The volume-weighted concentration of ^7Be for various sites around the world (data summarized by McNeary [2002]) was plotted against the annual rainfall and there was no significant correlation between the two (correlation coefficient = 0.13 , $n = 15$).

[23] The volume-weighted concentration for ^{210}Pb was calculated to be 21.2 dpm L^{-1} and is the highest reported so far. This high value could be due to a number of factors including the relative proportion of the air masses (oceanic versus continental), distance from the coastline, amount and frequency of rainfall, and altitude of the clouds. There was no significant correlation ($r = 0.35$; $n = 15$) between the volume-weighted concentration of ^{210}Pb and the amount of annual precipitation for the data compiled for various sites around the world [McNeary, 2002]. It is likely that the

Table 5. Percentage of Bulk Depositional Fluxes of ^7Be and ^{210}Pb During Heavy Rain at Detroit, Michigan, During 1999 and 2000^a

Sample	Individual Rain Event	Rainfall, cm	Percent of Annual Rainfall ^b	Percentage of Annual ^7Be Flux	Percentage of Annual ^{210}Pb Flux
RWB 1	Sept. 28–29, 1999	4.22 (96.6)	5.78	15.8 ^c	8.6 ^c
RWB 10	April 20, 2000	5.49 (73)	7.52	9.6	5.4
RWB 16	June 24–25, 2000	4.60 (90.6)	6.30	5.0	3.9
RWB 20	Aug. 22–23, 2000	2.59 (100)	3.55	3.3	4.4
RWB 21	Sept. 10–12, 2000	6.86 (72.2)	9.40	6.6	7.6
RWB 25	Dec. 15–16, 2000	3.63 (71.9)	4.97	2.9	6.9

^aNumbers in parenthesis denote the percentage of event rainfall during the deployment period.

^bEvents with >2.5 cm of precipitation and where the rainfall event was greater than 70% of total rainfall during the deployment period were chosen.

^cThe percentage of annual depositional fluxes of ^7Be and ^{210}Pb were calculated using the total fallout during the 4-month period the rain collector was deployed.

frequency of precipitation plays a significant role on the volume-weighted concentration of ^{210}Pb , rather than the amount of precipitation. In order to assess the importance of the frequency of precipitation, we compared the percentage of rainy days during the deployment period at the study site to those in other places. The only published data set that is available in literature on the number of rainy days during the deployment period is for Galveston, Texas [Baskaran *et al.*, 1993]. The percentage of rainy days in Detroit (32%) is higher than those for Galveston, Texas (27%), although the total annual rainfall is significantly lower (43%, average over three years, 1989–1991) in Detroit than Galveston, Texas. The coastal site (Galveston) is likely to have a lower ^{210}Pb value, due to significant input of oceanic air mass (expected to be depleted in ^{210}Pb due to low exhalation of ^{222}Rn from the water). Detroit is farther away from the nearest coastal site and hence the relative proportion of the marine air is likely to be small. However, even a small portion of the marine air could pick-up significant amounts of ^{210}Pb enroute to Southeast Michigan and hence the concentration of ^{210}Pb in air is likely higher.

3.5. Variations of Specific Concentrations and Fluxes of ^7Be and ^{210}Pb in Individual Rain Events and Fraction of the Annual Depositional Fluxes of These Nuclides From Pulse Rain Events

[24] In certain regions of the Earth, there are lots of thunderstorms, hurricanes and tornadoes that bring large amounts of rainfall leading to higher deposition of ^7Be and ^{210}Pb . Pulse injection of these nuclides in the coastal waters is useful to investigate the particle dynamics and hence information on the pulse input of these nuclides due to heavy rainfall is warranted. The pulse injection could result from one single injection lasting less than 12 hours or could last as long as 2–3 days with continuous rainfall. In the Gulf Coast, ~5% of the rainfall events bring about 20–25% of the bulk depositional fluxes of ^7Be and ^{210}Pb [Baskaran *et al.*, 1993]. Beks *et al.* [1998] reported that the variations in the annual depositional flux at two sites in Netherlands (Textel and Groningen) appear to be correlated to the number of heavy rains or thunderstorms. Dibb [1989] proposed that the thunderstorm rains maybe more effective than snow in stripping ^7Be from the atmosphere. In general, in southeastern Michigan, thunderstorms (>5 cm rainfall in a day) are less common. The percentages amounts of rainfall and depositional fluxes of ^7Be and ^{210}Pb during heavy rain events are given in Table 5. In 2000, 5 heavy

rainfalls with more than 2.5 cm of precipitation deposited 27.4 and 28.1% of the total annual fluxes of ^7Be and ^{210}Pb , respectively (Table 5). It appears that 5 heavy rain events per year (~4% of the total 134 rainy days during year 2000) account for about 25 to 30% of the total annual deposition rate of ^7Be and ^{210}Pb . This is likely to be true for other atmospherically delivered stable elements and chemicals that behave similar to Be and Pb.

3.6. Dry Depositional Flux of ^7Be and ^{210}Pb and Activity Ratio of $^7\text{Be}/^{210}\text{Pb}$

[25] The dry only depositional fluxes of ^7Be and ^{210}Pb were measured from October 14, 1999 to January 13, 2001. The collection interval, depositional fluxes and activity ratios of ^7Be and ^{210}Pb are listed in Table 2. Three dry deposition samples (RW-D5, RW-D9 and RW-D14) were lost during collection and in one sample (RW-D13), the sample was counted about 6 months after collection and hence most of the ^7Be activity reached below detection limit. In four samples (RW-D3, RW-D4, RW-D7 and RW-D17) very light showers contributed some amount of wet fallout into the dry collector and thus the data from these four samples will not be used for any further discussion and interpretation. The dry depositional flux of ^7Be varied between $0.36 \text{ dpm cm}^{-2} \text{ y}^{-1}$ (11.5% of the bulk) to $1.68 \text{ dpm cm}^{-2} \text{ y}^{-1}$ (8.4% of the bulk), with a mean of $0.86 \text{ dpm cm}^{-2} \text{ yr}^{-1}$. The fractional dry depositional flux of ^7Be varied between 2.1 and 19.8% (Table 2). The dry depositional flux of ^{210}Pb varied between $0.12 \text{ dpm cm}^{-2} \text{ y}^{-1}$ (9.0% of the bulk) and $0.40 \text{ dpm cm}^{-2} \text{ yr}^{-1}$ (13.4% of the bulk), with a mean of $0.28 \text{ dpm cm}^{-2} \text{ yr}^{-1}$. The fractional dry depositional flux of ^{210}Pb varied between 3.6 and 48.6% (Table 2). The average dry depositional flux of ^{210}Pb (22.7%, excluding one with >100% in Table 2) is significantly higher than ^7Be (10.8%) and this difference is attributed to the differences in sources of ^7Be and ^{210}Pb . The $^7\text{Be}/^{210}\text{Pb}$ activity ratio varied between 1.59 and 6.00, with a mean value of 3.55.

[26] Previous studies on the wet and bulk deposition of ^{210}Pb and ^7Be indicate that dry deposition is generally less than 10% of bulk deposition [Turekian *et al.*, 1977; Talbot and Andren, 1983; Olsen *et al.*, 1985; Todd *et al.*, 1989; Brown *et al.*, 1989]. Nevissi [1985] reported the dry fallout to be less than 14% of the total ^{210}Pb flux on an annual basis. Our results agree with earlier studies that the dry fallout is a relatively minor component of the bulk fallout. Our results support the previous studies that the fraction of

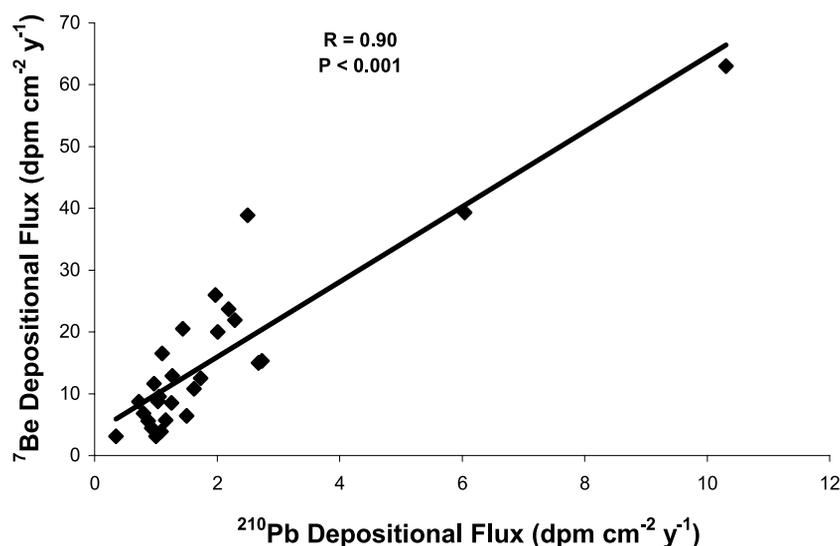


Figure 4. Bulk depositional fluxes of ^7Be versus ^{210}Pb in Detroit, Michigan, from September 1999 through February 2001.

the dry depositional flux of these nuclides is highly variable. For example, the dry depositional flux of ^7Be in Galveston, Texas, varied between 3.0 and 7.4% of the bulk depositional flux in October 1990, but it amounted to $\sim 87\%$ during October 1991 and this was attributed to very low rainfall [Baskaran *et al.*, 1993]. The dry fallout of ^{210}Pb (6 to 22% in October 1990 and 41% in October 1991) was found to be higher than those for ^7Be and this higher fallout for ^{210}Pb was attributed to either leaching of ^{210}Pb from the resuspended soil from the ground or importance of dry fallout for ^{210}Pb than ^7Be [Baskaran *et al.*, 1993]. Benitez-Nelson and Buesseler [1999] showed that the dry deposition accounts for less than 1% of the ^7Be flux, but accounts for 12% of the ^{210}Pb flux. There was no significant correlation between the dry fluxes (for both ^7Be and ^{210}Pb) and the amount of precipitation during that month on those 9 samples analyzed. The $^7\text{Be}/^{210}\text{Pb}$ activity ratios are significantly lower than those in the corresponding bulk precipitation samples and this is attributed to the difference in the source function of these nuclides. At the ground level, higher amounts of ^{210}Pb is produced from the decay of ^{222}Rn which can get attached to aerosols and get removed by dry depositional process, while most of the ^7Be is removed by precipitation.

3.7. Annual Bulk Depositional Fluxes of ^7Be and ^{210}Pb and Comparison of These Fluxes With Other Similar Data

[27] The annual bulk depositional flux of ^7Be at Detroit during the year 2000 was $13.0 \text{ dpm cm}^{-2} \text{ yr}^{-1}$ and this value can be compared to other sites in the mid-latitudes such as Woods Hole, MA ($12.7 \text{ dpm cm}^{-2} \text{ yr}^{-1}$ [Benitez-Nelson and Buesseler, 1999]), Portsmouth, MA ($16.6 \text{ dpm cm}^{-2} \text{ yr}^{-1}$ [Benitez-Nelson and Buesseler, 1999]), and Geneva, Switzerland ($12.5 \text{ dpm cm}^{-2} \text{ yr}^{-1}$ [Caillet *et al.*, 2001]). The annual bulk depositional flux of ^{210}Pb in the year 2000 was $1.56 \text{ dpm cm}^{-2} \text{ yr}^{-1}$. This annual ^{210}Pb deposition flux can be compared to other mid-latitude stations such as Seattle, Washington ($0.44 \text{ dpm cm}^{-2} \text{ yr}^{-1}$ [Nevissi, 1985]), Woods Hole, MA ($1.43 \text{ dpm cm}^{-2} \text{ yr}^{-1}$ [Benitez-Nelson and Buesseler, 1999]), Portsmouth, MA

($0.95 \text{ dpm cm}^{-2} \text{ yr}^{-1}$ [Benitez-Nelson and Buesseler, 1999]), Munich, Germany ($1.08 \text{ dpm cm}^{-2} \text{ yr}^{-1}$ [Winkler and Rosner, 2000]), and Geneva, Switzerland ($0.90 \text{ dpm cm}^{-2} \text{ yr}^{-1}$ [Caillet *et al.*, 2001]). The value reported in the present study can also be compared to the value obtained from the inventory of ^{210}Pb reported from a set of sediment cores collected from four lakes in Minnesota, $0.91 \text{ dpm cm}^{-2} \text{ yr}^{-1}$ by Urban *et al.* [1990]. It appears that the value of $1.56 \text{ dpm cm}^{-2} \text{ yr}^{-1}$ is considerably higher than other sites with similar amounts of rainfall. This higher value could partly be attributed to the increased frequency of precipitation. Baskaran *et al.* [1993] reported that the annual depositional flux varied with the amount of rainfall and the precipitation-normalized depositional flux of ^{210}Pb has remained constant. With one-year complete dataset, we are unable to determine inter-annual variations in the depositional fluxes of ^{210}Pb .

3.8. Relationship Between the Depositional Fluxes of ^7Be and ^{210}Pb in the Bulk Precipitation

[28] The removal mechanisms of ^7Be and ^{210}Pb are very similar, although their modes of production are different. If the depositional fluxes of these nuclides are related, then, these two tracers cannot be used as independent atmospheric tracers for air of upper and lower atmospheric origin. There is a strong correlation between the bulk depositional fluxes of ^7Be and ^{210}Pb (Figure 4, $r = 0.90$, $P < 0.001$). Similar correlations have been reported for many continental stations while in island stations, such as Bermuda, the correlation is weak (0.43 , $n = 8$; all the data were compiled by Baskaran *et al.* [1993]). However, more recent results published by Kim *et al.* [2000] on the depositional fluxes of ^7Be and ^{210}Pb in Bermuda indicate a strong correlation ($r = 0.92$, $n = 16$) between these nuclides and the differences between these two contrasting observations were attributed either to higher altitude sources of air masses during the sampling period of Turekian *et al.* [1983] or the errors associated with the extrapolation of a limited data set for annual flux calculation [Kim *et al.*, 2000]. Our finding confirms earlier observations that in most of the continental

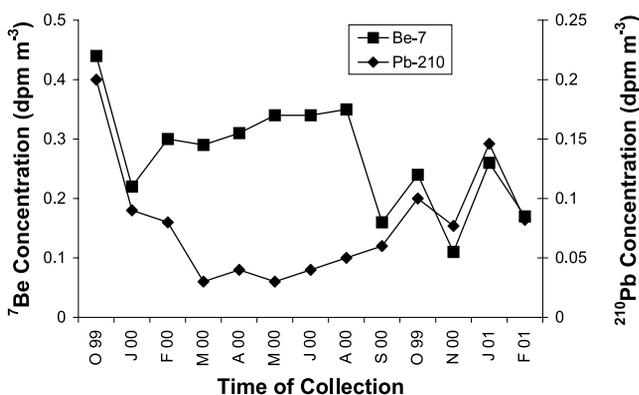


Figure 5. Temporal variations of ^7Be and ^{210}Pb concentration in aerosols with time in Detroit, Michigan.

and coastal stations, ^7Be and ^{210}Pb cannot be used as independent atmospheric tracers.

3.9. Concentrations of ^7Be and ^{210}Pb and Activity Ratios of $^7\text{Be}/^{210}\text{Pb}$ in Surface Air

[29] The volume of air passed through the glass fiber filter varied between 1,525 and 6,636 m^3 , with a mean of 3,975 m^3 (Table 3). The mass concentrations of the aerosol retained on the filter paper varied between 45 and 183 $\mu\text{g m}^{-3}$, with a mean value of 91 $\mu\text{g m}^{-3}$ (Table 3). In months when more than one air sample was collected, the average concentration value for ^7Be and ^{210}Pb for that month was calculated. The concentration of ^7Be in surface air ranged from 0.09 to 0.59 dpm m^{-3} , with a mean of 0.29 dpm m^{-3} (Table 3). In about 80% of the samples, the ^7Be concentration varied between 0.1 and 0.3 dpm m^{-3} . This range is comparable to other mid-latitude regions such as New Haven, Connecticut (0.27 dpm m^{-3} [Turekian *et al.*, 1983]), Argonne, Illinois (0.25 dpm m^{-3} [Feely *et al.*, 1989]), Sterling, Virginia (0.26 dpm m^{-3} [Feely *et al.*, 1989]), New York City, New York (0.27 dpm m^{-3} [Feely *et al.*, 1989]), and Thessaloniki, Greece (0.32 dpm m^{-3} in a 3-year period, January 1987 to April 1990 [Papastefanou and Ioannidou, 1991]). The temporal variations of ^7Be (monthly means) indicate that the concentra-

tions are generally higher during the spring and summer months (Figure 5).

[30] The concentration of ^{210}Pb in aerosol samples ranged between 0.018 and 0.253 dpm m^{-3} , with a mean value of 0.069 dpm m^{-3} (Table 3). This range is comparable to the values reported for other sites, such as New Haven, Connecticut (0.035 dpm m^{-3} [Turekian *et al.*, 1983]) and Munich-Neuherberg, South Germany (0.034 dpm m^{-3} [Winkler and Rosner, 2000]). The frequency distribution of ^{210}Pb concentration indicates the variations are more scattered, unlike ^7Be (figure not shown). The temporal variations of ^{210}Pb in the aerosol samples do not indicate higher concentrations during the spring and summer months (Figure 5) and highest ^{210}Pb concentration was found in October 1999 with the lowest concentration in May 2000. On the other hand, the concentration generally appears to be higher during the winter months. The lower concentration of ^{210}Pb in summer is likely due to dynamic vertical upward movement of ^{210}Pb -laden aerosols while higher concentration during winter months is likely due to stratification. The $^7\text{Be}/^{210}\text{Pb}$ activity ratio in surface air varied between 1.36 and 18.9, with a mean value of 6.07 (Table 3). Generally, the higher values were observed during late spring and summer months. In summer, convection mixes ^{210}Pb upward and ^7Be downward, leading to an annual peak in $^7\text{Be}/^{210}\text{Pb}$ ratio [Koch *et al.*, 1996]. Overall, most of the temporal variations in the atmospheric concentrations of ^7Be and ^{210}Pb are likely due to changes in the mixing height of surface level air. There is no significant correlation between the specific concentrations of ^7Be and ^{210}Pb in aerosols (Figure 6), unlike in the precipitation samples (Figure 4).

[31] The concentrations of ^7Be and ^{210}Pb in air samples at any season depend upon three factors. They include: (1) changes in the rate of transport of stratospheric air containing high concentrations of ^7Be and low concentrations of ^{210}Pb into the troposphere during the late winter and early spring seasons; (2) decreased stability of the troposphere during the warmer months leading to an increased rate of vertical transport of ^7Be (lower rate of vertical transport for ^{210}Pb) from the upper troposphere to the middle and lower troposphere and into the surface air [Feely *et al.*, 1989]; and

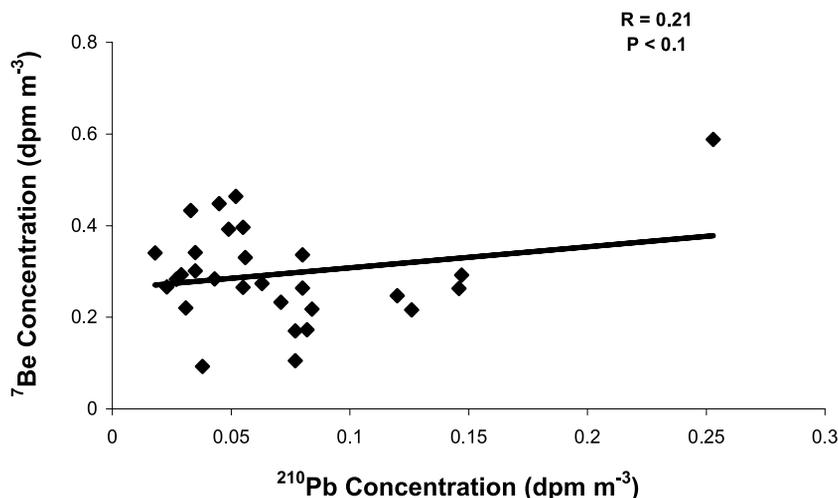


Figure 6. Correlation between specific concentrations of ^7Be and ^{210}Pb (Table 3) in aerosol samples in Detroit, Michigan, from October 1999 through February 2001.

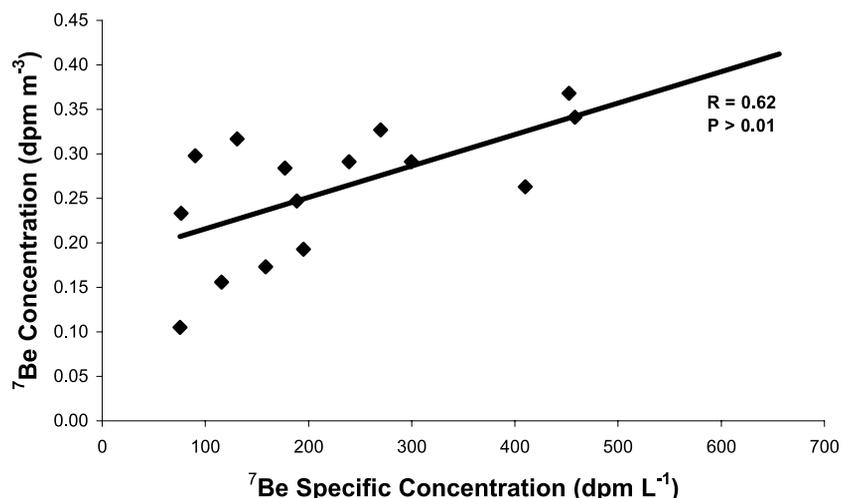


Figure 7. ^7Be concentration in surface air versus ^7Be specific concentration in precipitation from October 1999 through February 2001 in Detroit, Michigan.

(3) variations of the rate of washout of the atmospheric aerosols that carry the ^7Be and ^{210}Pb as seasonal changes in precipitation occur in the air masses reaching the sampling site. Although the ^{210}Pb was the lowest during early spring (Figure 5), there was no significant increase of ^7Be during that period (Figure 5). The co-variation between ^7Be and ^{210}Pb in the air is fairly tight (Figure 5) during October 1999 and January 2000 and from September 2000 to February 2001. From February to August 2000, if there was any increased rate of transport of air, due to factors 1 and 2 listed above, the ^{210}Pb concentration is expected to be lower, as the tropospheric air mixes with the stratospheric air containing low levels of ^{210}Pb while the opposite is expected to be true for ^7Be (higher concentration of ^7Be is expected, as the tropospheric air mixes with the stratospheric air containing higher concentrations of ^7Be). The amounts of precipitation were higher during the months of April–June, and August–October, 2000. During these months, there was no significant decrease in the concentration of ^7Be (Figure 5) and is likely due to additional input of ^7Be from the upper atmosphere. The concentrations of ^{210}Pb during these peri-

ods appear to be low (Figure 5). In order to see if the fluctuations in the concentrations of ^7Be and ^{210}Pb in aerosols are reflected in the bulk precipitation samples collected during the corresponding periods, the specific concentrations of these nuclides in the aerosols are plotted against those in the precipitation samples. There is a significant correlation between the specific concentration of ^7Be and ^{210}Pb in air to those in the bulk deposition samples ($r = 0.62$, $P > 0.01$ for ^7Be , Figure 7 and $r = 0.69$, $P > 0.01$ for ^{210}Pb , Figure 8), suggesting the concentrations of these nuclides in the air and precipitation are coupled, as expected.

3.10. Total Deposition Velocity of Aerosols Using ^7Be and ^{210}Pb

[32] The total deposition velocity (V_d) for any nuclide is determined by the following equation:

$$V_d = F/C_s \quad (4)$$

where F is the total flux of a nuclide to the Earth's surface and C_s is the concentration of that nuclide in air at the

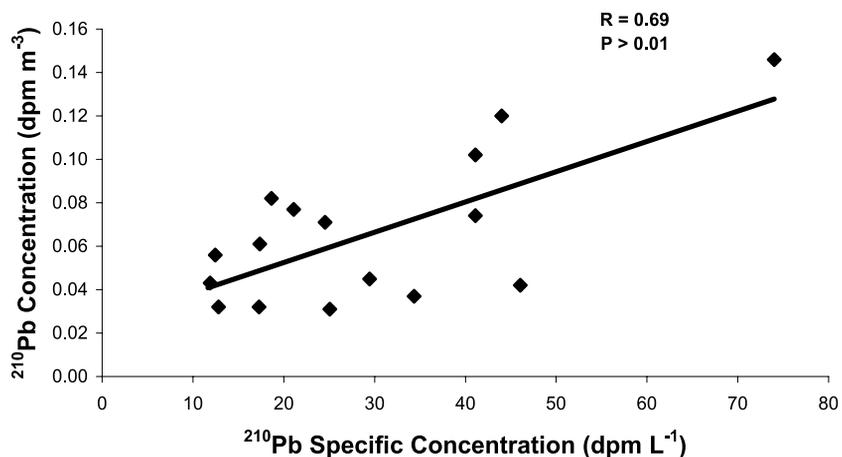


Figure 8. ^{210}Pb concentration in surface air versus ^{210}Pb specific concentration in precipitation from October 1999 through February 2001 in Detroit, Michigan.

Table 6. Total Deposition Velocity of Aerosols Using ^7Be and ^{210}Pb

Mid-Collection Time	^{210}Pb Flux, dpm $\text{m}^{-2} \text{d}^{-1}$	^{210}Pb Concentration, dpm m^{-3}	^7Be Flux, dpm $\text{m}^{-2} \text{d}^{-1}$	^7Be Concentration, dpm m^{-3}	^7Be Deposition Velocity, cm s^{-1}	^{210}Pb Deposition Velocity, cm s^{-1}
Oct. 29, 1999	9.7	0.253	85.6	0.589	0.17	0.04
Nov. 1, 1999	9.7	0.147	85.6	0.292	0.34	0.08
Jan. 11, 2000	31.9	0.077	15.6	0.170	1.06	0.48
Jan. 23, 2000	31.9	0.126	15.6	0.216	0.84	0.29
Jan. 27, 2000	31.9	0.063	15.6	0.274	0.66	0.59
Feb. 4, 2000	24	0.080	154	0.264	0.68	0.35
Feb. 8, 2000	24	0.080	154	0.336	0.53	0.35
March 9, 2000	26.5	0.027	31.7	0.283	1.29	1.15
March 22, 2000	26.5	0.035	31.7	0.301	1.22	0.88
April 15, 2000	32.2	0.043	480	0.284	2.0	0.86
May 1, 2000	34.7	0.035	354	0.341	1.20	1.16
May 3, 2000	34.7	0.029	354	0.292	1.40	1.37
May 13, 2000	73.5	0.031	1147	0.220	6.05	2.78
May 15, 2000	73.5	0.033	1147	0.433	3.07	2.59
May 23, 2000	61.4	0.055	809	0.396	2.36	1.29
May 25, 2000	61.4	0.019	809	0.340	2.76	3.64
July 1, 2000	57.3	0.049	570	0.392	1.68	1.36
July 9, 2000	57.3	0.023	570	0.266	2.48	2.92
July 14, 2000	57.3	0.052	570	0.464	1.42	1.26
July 25, 2000	57.3	0.043	570	0.241	2.73	1.53
Aug. 13, 2000	29.3	0.045	267	0.448	0.69	0.75
Aug. 25, 2000	47.4	0.253	343	0.330	1.20	0.97
Aug. 30, 2000	47.4	0.147	343	0.265	1.50	1.00
Sept. 20, 2000	44.5	0.077	296	0.093	3.70	1.35
Sept. 30, 2000	44.5	0.126	296	0.218	1.58	0.61
Oct. 12, 2000	52.8	0.063	226	0.246	1.06	0.51
Oct. 27, 2000	27.4	0.080	85.2	0.233	0.42	0.45
Nov. 30, 2000	29.6	0.080	106	0.105	1.16	0.44
Jan. 21, 2001	25.6	0.026	142	0.263	0.62	0.20
Feb. 3, 2001	21.9	0.082	186	0.173	1.25	0.31
Range				0.17–6.05	0.04–3.64	
Mean				1.57	1.05	

Earth's surface. The advantages of using ^7Be and ^{210}Pb to determine the total deposition velocities are the following: (1) The production rate of ^7Be and ^{210}Pb at any given site remains constant over long periods of time; (2) The size distributions of ^7Be and ^{210}Pb in aerosols are very similar to other particulate contaminants of interest, and therefore can be used to determine the fluxes of these contaminants to the Earth's surface, from a knowledge of the deposition velocities of these nuclides and the concentrations of these contaminants in air; and (3) The concentrations of ^7Be and ^{210}Pb in the aerosols and precipitation samples can be easily measured. As was discussed earlier, the vertical distribution of ^7Be in the atmosphere is such that it will increase with altitude from the Earth's surface while the opposite is true for ^{210}Pb . Thus, the fluxes to the Earth's surface of other atmospheric contaminants that have sources of either the Earth's surface or stratosphere can be calculated using the deposition velocity of ^7Be and ^{210}Pb .

[33] The total deposition velocity for ^7Be varies between 0.2 and 6.1 cm s^{-1} , with a mean value of 1.6 cm s^{-1} (Table 6). The corresponding values for ^{210}Pb varied between 0.04 to 3.6 cm s^{-1} , with a mean value of 1.1 cm s^{-1} (Table 6). During periods when there is heavy precipitation, the deposition velocity of aerosols is expected to be significantly higher than those during drier periods. The deposition velocity calculated by previous researchers is listed in Table 7 and our values are comparable to the values reported by other investigators. For example, *Preiss et al.* [1996] have reported a mean deposition velocity of 0.6 cm s^{-1} for North America, based on ^{210}Pb .

[34] In 87% of our calculated values (Table 6), the total deposition velocity obtained from ^7Be values are higher than those obtained from ^{210}Pb . The data on the deposition velocity obtained by both nuclides are limited. However, the few published data do indicate that the deposition velocity obtained from ^7Be is higher than those obtained using ^{210}Pb .

Table 7. Summary of Deposition Velocity of Aerosols Using ^7Be and ^{210}Pb

Collection Time	Coordinates Latitude/Longitude	^7Be , cm s^{-1}	^{210}Pb , cm s^{-1}	Reference
Hawaii May–June 1979	21°N, 157°W	3.0	–	<i>Turekian et al.</i> [1983]
Bermuda May–June 1979	33°N, 64°W	4.0	–	<i>Turekian et al.</i> [1983]
Quillayute, Washington 1976–1977	47°N, 124°W	1.0	–	<i>Crecelius</i> [1981]
New Haven, Connecticut March 1977–Feb. 1978	41°N, 73°W	2.8	0.95	<i>Turekian et al.</i> [1983]
Bermuda March 1977–Feb. 1978	33°N, 64°W	–	1	<i>Turekian et al.</i> [1983]
Eastern and Mid–western U.S. 1978	–	–	0.62	<i>Graustein and Turekian</i> [1986]
Norfolk, Virginia 1983–1984	35°N, 84°W	1.3	0.7	<i>Todd et al.</i> [1989]
Groningen, Netherlands 1990–1993	53°N, 6°E	–	1.0	<i>Beks et al.</i> [1998]
Western North Atlantic April 1989	–	–	1.9	<i>Hussain et al.</i> [1998]
Munich, Germany	48°N, 11°E	–	1.0	<i>Winkler and Rosner</i> [2000]
Detroit, Michigan Sept., 1999–Feb. 2001	42°N, 83°W	1.6	1.1	present study

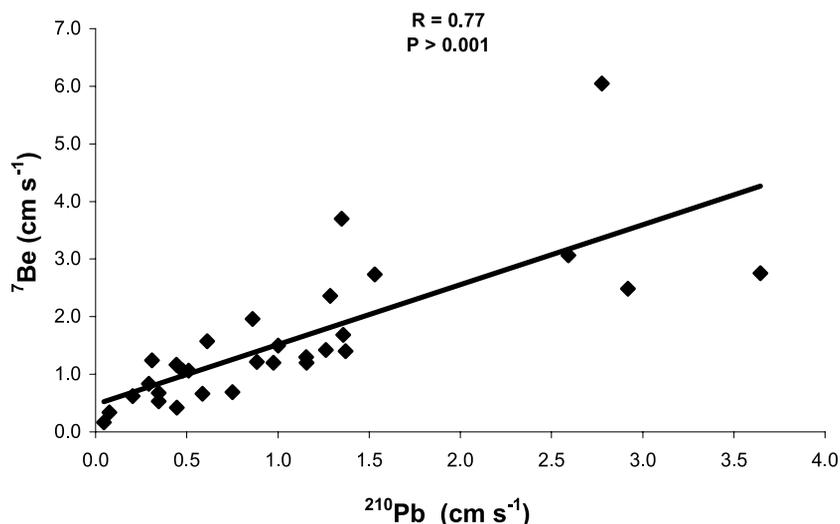


Figure 9. Deposition velocity of aerosols of ^7Be versus ^{210}Pb .

(Table 7, *Turekian et al.* [1983], and *Todd et al.* [1989]). *Todd et al.* [1989] have reported that the deposition velocity of ^7Be is about a factor of 2 higher than that for ^{210}Pb and suggested that this difference is mainly due to artifacts of the manner in which they are calculated. This difference in values reported in this study is beyond the propagated two sigma errors (typically about 10%) associated with the depositional fluxes of ^7Be and ^{210}Pb as well as the concentrations of these nuclides in the air samples. Since the source of ^{210}Pb is the Earth's surface and the concentration of ^{210}Pb in the air closer to the Earth's surface is expected to be higher, the total deposition velocity is expected to be lower. The strong positive correlation between the total deposition velocities obtained using ^7Be and ^{210}Pb ($r = 0.77$, $P > 0.001$, Figure 9; slope value of 0.4) suggests that irrespective of the sources of these nuclides, the deposition velocity obtained by these two nuclides are similar. A positive intercept 0.4 cm s^{-1} possibly indicates even if the deposition velocity is zero for ^{210}Pb , ^7Be will have a finite deposition velocity because of its source. However there are no significant correlations between the total depositional velocities derived from either ^7Be or ^{210}Pb and the total particulate matter load collected onto the filter paper indicating aerosol mass does not control the amount of ^7Be or ^{210}Pb scavenged by the aerosols. We suggest that only a small portion of the aerosol scavenges effectively ^7Be and ^{210}Pb from the atmosphere and a major portion of the aerosols do not participate in the removal of particle-reactive nuclides.

3.11. Washout Ratio

[35] The washout ratio, W , that relates the average concentration of ^7Be or ^{210}Pb in surface level precipitation to its average concentration in unwashed surface level air, is calculated from the following relationship:

$$W = \rho C_{\text{rain}}/C_{\text{air}} \quad (5)$$

where ρ is the density of air at standard conditions (1.2 kg m^{-3} at 20°C and 0.76 m Hg) and C_{rain} and C_{air} are the radionuclide (^7Be or ^{210}Pb) concentrations in bulk deposition (in dpm kg^{-1}) and surface air (in dpm m^{-3}),

respectively. The washout ratio calculation is based on the assumption that the specific ^{210}Pb content of the air in the precipitating cloud is the same as that measured at the surface level in aerosol. However, if there is a new air mass intruding into the area and the precipitation is derived from that air mass, then, this value could result in considerable error. The washout ratios for ^7Be and ^{210}Pb varied between 210–2097, (mean: 948) and 55–2244, (mean: 637), respectively (Table 8). These values can be compared to the values in other places: mean value of 780 for ^{210}Pb in the Northern Wisconsin, *Talbot and Andren* [1983]; 370 and 215, for ^7Be and ^{210}Pb , respectively, *Todd et al.* [1989] for the Southeastern Virginia coast; 144 for ^7Be at Thessaloniki, Greece ($40^\circ 38' \text{N}$; $22^\circ 58' \text{E}$ [*Papastefanou and Ioannidou*, 1991]); and 30 for ^{210}Pb , for the Western North Atlantic, *Hussain et al.* [1998].

[36] The lower washout values for ^{210}Pb are likely due to its origin, the Earth's surface. The concentration of ^7Be is expected to be higher at the points where the cloud condensation takes place as opposed to near the Earth's surface. In the case of ^{210}Pb , the concentration is expected to be low at the point where the cloud condensation takes place and high near the Earth's surface. The possibility of differential washout of ^7Be and ^{210}Pb always exist and cannot be ruled out from our data. *Talbot and Andren* [1983] have shown that ^{210}Pb was removed from the atmosphere with greater efficiency than was stable Pb and attributed this difference to the source function of these species. In the case of atmospheric ^{210}Pb , most of it is derived from the decay of ^{222}Rn that reaches some heights in the atmosphere before its decay while most of the stable Pb was derived from gaseous emission sources and confined to limited vertical distances above the Earth's surface.

4. Conclusion

[37] The simultaneous measurements of atmospheric depositional fluxes of ^7Be and ^{210}Pb and concentrations of these nuclides in aerosols were conducted at Detroit, Michigan, during a 17-month period (September 1999 to February 2001) in order to understand the depositional characteristics of these nuclides. The variations in the

Table 8. Washout Ratio for ^7Be and ^{210}Pb

Sample	Mid-Collection Time	^7Be in Precipitation, dpm L ⁻¹	^7Be in Air, dpm m ⁻³	^7Be Washout Ratio	^{210}Pb in Precipitation, dpm L ⁻¹	^{210}Pb in Air, dpm m ⁻³	^{210}Pb Washout Ratio	$^7\text{Be}/^{210}\text{Pb}$ Air Ratio
AF-1	Oct. 28, 1999	103	0.588	210	11.7	0.253	55	2.32
AF-2	Oct. 31, 1999	103	0.292	424	11.7	0.147	95	1.98
AF-3	Jan. 10, 2000	195	0.170	1376	39.9	0.077	621	2.21
AF-4	Jan. 23, 2000	195	0.216	1083	39.9	0.126	379	1.71
AF-5	Jan. 26, 2000	195	0.274	855	39.9	0.063	761	4.35
AF-6	Feb. 4, 2000	234	0.264	1065	36.5	0.080	546	3.29
AF-7	Feb. 8, 2000	234	0.336	836	36.5	0.080	546	4.18
AF-8	March 9, 2000	300	0.283	1279	25.1	0.027	1133	10.7
AF-9	March 22, 2000	300	0.301	1194	25.1	0.035	869	8.69
AF-10	April 15, 2000	157	0.284	664	10.5	0.043	291	6.55
AF-11	May 1, 2000	131	0.341	462	12.8	0.035	442	9.81
AF-12	May 3, 2000	131	0.293	537	12.8	0.029	525	10.0
AF-13	May 13, 2000	233	0.220	1273	15.0	0.030	587	7.16
AF-14	May 15, 2000	233	0.433	646	15.0	0.033	549	13.2
AF-15	May 23, 2000	350	0.396	1060	26.6	0.055	578	7.18
AF-16	May 25, 2000	350	0.340	1236	26.6	0.019	1740	17.4
AF-17	July 1, 2000	422	0.392	1291	42.5	0.049	1044	8.03
AF-18	July 8, 2000	422	0.266	1906	42.5	0.023	2244	11.7
AF-19	July 13, 2000	422	0.464	1091	42.5	0.052	971	8.84
AF-20	July 25, 2000	422	0.241	2097	42.5	0.043	1178	5.58
AF-21	Aug. 13, 2000	257	0.448	688	28.3	0.045	751	9.91
AF-22	Aug. 25, 2000	90.3	0.330	328	12.5	0.056	266	5.86
AF-23	Aug. 30, 2000	90.3	0.265	408	12.5	0.055	274	4.84
AF-24	Sept. 21, 2000	116	0.093	1502	17.4	0.038	547	2.43
AF-25	Sept. 30, 2000	116	0.218	640	17.4	0.084	248	2.59
AF-26	Oct. 12, 2000	115	0.247	559	44.0	0.120	441	2.06
AF-27	Oct. 27, 2000	76.4	0.233	393	24.6	0.071	415	3.28
AF-28	Nov. 30, 2000	75.4	0.105	860	21.1	0.077	327	1.36
AF-29	Jan. 22, 2001	303	0.263	1385	54.7	0.146	450	1.80
AF-30	Feb. 3, 2001	159	0.173	1103	15.7	0.082	230	2.11
Range				210–2097			55–244	
Mean				948			637	

depositional fluxes (both dry and bulk) of ^7Be and ^{210}Pb as well as the deposition velocities and the washout ratios for these nuclides in this site are similar to other study sites in the Midwest. From this investigation, we draw the following conclusions:

1. The depositional fluxes of ^7Be and ^{210}Pb during the 17 months study period varied between 3.11–63.0 dpm cm⁻² yr⁻¹ (mean = 11.4 dpm cm⁻² yr⁻¹) and 0.72–10.3 dpm cm⁻² yr⁻¹ (mean = 1.41 dpm cm⁻² yr⁻¹), respectively. This variability in the depositional fluxes of ^7Be and ^{210}Pb are attributed to the frequency and amounts of precipitation, and seasonal variations on the depositional fluxes.

2. The dry depositional fluxes of ^7Be and ^{210}Pb contributed 2.1–19.8% and 3.6–48.6% of the bulk depositional fluxes, respectively. These varying percentages are attributed to the variable frequency of the rainfall. The difference between these nuclides is attributed to the differences in their source function.

3. The precipitation-normalized fluxes of ^7Be and ^{210}Pb show seasonal changes: increase during spring and summer and a significant depletion during the fall for ^7Be and depletion during spring with enrichment during winter and summer for ^{210}Pb . Higher precipitation-normalized enrichment factor for ^7Be during spring is attributed to the stratosphere-troposphere exchange of air masses and large-scale atmospheric mixing during summer.

4. It appears that snow is more efficient than rain in removing ^{210}Pb from the atmosphere and this is likely due to higher surface area of snow flurries compared to the rain droplets.

5. It appears that five heavy rain events, with 31% of the total rainfall and 4% of the total number of rainy days, in the year 2000, deposited 27.4% and 28.1% of the total annual depositional fluxes of ^7Be and ^{210}Pb , respectively, suggesting higher amounts of rainfall leads to efficient removal of these nuclides.

6. The volume-weighted concentrations of ^7Be (172 dpm L⁻¹) and ^{210}Pb (21.2 dpm L⁻¹) are the highest reported in literature so far, and we attribute this to the higher frequency of rainfall.

7. The concentrations of ^7Be and ^{210}Pb in aerosols varied between 0.09 and 0.59 dpm m⁻³ (mean = 0.29 dpm m⁻³) and 0.018 and 0.253 dpm m⁻³ (mean = 0.069 dpm m⁻³), respectively. The concentration of ^7Be showed a seasonal increase during the spring and summer months and no seasonal variation was observed for ^{210}Pb . There was no significant decrease in the concentration of ^7Be during months with higher precipitation amounts, suggesting that during these months there was additional input of ^7Be from the upper troposphere.

8. The deposition velocities of aerosols laden with ^7Be and ^{210}Pb varied between 0.2–6.1 cm s⁻¹ (mean: 1.6 cm s⁻¹) and 0.04–3.6 (mean: value 1.1 cm s⁻¹). The corresponding washout ratios varied between 210–2097 (mean: 948) and 55–2244 (mean: 637), respectively. There is no correlation between the aerosol mass and concentrations of ^7Be and ^{210}Pb . There is no significant correlation between the mass of the particulate matter retained on the filter paper and depositional velocities of ^7Be and ^{210}Pb in the air masses. We propose that relatively a minor portion of

the aerosols actively participate in the removal of these nuclides from the air masses.

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