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M. Baskaran

Texas A & M University - Galveston, baskaran@wayne.edu

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Comments on "Measurements of ^7Be and ^{210}Pb in Rain, Snow, and Hail"

M. BASKARAN

Department of Oceanography, Texas A&M University—Galveston, Galveston, Texas

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The authors have reported the first measurement of ^7Be and ^{210}Pb of a hail sample and deserve due credit for their fine work. However, it appears that there are some problems with their data as well as interpretations. Since the daughter products of ^{222}Rn and ^7Be (also other cosmogenically produced isotopes) that are produced in the lower and upper atmosphere, respectively, are powerful tracers of atmospheric and meteorological processes, it is important that the right perspective is given so that a wider community can possibly use these isotopes to investigate many other meteorological and atmospheric processes.

This paper reported measurements of ^7Be and ^{210}Pb on five rainwater samples during thunderstorms, one sample each from a stratus storm system and one snow. Even though it is a common practice now to acidify the rain collectors before the collection of samples to minimize the wall adsorption of ^7Be and ^{210}Pb , this probably was not a serious problem since the authors collected the samples on individual events and the sampling time lasted for at most 13 h. The complete information on when the samples were processed after collection is not given, as retaining the samples for longer periods of time will likely yield lower values of the concentrations and fluxes due to sorption of ^7Be and ^{210}Pb onto container walls. However, the paper states that the samples were stored until the bismuth was essentially in secular equilibrium with the lead (typically 10 days) and during this 10 days, significant amounts of ^{210}Pb (as well as ^7Be) could have been removed on to the container walls from the unacidified water sample. The authors have not given any information on the sampling site, how high the sampling site is from the ground, etc. It is very important to provide this information, as resuspended fine dust particles during the initial phase of the storm event can easily get into the rain collector. I suspect that the higher particulate fraction of ^{210}Pb is due to the resuspended material. Since there is no year-around data on the

depositional fluxes of ^7Be and ^{210}Pb for both places (Socorro, New Mexico, and West Chicago, Illinois) in published literature, a quantitative evaluation on the contribution of ^7Be and ^{210}Pb from resuspended particles can be made by taking fallout data from another site. Our published data on the depositional fluxes of ^7Be and ^{210}Pb in Galveston can be used for this calculation. The mean annual depositional fluxes of ^7Be and ^{210}Pb collected during 1989 to 1991 are 14.7 and 1.07 dpm (disintegrations per minute) $\text{cm}^{-2} \text{yr}^{-1}$, respectively (Baskaran et al. 1993). The corresponding expected inventories of ^7Be and ^{210}Pb in soil cores would be 3.08 and 34.1 dpm cm^{-2} , respectively. If most of ^7Be and ^{210}Pb are confined to the top 2 cm, and the soil dry density is 2.5 g cm^{-3} , then the expected ^{210}Pb concentration in the upper 2-cm soil would be 0.6 dpm g^{-1} of ^7Be and 6.8 dpm g^{-1} of ^{210}Pb . Thus, the contribution of detritus-derived ^{210}Pb concentration should be 11.3 times higher than that of ^7Be . Indeed, the data presented in Table 2 clearly show higher particulate ^{210}Pb compared to ^7Be ; the authors seem to have overlooked this important point. The particulate ^7Be varies between 1.2% and 12.7% with a mean of 4.5%, whereas that of ^{210}Pb varies between 74% and 86% with a mean of 74.5%. The ratio of ^{210}Pb to ^7Be in particles is 16.6, which can be compared to the value of 11.3 expected if the penetration depths of ^{210}Pb and ^7Be are the same. The resuspended fine particles that were collected in the rain collector could have removed some of the dissolved ^7Be and ^{210}Pb , between the time of collection and the time of acidification of the sample (at least 10 days). It is pertinent to point out that the K_d of ^7Be and ^{210}Pb are similar in surface waters (Baskaran and Santschi 1993).

Higher ^7Be concentrations in the rain samples due to increased stratosphere-troposphere mixing. The authors have reported four measurements that were made during spring (22 March 1990, 29 March 1990, 9 April 1990, and 15 April 1992). Except in one sample (29 March 1990), all other three values of the concentrations are not distinguishable from the concentrations during nonspring months. Thus, the presented data do not seem to show any trend with season. Also, only Chicago, Illinois (41.8°N), is in the midlatitude,

Corresponding author address: Dr. M. Baskaran, Dept. of Oceanography, Texas A&M University—Galveston, Fort Crockett Campus, 5007 Avenue U, Galveston, TX 77551.
E-mail: Baskaran_M@tamug2.tamu.edu

whereas Socorro, New Mexico (31.5°N), is not in the midlatitude (38° – 51°).

There is a general tendency among researchers who work in the fallout of atmospheric radionuclides to attribute any increase in the depositional fluxes to the troposphere–stratospheric exchange (Olsen et al. 1985; Todd et al. 1989; Gaffney et al. 1994). Recent study shows that several meteorological variables, such as seasonal variability of the amounts of precipitation, vertical mixing of lower and upper troposphere, seasonal variations on the cloud altitude, and seasonal variability on the sources of water vapor, could explain the seasonal variability of the depositional fluxes of ^7Be at any given location (Baskaran 1995). Since at least 90% of the bulk fallout of ^7Be and ^{210}Pb are through precipitation, the amount of precipitation primarily controls the depositional fluxes of these radionuclides. When the amount of rainfall increases, in a season, or a year, the depositional flux also increases for that season or year. For example, the depositional flux of ^7Be increased from 8.9 to 23.2 $\text{dpm cm}^{-2} \text{yr}^{-1}$ when the amount of precipitation increased from 103 cm in 1989 to 150 cm in 1991 at Galveston (Baskaran et al. 1993).

The stratospheric source of ^7Be becomes significant during the spring when midlatitude (38° – 51°) folding of the tropopause enhances the stratospheric–tropospheric exchange. The ^7Be concentration in the air samples increase during this time. When such an increase of ^7Be takes place, the concentration of ^{210}Pb in air should theoretically remain the same. This increase in ^7Be concentration in air will be likely reflected in

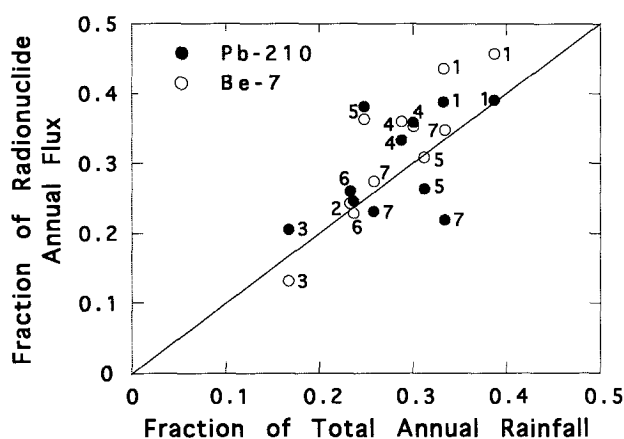


FIG. 1. Fractional amounts of ^7Be and ^{210}Pb fluxes (depositional flux in a season/annual depositional flux) plotted against fractional amount of rainfall (rainfall in a season/annual rain fall) for spring. Those data points that fall above the 45° diagonal line (slope = 1) have seasonal increase in the depositional flux of that nuclide (^7Be or ^{210}Pb). The key for the numbers given in the figure are as follows (each time a number appears belong to one year): 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). Figure is taken from Baskaran (1995).

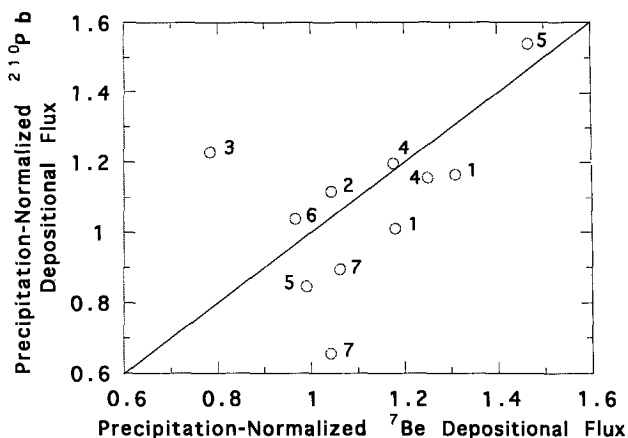


FIG. 2. The precipitation-normalized ^{210}Pb flux data in spring [α = annual rainfall \times seasonal flux of ^{210}Pb /(amount of rainfall in spring \times annual flux of ^{210}Pb)] is plotted against precipitation-normalized ^7Be flux data in spring [α = annual rainfall \times seasonal flux of ^7Be /(amount of rainfall in spring \times annual flux of ^7Be)]. Data points below 45° diagonal line have lower depositional flux for ^7Be , whereas those above the line have higher ^{210}Pb flux. The key for the numbers given in Fig. 2 is as follows (each time a number appears belong to one year): 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). Figure is taken from Baskaran (1995).

the depositional fluxes of ^7Be . Thus, any seasonal increase of ^7Be in air due to tropospheric–stratospheric exchange should be clearly seen on precipitation-normalized depositional fluxes, and such relationships should not be seen for ^{210}Pb .

All the published data on the simultaneous depositional fluxes of ^7Be and ^{210}Pb since 1980 were recently analyzed (Baskaran 1995). The fractional depositional fluxes (= depositional flux during spring/annual flux) of ^7Be and ^{210}Pb are replotted against the fractional amount of precipitation (= rainfall during spring/total annual rainfall) in Fig. 1 for all the stations where simultaneous depositional fluxes of ^7Be and ^{210}Pb are available (Baskaran 1995). All the data points with slopes greater than 1 correspond to higher precipitation-normalized depositional fluxes and those slopes less than 1 to lower values. For ^7Be , 8 points are above the 45° diagonal line, whereas for ^{210}Pb 7 points lie above the line. Thus, it appears that the depositional fluxes of both ^7Be and ^{210}Pb are higher in spring.

The precipitation-normalized depositional flux of ^{210}Pb is plotted against the precipitation-normalized depositional flux of ^7Be in Fig. 2. When the data points lie below the 45° diagonal line, there is a seasonal increase of ^7Be over ^{210}Pb . There are 6 points out of a total of 11 fall below this line. The slight increase in depositional fluxes of ^7Be and ^{210}Pb during spring was hypothesized as due to changes in atmospheric processes, such as vertical mixing of the troposphere due

to heating in late spring, rather than the contribution from stratosphere-troposphere exchange of air masses (Baskaran 1995).

Due to the distinct sources of ^7Be and ^{210}Pb , the $^7\text{Be}/^{210}\text{Pb}$ ratios have great potential of being used for many meteorological and atmospheric processes. More individual interval sampling of rainfall, snow, and hail samples are needed for these isotopes to be used as powerful tracers.

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