Measurements of ^{7}Be and ^{210}Pb in cloudwaters: Toward a better understanding of aerosol transport and scavenging

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[1] This paper presents the first results of ^{7}Be and ^{210}Pb specific activities in cloudwater and investigates their respective differences in cloudwater and rainwater. Our data and analysis show that temporal variation of the ^{7}Be/^{210}Pb activity ratio in cloudwater are caused by multiple sources of air masses, with increased ^{7}Be/^{210}Pb activity ratios corresponding to the advent of maritime or high-altitude air masses. The measured specific activities of ^{7}Be and ^{210}Pb in cloudwater samples span three orders of magnitude and correlate strongly with the acidity of the samples. From this correlation and the observation that specific activities of ^{7}Be and ^{210}Pb are higher in the more acidic cloudwater, we suggest that ^{7}Be and ^{210}Pb in aerosol particles can be desorbed by H^{+} ions. **Citation:** Su, C.-C., and C.-A. Huh (2006), Measurements of ^{7}Be and ^{210}Pb in cloudwaters: Toward a better understanding of aerosol transport and scavenging, *Geophys. Res. Lett.*, *33*, L04807, doi:10.1029/2005GL025042.

1. Introduction

[2] Be-7 ($T_{1/2}$ = 53.3 d) and ^{210}Pb ($T_{1/2}$ = 22.3 yr) have been widely used as tracers for studying atmospheric transport, exchange, and aerosol removal processes, and to test global circulation models [*Junge*, 1963; *Balkanski et al.*, 1993; *Baskaran et al.*, 1993; *Baskaran*, 1995; *Rehfeld and Heimann*, 1995; *Koch et al.*, 1996; *Tokieda et al.*, 1996; *Caillet et al.*, 2001]. Following their production in the atmosphere, ^{7}Be and ^{210}Pb are quickly adsorbed to aerosol particles and are removed by radioactive decay and by wet and dry deposition, with mean residence times in the troposphere ranging from days to more than one month [*Bleichrodt*, 1978; *Tokieda et al.*, 1996; *Caillet et al.*, 2001; *Su and Huh*, 2002]. With more data accumulating, there is a general consensus that wet deposition is the primary removal process for these radionuclides [*Small*, 1959; *Todd et al.*, 1989; *Baskaran*, 1995; *Su et al.*, 2003]. However, details of the scavenging process by wet deposition are not yet fully understood.

[3] Atmospheric aerosols may be scavenged by precipitation due to Brownian diffusion, thermo- and diffusiophoretic forces, inertial impaction, electrical forces, and cloud condensation nuclei [*Chate et al.*, 2003]; all these processes may be broadly divided into rainout (in-cloud) and washout (below-cloud) processes. In most, if not all, previous studies, measurements of ^{7}Be and ^{210}Pb were made on either aerosol (i.e., air filter) or rainwater samples whereby it was not possible to discriminate between these two processes. For a better understanding of scavenging of ^{7}Be and ^{210}Pb by wet deposition, it is necessary to collect and analyze cloud water as well.

[4] Yang-Ming Shan, a national park in northern Taiwan, is often covered by clouds and has unusually high fluxes of ^{7}Be and ^{210}Pb [*Huh and Su*, 2004]. It is therefore ideally suited for this study.

2. The Sampling and Analytical Methods

[5] Cloudwater and rainwater samples were collected between February 2004 and March 2005 at the Leng-Shui Keng (LSK) experimental site (25°9′54″, 121°33′55″, 730 m a.s.l.), which is located at the foot of the Seven Star Mountain (1120 m a.s.l.) in the Yang-Ming Shan National Park (Figure 1). The experimental site is located at the saddle between the Seven Star Mountain and Zhu-Gao Shan (830 m a.s.l.), overlooking a valley in the northeast direction. Such an orographic terrain makes the site frequently shrouded in cloud mists due to the passage of fronts, especially during the northeast monsoon season.

[6] Cloud mists were sampled using a single stage Caltech Active Strand Cloud water Collector (CASCC2) which consisted of a bank of 3 rows each containing 32 Teflon strands (508 μm diameter) spaced 2.5 mm apart and a 100/120V, 50/60Hz fan (flow rate 6.3 m^{3} min^{-1}). During operation, cloud mists were condensed and dripped into a 2-L polyethylene bottle. A more detailed description of the CASCC2 is given by *Demoz et al.* [1996]. In this study, we also collected rainwater concurrent with cloud water. The rainwater collector consisted of a polyethylene bucket (29 cm in diameter) with a nylon mesh at the top to prevent insects and plant debris from falling in. Each time after the sample was poured out, the bottle and bucket were cleaned with 5% neutral detergent solution (Merck Extran® MA 02), then filled with 5% HNO_3 and left to stand for at least one week. Prior to using, all apparatus and containers were rinsed with distilled water and dried in a laminar flow bench.

[7] Most of the cloudwater and rainwater samples were collected for a period of 1-3 days. When the water levels were exceptionally low, the sampling time was extended up to one week. Upon collection, the samples were immediately shipped to the laboratory for the measurement of pH using a pH meter (Radiometer, Model PHM210) with the combination electrode (Radiometer, Model PHC3001) calibrated with pH 4 and 7 standard buffers. The samples were then filtered (0.45μm, Millipore HA) and evaporated on a hot plate for volume reduction. During the evaporation and sample transfer processes,

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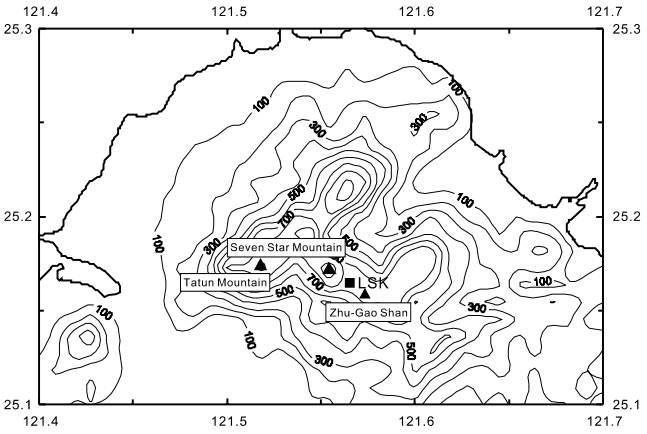

Figure 1. Map showing the location of Leng-Shui Keng (LSK), the experimental site, (solid square) in northern Taiwan.

the sizes of the beakers were also reduced step-by-step. Whenever a beaker was emptied, it was acid-washed (by refluxing with HNO_3 and $HClO_4$) and the washings were combined with the condensate in the next, smaller beaker for further evaporation. Eventually, the sample was reduced to a nominal size of ~0.5 ml, from there the final condensate and beaker washings were transferred to a Kimax tube with a final volume of about 1 ml.

[8] Gamma counting was performed using a well-type HPGe detector (EG&G ORTEC GWL-100230) interfaced with a digital γ-ray spectrometer (EG&G ORTEC DSPec™) for the determination of ^{210}Pb and ^{7}Be. Efficiencies of the detector as functions of sample volume and γ-ray energy have been carefully calibrated using Harwell uraninite and IAEA-133B standard solutions. For 1-ml samples, the absolute counting efficiency is 63.7 ± 0.4% for ^{210}Pb (at 46.52 keV) and 20.3 ± 0.3% for ^{7}Be (at 477.56 keV). The data reported here have been decay-corrected to the midpoint of sample collection.

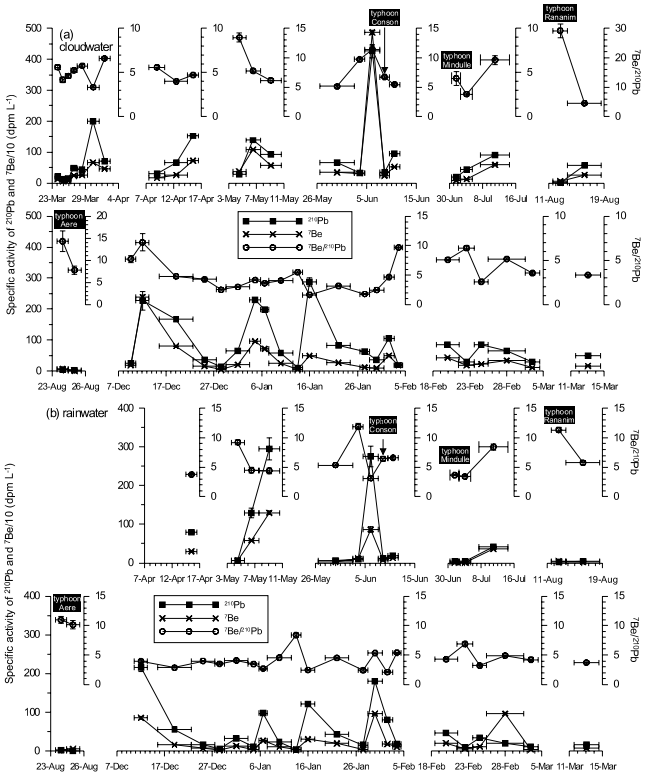

Figure 2. Time series of ^{7}Be and ^{210}Pb specific activities and ^{7}Be/^{210}Pb activity ratios in (a) cloudwater and (b) rainwater samples at the LSK experimental site.

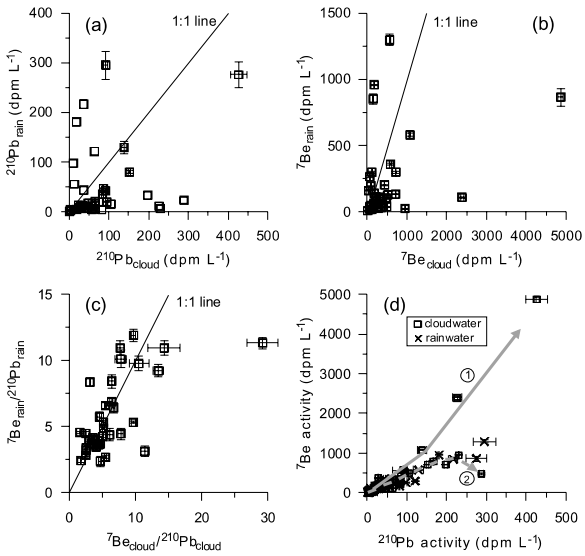

Figure 3. Scatter diagrams of the specific activities of (a) ^{210}Pb$_{rain}$ vs. ^{210}Pb$_{cloud}$, (b) ^{7}Be$_{rain}$ vs. ^{7}Be$_{cloud}$, (c) ^{7}Be$_{rain}$/^{210}Pb$_{rain}$ vs. ^{7}Be$_{cloud}$/^{210}Pb$_{cloud}$, and (d) ^{7}Be vs. ^{210}Pb in cloudwater and rainwater samples. In Figure 3d, the cloudwater samples can be divided into high (1, maritime air masses; gray solid line) and low (2, NE monsoon; gray dashed line) ^{7}Be/^{210}Pb activity ratio groups.

3. Results and Discussion

[9] There are several meteorological systems controlling the variation of weather in Taiwan. In winter and early spring when the northeast monsoon prevails over East Asia, the wind direction at our experimental site is up to 51% from the northwest in the 315–360° sector of the wind rose. In summer and fall when the southwest monsoon introduces air masses into Taiwan, the wind direction at the experimental site is from the southeast (25%, 135–180°). In-between the NE and SW monsoons are the transitional periods of frontal surface and Mei-yü (plum rain) from mid-March until June. Typhoon is another perennial phenomenon frequently invading Taiwan from July to September. The on-site meteorological data indicate that the wind direction is mainly steered by the topography surrounding the experimental site. According to our meteorological data and 7-day back trajectory analyses (AERONET program; http://croc.gsfc.nasa.gov/aeronet/), the cloudwater samples can be divided into three groups based on their sources: (1) northeast monsoon samples (November–February), (2) frontal passage and Mei-yü samples (March–May), and (3) typhoon and southwest airflow samples (June–August).

[10] Figure 2 shows the temporal variations of ^{7}Be and ^{210}Pb specific activities and ^{7}Be/^{210}Pb activity ratios in

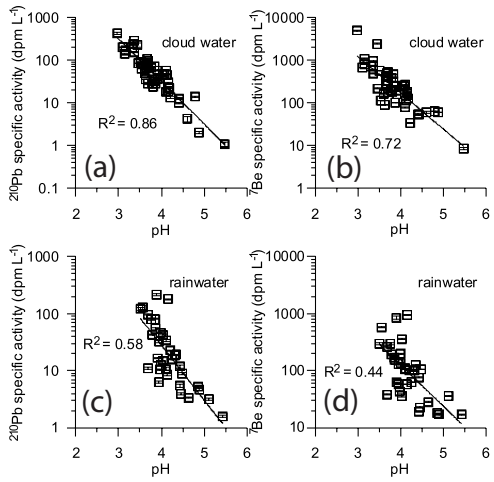

Figure 4. Specific activities of ^{210}Pb and ^{7}Be vs. pH in (a and b) cloudwaters and (c and d) rainwaters.

cloud and rain water at the LSK experimental site. During the NE monsoon season (Dec.–Feb.), ^{7}Be and ^{210}Pb specific activities show a cyclic pattern (Figure 2a), with each peak representing a cold spell due to the intrusion of cold fronts. The time series of ^{7}Be and ^{210}Pb specific activities exhibits a trend opposite to that of cloudwater volume and rainfall. This phenomenon is commonly observed for rainwater and it may be caused by the dilution effect of rainwaters [*Tang et al.*, 2005]. Alternatively, it may be related to the sizes of cloudwater droplets and rainwater drops, which tend to be larger for dense clouds and heavy rainfalls. Consequently, cloud and raindrops with smaller sizes have longer residence time and larger surface areas to scavenge more aerosol particles and gases [*Lee et al.*, 2000] thus raising their ^{7}Be and ^{210}Pb specific activities. Another important point is the relationship between the ^{7}Be/^{210}Pb activity ratio and the source of air masses. Figure 2 shows that, of the 47 cloudwater samples analyzed most have ^{7}Be/^{210}Pb activity ratios in the range of 5 ± 2, with only 5 exceeding 10 and the highest one reaching 30. Through a close examination of the weather system and back trajectory analysis, we found that these high ^{7}Be/^{210}Pb activity ratios were associated with maritime air masses introduced either by the Mei-yü front (5/3/04–5/5/04) or by typhoons (Rananim: 8/11/04–8/13/04 and Aere: 8/23/04–8/24/04). The lower ^{7}Be/^{210}Pb values in cloudwaters collected during the invasion of typhoon Conson (6/8/04–6/10/04) and typhoon Mindulle (7/1/04–7/3/04) are due to substantial modification of marine air by continental air masses on their movement tracks though land masses in the southeast Asia and Philippines. The high ^{7}Be/^{210}Pb ratio during the period of 4/6/04–7/6/04 prior to the arrival of typhoon Conson can be attributed to the transport at high altitude (5–6 km) of high latitude (60–70°N) air mass derived from Siberia. As for the high ^{7}Be and ^{210}Pb specific activities at the same time, it may be due to the low water content of the cloudwater and rainwater.

[11] For most of the time, specific activities of ^{7}Be and ^{210}Pb in cloudwater samples are higher than those in concurrent rainwater samples (Figures 3a and 3b). This may be due to inefficient scavenging of ^{7}Be and ^{210}Pb by rapidly falling raindrops, resulting in lower concentrations of these two radionuclides in rainwater compared with cloudwater samples. In contrast to the difference in specific nuclide activities between rainwater and cloud water, the ^{7}Be/^{210}Pb ratios in rainwater and corresponding cloudwater samples agree well with each other, except that there is no value higher than 12 in rainwater samples (Figure 3c). It can also be noted in Figure 3d that the correlation between ^{7}Be and ^{210}Pb is better for rainwater samples than for cloudwater samples. Based on Figure 3d, it appears that cloudwaters can be divided into two distinct groups; samples in the group with a higher ^{7}Be/^{210}Pb ratio came from maritime air masses, while those fitting a lower ^{7}Be/^{210}Pb activity ratio were collected during the northeast monsoon season. In contrast to the apparent disparity in ^{7}Be/^{210}Pb for cloudwaters of different sources, there is less variation in ^{7}Be/^{210}Pb for rainwaters, probably suggesting the efficacy of convective mixing below clouds.

[12] Figure 4 shows the correlation between the pH and specific activities of ^{7}Be and ^{210}Pb in cloudwater and rainwater samples. The pH values in cloudwater and rainwater samples range between 3 to 5.5 and 3.5 to 5.4, respectively. It is interesting to point out that, pH of cloudwater is systematically lower than pH of concurrent rainwater samples by half a unit on average. It is also important to note the correlation between nuclide specific activities and pH, which is especially strong for cloudwater samples, with nuclide specific activities spanning three orders of magnitude while pH spanning 2.5 units. This result implies that ^{7}Be and ^{210}Pb are not necessarily scavenged from the atmosphere by aerosol particles irreversibly. As cloudwaters become more acidic, ^{7}Be and ^{210}Pb will be desorbed from particles and released into cloud mist/vapor. In a previous study by us [*Huh and Su*, 2004], it's found that inventories of ^{7}Be and ^{210}Pb in soils at the Yang-Ming Shan area are much higher than expected from the amount of rainfall [*Huh and Su*, 2004]. From this study, we have gleaned data to support our previous speculation that low flying clouds may play an important role in transporting fallout nuclides directly into soils in mountainous areas.

4. Conclusions

[13] Forty-seven cloudwater and 39 rainwater samples collected during the period from February 2004 to March 2005 were analyzed for ^{7}Be and ^{210}Pb. The results show that cloudwater samples have higher ^{7}Be and ^{210}Pb specific activities and lower pH values than concurrent rainwater samples. The temporal variation of the ^{7}Be/^{210}Pb activity ratio is controlled by different air masses from multiple source region, altitude and transport pathways. Our results show that ^{7}Be and ^{210}Pb can be desorbed from aerosol particles by H^{+} ions; their sorption behaviors and concentrations in cloud and rain waters are strongly dependent on pH.

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