The Use of Naturally Occurring Radionuclides (^{7}Be and ^{210}Pb) as Atmospheric Tracers

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The ^{210}Pb and ^{7}Be activities in daily aerosol samples from three stations in the North Atlantic [Bermuda, Barbados, and Izaña (Tenerife, Canary Islands)] were combined with meteorological information to investigate how the composition of aerosol particles varies in response to transport pathway and precipitation scavenging. These two nuclides are useful tracers of air mass history owing to their unique sources: ^{210}Pb (half life, $\tau_{1/2}$ = 22 years) is supplied to the atmosphere by the radioactive decay of ^{222}Rn, which is a noble gas produced in turn from the decay of primordial ^{238}U. In contrast ^{7}Be ($\tau_{1/2}$ = 53-days) is produced *via* cosmic ray spallation reactions with nitrogen and oxygen in the stratosphere and upper troposphere. After production, both nuclides rapidly attach to existing aerosol particles, and the relative activities of the two nuclides provide an indication of the strengths of continental emissions vs. air from upper troposphere and lower stratosphere (UT/LS).

For this study, over three years of radionuclide data, amounting to more than 1200 aerosol samples from each site, were analyzed as part of the Atmosphere/Ocean Chemistry Experiment (AEROCE), a multidisciplinary program sponsored by the National Science Foundation. One set of studies for AEROCE is concerned with evaluating and comparing the impacts of anthropogenic emissions vs. natural processes on the ozone budget and oxidizing capacity of the troposphere over the North Atlantic. The pair of nuclides studied here provides data particularly relevant for the interpretation of tropospheric O_3 because the O_3 mixing ratios are determined by the same two sources that control the relative abundances of the two nuclides, i.e., continental emissions and transport from the UT/LS. A second focus of AEROCE is on the physical and chemical characteristics of aerosols in the marine atmosphere, especially pollutants and those important to the radiative properties of the atmosphere and hence climate. Our studies of radionuclides also are relevant to the AEROCE aerosol focus because they provide information on sources and on particle removal by precipitation scavenging.

The activities of ^{7}Be and ^{210}Pb in the atmosphere over the North Atlantic basin vary geographically and change from year-to-year; furthermore the two nuclides' relative proportions often change abruptly. Both radionuclides exhibit substantial variability with season, but the amplitude of the ^{7}Be cycle is compressed relative to that of ^{210}Pb, especially at Barbados and

Izaña. Short-term effects on the radionuclide loadings due to precipitation scavenging are superimposed on the seasonal cycles, and both types of influences are tied to large-scale circulation patterns.

Significant interannual variability in ^{210}Pb was found at all sites, and the ^{210}Pb activities at Barbados and Izaña were higher than at Bermuda, a pattern consistent with the loadings of atmospheric mineral dust but different from anthropogenic substances. At Bermuda high values for a combined radionuclide loading index (RLI) were matched to air-mass trajectories from the northwest while low RLIs, due at least in part to precipitation scavenging, generally occurred under easterly flow. The activities of ^{7}Be at the Izaña observatory (elevation 2367 m) were much higher than at Bermuda or Barbados, which are situated near sea level, demonstrating the stronger UT/LS influence at Izaña. The ^{7}Be activities at Izaña generally decreased in the late fall and early winter, but wintertime peaks in the ^{7}Be/^{210}Pb ratio resulted from even sharper decreases in ^{210}Pb; this illustrates the necessity for considering both nuclides when evaluating sources. At all sites the strongest UT/LS influence (high ^{7}Be/^{210}Pb ratios) occurred in winter, consistent with the descent of air from high altitudes over the North Atlantic basin, but differences among sites reflect heterogeneity over the basin. The RLIs vary independently of the ^{7}Be/^{210}Pb ratios at all sites, indicating that different processes control the loadings and relative abundances of these two radionuclides over the North Atlantic.

The high loadings of ^{7}Be and ^{210}Pb at Bermuda under northwesterly flow are noteworthy because high mixing ratios of ozone at Bermuda also are associated with transport from North America, and the same is true for various types of aerosol pollutants. The interpretation of the data is complex, however, because these various substances originate from different sources, and ^{7}Be in particular is not continental in origin. Thus, the high mixing ratios of ozone at Bermuda are clearly associated with mixed air masses containing both continental boundary layer air and UT/LS air. A further complication is that the radionuclide aerosol loadings vary in response to precipitation scavenging, but that effect too, is tied to the trajectory pathways.

Comparisons between the rainy day vs. non-rainy day aerosol samples from Bermuda support the theory that precipitation scavenging removes both ^{7}Be and ^{210}Pb from the atmosphere, but the scavenging process evidently does not affect the relative proportions of the two nuclides. One set of comparisons showed that the activities of both nuclides and RLIs were lower in the samples collected on rainy days vs. those collected on non-rainy days, and this is what one would expect if the aerosols carrying the nuclides were scavenged from the atmosphere by rain. A second comparison showed the mean log(^{7}Be/^{210}Pb) ratios were not different between the two sets of samples, indicating there was no selective scavenging of nuclides. Little or no evidence for a day-of-the-week (DOW) effect on aerosol loadings or precipitation was observed for the samples from Bermuda. Even less evidence supports the hypothesis that a weekly cycle has been imposed on radionuclide activities as a result of a DOW cycle in precipitation scavenging.