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Differential Degassing Behavior of Radiocarbon (^{14}C) and Stable Inorganic Carbon in Surface Waters: Effects of pH and Surface-Area-to-Volume Ratio

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Carbon-14 (^{14}C) is a long-lived beta-emitting radionuclide and a key contributor to radiation dose assessments for radioactive waste disposal facilities. Therefore, an accurate safety assessment requires appropriate biosphere modeling for ^{14}C migration. Potential migration pathways of ^{14}C include transport through the vadose zone and aquifers via groundwater, ultimately reaching surface water and causing aquatic contamination. In current dose assessments, ingestion of contaminated aquatic products is considered one of the primary exposure pathways. However, for an accurate safety assessment, the behavior of ^{14}C in surface water must be carefully examined. In surface water bodies such as oceans and lakes, carbonate-form ^{14}C undergoes degassing into the atmosphere. As a result, ^{14}C concentration in surface water should decrease, a phenomenon not currently accounted for in existing safety assessments. For waste forms containing inorganic carbon, the impact of ^{14}C degassing into the atmosphere should be incorporated into safety assessments. To ensure proper integration, key factors affecting the degassing rate of ^{14}C in surface water must be identified.

This study examines two key factors: solution pH and the surface-area-to-volume ratio of the solution in contact with the atmosphere. The study employed both experimental observations and computational analysis. In the first experiment, $Na_2{}^{14}CO_3$ was added to solutions with different pH levels, and the decrease in ^{14}C concentration over time was measured using a Liquid Scintillation Counter (LSC). The results demonstrated that lower pH levels led to a more rapid decrease in ^{14}C concentrations. In the second experiment, the surface-area-to-volume ratio was adjusted, and the results indicated that a higher ratio(corresponding to shallower solution depth) led to a more rapid loss of ^{14}C. The experimental results were analyzed using a computational model based on equilibrium dynamics among carbonate species, the dehydration kinetics of carbonic acid, and the transport of $^{14}CO_2$ (aq) to the atmosphere based on Fick's law. Model predictions closely matched the experimental observations. The findings of this study are expected to contribute to the enhancement of safety assessments by providing a better understanding of ^{14}C behavior in surface water environments.

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