The Effect of Radon-222 Escape on ^{210}Pb-Based Sediment Chronology: Implications for Dating Accuracy in Lacustrine Systems

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The radiochronology method applied to investigate changes in lake ecosystems over the past 150 years is based on the ^{210}Pb isotope from the ^{238}U decay series. When using this method, measurable variations in the concentration of ^{210}Pb deposited from the atmosphere into lake sediments indicate the age of sediment layers (excess ^{210}Pb). However, the measured ^{210}Pb activity also includes the in situ produced ^{210}Pb generated by ^{226}Ra present in the sediment, assumed to be in equilibrium. In analytical calculations, the atmospheric ^{210}Pb inventory is determined by subtracting the ^{226}Ra concentration from the total measured ^{210}Pb. This concept is only valid if the system is considered closed and the gaseous intermediate decay product, ^{222}Rn, does not escape from the sediment layers. This assumption holds true mainly for systems with rapid sedimentation, where diffusion distances from the pore space are short. In such cases, radon loss is confined to the topmost centimeter, which does not significantly affect dating accuracy. However, in systems where sedimentation is slow and organic matter content is high, the sediment cannot be regarded as a closed system regarding gas escape.

The present study examined this issue in the sediments of Lake Balaton (Hungary) and two Romanian lakes, Lake St. Anna and Red Lake (Gyilkos). These lakes were selected deliberately: Balaton and Saint Anna are both characterized by low sedimentation rates due to their specific depositional mechanisms. Lake Balaton's catchment area contains rocks with high ^{226}Ra content. In contrast, Lake St. Anna receives water solely from precipitation, with no inflows, and its upper 10 cm sediment layer has high water content. Meanwhile, Red Lake features higher sedimentation rates and more compact sediments. Thus, these three sites, with differing sediment characteristics, demonstrate that the observed phenomenon is not local but more widespread. We hypothesize that ^{222}Rn gas escapes from these systems, leading to modified dating results. To test this, we measured radon concentrations in sediment pore water using liquid scintillation counting, taking advantage of its low detection limits.
In Lake Balaton's sediment, radionuclide distributions indicate elevated ^{226}Ra levels ranging between 25 and 55 Bq/kg. If this enhanced ^{226}Ra activity is directly used in dating calculations, the dating horizon would appear at only 10 cm depth, which contradicts other independent sedimentation measurements. The pore water ^{222}Rn concentration profiles confirmed significant radon loss within the upper 30 cm of the sediment column, with values ranging from 3 ± 0.2 to 37 ± 4 Bq/l. This loss is attributed to the high porosity (70-90%) and low sedimentation rate. In Lake St. Anna, radon concentrations ranged from 0.2 ± 0.01 to 3 ± 0.2 Bq/l, corresponding to its lower average ^{226}Ra content (around 25 Bq/kg). For Red Lake, values ranged from 2 ± 0.2 to 28 ± 4 Bq/l, with ^{226}Ra content averaging around 35 Bq/kg—lower than that of Balaton sediments. The radon loss and transport processes can be described by a multivariable differential equation.

Our results clearly show that a significant portion of radon escapes from the upper sediment layers, causing a disequilibrium. Consequently, the sediments of Lake Balaton, Lake St. Anna, and Red Lake cannot be regarded as closed systems concerning radon gas. As a result, this research demonstrates that pore water analysis is crucial for reliable ^{210}Pb dating, especially in systems with slow sedimentation and high porosity accumulated over the last 150 years.