論 文 の 内 容 の 要 旨

論文題目

Iodine-129 in natural archives as historical records of human nuclear activities

（天然アーカイブに記録された人為起源ヨウ素129の分析）

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Iodine-129 ($T_{1/2}$ = 15.7 Ma) is a long-lived fission product that, while produced naturally in the environment, is currently released by human nuclear activities (HNA) such as nuclear bomb testing, nuclear fuel reprocessing (NFR), and nuclear accidents in massive amounts. Its behavior in the environment is conservative and generally known, where it is mainly transported through atmosphere and ocean circulations. Given its long half-life and the current understanding of its sources and behavior in the environment, ^{129}I is considered a promising environmental tracer of HNA-derived radionuclides and the earth processes associated with its transport and cycling. This tracer capability is demonstrated in the analysis of ^{129}I in natural archives, where it can be used to reconstruct HNA influence in locations where archives are taken, to study atmospheric and oceanic circulations that transported ^{129}I to these locations, and to verify age models by using ^{129}I signals of known origins as event markers. However, the current number of ^{129}I records in natural archives is sparse and a lot remains to be studied to fully explore this application of ^{129}I.

In particular, only two ^{129}I coral records exist at present and both are located in the southern hemisphere, in the Solomon and Easter Islands. ^{129}I in these corals reflect nuclear bomb testing and NFR signals, as well as the prevailing ocean circulation patterns in their respective locations. Given that nearly all HNAs were conducted in the northern hemisphere, stronger ^{129}I signals should be observed in coral records from the northern hemisphere. In this regard, the main objective of this dissertation is to construct the ^{129}I time series of two coral cores coming from the northern hemisphere (Philippines). This is done to explore the extent of applications of ^{129}I in (1) reconstructing HNA influence in the Philippines, where currently little is known; (2) tracing atmospheric and oceanic circulations that transport ^{129}I to the study locations; and, (3) as an event marker for establishing/confirming the age models of these coral archives.

To achieve this goal, a method capable of measuring ^{129}I and ^{127}I in about 1-4g of coral samples first needed to be developed. Once achieved, ^{129}I time series of two coral cores from the Pacific Ocean or PO (Baler) and South China Sea or SCS (Parola) sides of the Philippines were constructed and compared to historical records of known HNAs and the existing southern hemisphere ^{129}I coral records. Subsequently, ^{14}C of the corals were analyzed in comparison with the ^{129}I signals as well as other published ^{14}C coral records. ^{14}C in corals are more extensively studied as an event marker and an oceanographic tracer and thus will put context into ^{129}I's capabilities in the following applications. Comparison between ^{129}I and ^{14}C is subsequently done in more detail through a box mixing model capable of simulating radionuclide concentrations in Baler and Parola. The model is also capable of quantitatively demonstrating several mechanisms that explain the observed coral ^{14}C and ^{129}I trends.

Results show that the coral ^{129}I measurement method developed in this study was capable of measuring ^{129}I/^{127}I in 1-4g of samples with errors of about 23% for natural-aged (pre-1950) samples and about 9.6% and 13.5% for anthropogenic-aged (post-1950) samples in Baler and Parola, respectively. This is comparable to the work of Biddulph et al. (2006), which had errors of about 35% and 8% natural and anthropogenic-aged samples, respectively, using 10-30g of corals. This was accomplished by modifying the method of Biddulph et al. (2006) in two aspects: (1) the use of inductively coupled plasma mass spectrometry (ICP-MS) instead of ISE. ICP-MS matrix effect was minimized by using ^{133}Cs internal standard and finding an optimal dilution factor for sample solutions. A 286x dilution was eventually used, which was the largest dilution that resulted to a concentration higher than the method's limit of quantification of 0.27 ppb; and (2) the addition of 0.66 mg of low-ratio iodine carrier in the preparation of target for AMS analysis, instead of using carrier-free method. 0.66 mg of carrier was the smallest amount that forms sufficient amount of AgI for AMS measurement. It was found that error levels increase exponentially with decreasing sample size, as estimated by the equation $y = 3.6338\ln x + 15.609$ of 1/RSD vs. resulting ratio of the AgI precipitate (sample + carrier). Given this, this method can achieve a target error level of about 10% with as little as 1.5 g and 22.5 g for anthropogenic- and natural-age coral samples.

^{129}I in corals from the Philippines was found to record human nuclear activities in good detail. Nearly identical bomb peaks in 1962 were observed in both Baler and Parola (^{129}I/$^{127(stable)}$I ~ 31.5×10^{-12}) – a potential event or age marker, comparable or possibly even better than the coral ^{14}C bomb curve. Nuclear fuel reprocessing and Chernobyl accident signals in 1977, 1980, and 1986 were also observed concurrently in Parola and with a 9 to 11-year lag in Baler. This time lag suggests that ^{129}I reaches the South China Sea and Pacific Ocean sides of the Philippines from the atmosphere and through Pacific Ocean currents, respectively. Lastly, anomalously high $^{129}I/^{127}I$ levels were found in Parola (i.e., 22.8 to 38.9 x 10^{-12}), contrary to the decreasing trend observed in Baler and in published ^{129}I emission records. This finding possibly reveals unknown nuclear activities performed around the South China Sea region. These results demonstrate how ^{129}I in coral samples the Pacific Ocean (Baler) and South China Sea (Parola) sides of the Philippines provide information about how HNAs affected the country, given the different atmospheric and oceanic regimes around the country and the region.

Unlike ^{129}I, which has a variety of sources, ^{14}C only majorly comes from nuclear bomb testing. ^{14}C bomb curves were observed for both Parola and Baler. The onset of the ^{14}C bomb curve in Parola starts at around 1956-1958 and peaks in 1972, similar to ^{14}C of corals located inside the North and South Pacific Gyres (i.e., French Frigate Shoals and Rarotonga, respectively). This is expected because Parola is likewise situated in the middle of the SCS circulation pattern and also because SCS is a semi-enclosed. These factors should enhance the surface water residence time and the atmosphere-to-surface ocean concentration of ^{14}C, resulting to the rapid rise and peaking of $\Delta^{14}C$. The onset of the ^{14}C bomb curve in Baler, meanwhile, also starts at around 1956-1958 but decreases again after 1962. Consequently, a small pre-bomb curve peak is formed in 1962 and the bomb curve of Baler appears to lag that of Parola. Two possible mechanisms are proposed to explain this: (1) “quick input” – a small portion of the bomb-produced ^{14}C with a form other than CO_2 that has a significantly quicker atmospheric lifetime; (2) “SP intrusion” – the intrusion of lower-^{14}C South Pacific waters to the North Equatorial Current (NEC), which supplies the source water in Baler. Features of the Baler ^{14}C record also coincide with phases of both El Niño Southern Oscillation (ENSO; particularly in years 1959, 1966, and 1973/74) and the Pacific Decadal Oscillation (PDO; 1976). Variations of coral ^{14}C, in relation to ENSO and PDO, is possibly explained by the northward shift of the NEC bifurcation latitude during El Niño years and the positive phase of the PDO, which causes increased contribution of northern subtropical waters through the Kuroshio recirculation gyre. Additionally, strengthened upwelling in the Central American region is also considered in the case of the 1976 PDO shift.

Comparison between ^{129}I and ^{14}C is demonstrated through the box mixing model that simulated the two radionuclides in Parola and Baler. Results show that the difference between the atmosphere-to-ocean lifetimes of ^{129}I and ^{14}C is the largest factor for the observed trends in the two nuclides. In particular, the quicker (i.e., 2 years) atmospheric lifetime of ^{129}I causes a sharper, more robust peak that is uniform in timing and shape, regardless of the oceanic processes in the location of the coral. The longer atmospheric lifetime (i.e., 300 years, for this model) of ^{14}C causes the broad peak, which is sensitive to ocean processes, causing variations in the ^{14}C bomb curve’s timing and shape. The plausibility of specific mechanisms for both ^{129}I and ^{14}C was also demonstrated, particular: (1) “SP intrusion” and “quick input”, which causes the small pre-bomb curve peak and the delay of the simulated Baler ^{14}C bomb curve; (2) the 1976 PDO shift, i.e., the increased contribution of northern subtropical waters and the increased upwelling in the Central American region, both due to the northward shift of the NEC bifurcation latitude; and (3) pre-1962 ^{129}I signal discrepancy and the delay of the NFR and Chernobyl ^{129}I signals between Parola and Baler, as a result of different pathways wherein ^{129}I entered the South China Sea directly from the atmosphere, while it traveled from the North Pacific to the Baler site through the prevailing ocean currents.

These results demonstrate that coral ^{129}I records human nuclear activities, particularly nuclear bomb tests, nuclear fuel reprocessing, and nuclear accidents in unprecedented detail. As a coral age marker, it is clearly better than the better-studied coral ^{14}C because, as shown in the measurements and validated with the box mixing model, ^{129}I oceanic peak appears almost concurrently with the time of atmospheric release and it is hardly affected by ocean processes primarily because of its short atmospheric lifetime. ^{14}C oceanic signal, in contrast, varies in timing, shape, and magnitude depending on the oceanic processes in the location of the coral, as a consequence of the long atmospheric lifetime of ^{14}C. Nonetheless, the large difference in atmospheric lifetimes of ^{129}I and ^{14}C lead to distinct oceanographic tracer applications for these radionuclides: coral ^{129}I signals are able to trace the speed of ocean circulation, as demonstrated by the timing discrepancy in NFR and Chernobyl signals between Parola and Baler; coral ^{14}C, on the other hand, are able to reflect large and long-term scale changes in circulation such as SP intrusion and the effects of the ENSO phases and the 1976 PDO shift.

These results will likely interest studies to confirm the ubiquity of observed ^{129}I signals across the northern hemisphere, as well as investigations to ascertain the source and the possible oceanographic tracer application of the high ^{129}I levels in the South China Sea. It could also interest a review of the available coral ^{14}C records, in light of the new mechanisms proposed here, particularly the SP intrusion and the quick input. These findings merit that both ^{129}I and ^{14}C be studied side-by-side in more corals from other locations to fully explore the potential of their applications. We also recommend that higher resolution and more sophisticated modeling studies be conducted for these radionuclides, given that some aspects (e.g., SP intrusion) were not represented accurately in this work. I anticipate that more in-depth studies and representation of the mechanisms proposed in this dissertation would largely contribute to the better understanding of the dynamics and possible applications of these radionuclides in the management of nuclear energy and technology and in the study of earth processes and the environment.