

Thesis Summary

Actinide Elements as Oceanographic Tracers in the Pacific Ocean

(太平洋におけるアクチノイドの海水循環トレーサーとしての利用)

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The ocean circulation is closely linked to our Earth's climate and shifts in its patterns or magnitude will inevitably lead to changes in our living environment, which makes research on ocean circulations extremely important. However, the detailed description of the movement of water masses is difficult to assess using only the typical routine oceanographic parameters (density, salinity, dissolved oxygen, nutrients, etc.). This is why researchers have started in recent decades to use artificial chemical substances (e.g. CFCs) or radionuclides in order to gain additional information on circulation patterns. The advantages of these "tracers" are that their input sources are clear or can be reconstructed and that, in many cases, natural inventories are negligibly small compared to the amounts of anthropogenic production.

The actinide isotopes uranium-236 (^{236}U) and neptunium-237 (^{237}Np), which are the focus of this work, have comparably long half-lives (2.34×10^7 and 2.14×10^6 years, respectively) and allow to describe ocean processes happening on long time scales, like the deep-water circulation in the Pacific Ocean or the global overturning circulation, which are known to take place in a time frame of several hundreds to thousands of years. Furthermore, ^{236}U and ^{237}Np have only been introduced into the world's surface in recent decades, meaning that they are not yet in steady state in seawater, which makes it possible to differentiate between water masses according to their different concentrations of these artificial radionuclides.

However, ^{236}U and ^{237}Np occur only at ultra-trace levels (in the order of attograms per 1 kg) in seawater and advanced measurement equipment, as well as sophisticated chemical separation techniques to reject interfering matrix material are necessary for their detection. The present work first focuses on ^{236}U and describes a co-precipitation method that allows to separate uranium isotopes from seawater with high, stable chemical yields (Chapter 2). Next, a new method for the preparation of target material for the measurement of ^{236}U from seawater samples by accelerator mass spectrometry

(AMS) is developed. The procedure allows to concentrate the uranium isotopes of seawater samples in a tiny amount (100 µg) of iron carrier material. In the AMS measurement, this leads to faster ionization of the sample material, higher ^{238}U currents and a five-fold reduction in measurement times.

In the following, the chemical methods described in Chapter 2 and 3 are applied to 163 seawater samples from different depths at 15 sampling stations in the North, Equatorial and South Pacific Ocean (Chapter 4 and 5). The obtained data on ^{236}U distributions is compared to routine oceanographic parameters (e.g. salinity, temperature), data published on other tracer isotopes, as well as known circulation patterns. Similarities in the latitudinal and depth distribution of ^{236}U concentrations to the relatively well established oceanographic tracer isotope ^{137}Cs become obvious. Also, the ^{236}U concentrations on different depths are in correlation to regional circulation patterns and the movement of water masses, which is proof for the good suitability of ^{236}U as ocean circulation tracer. Additionally, an analysis of the ^{236}U inventory in sediment samples from three sampling stations in the Equatorial and South Pacific Ocean is conducted. The low ^{236}U inventories in the sediment samples, compared to the ^{236}U inventory in the water column above, confirm that this isotope stays in the dissolved phase in seawater and can thus be used as a conservative tracer in the analysed ocean region. In the part of this work that describes the distribution of ^{236}U in the North Pacific Ocean (Chapter 4), the possibility of additional input of ^{236}U isotopes from the accident at the Fukushima Dai-ichi Nuclear Power Plant is shortly discussed, but can be ruled out as contamination source for the sampling points in this study.

In the last chapter of this PhD thesis, the focus is on the use of ^{237}Np and ^{236}U as a tracer pair for oceanographic research. The simultaneous measurement of different actinide isotopes can help in the source determination of a specific contamination. A tracer substance is used to describe the chemical yield of ^{237}Np in the co-precipitation of actinide elements with iron hydroxide from seawater. Afterwards, $\text{K}_2\text{S}_2\text{O}_5$ is added as a reducing reagent in an attempt to improve the chemical yields of the procedure. Finally, a simplification of the method is tried by omitting the heating step. The final method presented in this work provides stable, high chemical yields for both ^{237}Np and uranium isotopes and may help in future towards the application of the isotopic ratio $^{237}\text{Np}/^{236}\text{U}$ as new tracer in ocean research.