# Input and transport of $^{129}$I in the Canadian Arctic: evidence of a Fukushima pulse

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Since the 1950s, nuclear fuel reprocessing and nuclear accidents have released large amounts of anthropogenic $^{129}$I into the atmosphere. This $^{129}$I has spread throughout the northern hemisphere, increasing concentrations above natural fallout and geogenic levels in these regions. Because it is very mobile and readily absorbed by living organisms, $^{129}$I is considered a contaminant of concern, especially in terms of the long term disposal of radioactive waste.

The Arctic has been identified as a location that may be affected by $^{129}$I fallout from nuclear activities in Europe and Asia. A multi-year comparison of $^{129}$I concentrations, ratios and fluxes between Arctic watersheds is a valuable tool to assess the origin and extent of contamination, and to understand $^{129}$I transport dynamics. Water samples from a number of watersheds were collected in the Yukon Territory, Canada in 2010 and 2011. Sampling was conducted in Whitehorse (60°43′00″N) and continued north along the Dempster Highway as far as Tsiigehtchic, Northwest Territory (67°26′26″N). $^{129}$I was extracted from water samples by redox separation, precipitated as AgI, and analyzed using accelerator mass spectrometry (AMS). Total iodine was determined using ICP-MS. Mean concentrations for $^{129}$I northwest Canadian rivers were $16.1 \mathrm{x} 10^6$ atoms/L in 2010 and $66.8 \mathrm{x} 10^6$ atoms/L for 2011. These values significantly exceed previously published values for waters containing only geogenic or cosmogenic $^{129}$I. A comparison of $^{129}$I flux over the two years for a subsection of the sampling area also shows a 10-fold increase in the fallout of $^{129}$I (atoms/$m^2$/yr) in the watersheds between 2010 and 2011. The source of $^{129}$I in the Arctic is mainly from nuclear fuel reprocessing releases which are transported atmospherically through a cycle of deposition and revolatilization to remote regions throughout the northern hemisphere. The substantial increase in $^{129}$I fallout in these watersheds from 2010 to 2011 is consistent with elevated levels of $^{131}$I measured on the western coast of North America in the days following releases from the Fukushima Daiichi nuclear power plant in March 2011.

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# U isotopes in marine calcareous algae: a paleo-redox proxy?

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Significant variability in the uranium isotopic composition of natural samples [1] is largely due to isotope fractionation during redox transformations associated with uranium deposition in reducing sediments. This suggests that the uranium isotopic composition of seawater may be determined by the redox state of the global ocean. Limited evidence indicates that scleractinian corals record the uranium isotopic composition of seawater [1]. Thus, changes in the uranium isotopic composition of carbonates through time may reflect changes in the redox conditions of the global oceans. More work is needed, however, to confirm that uranium values of biogenic carbonate sediments in general reflect seawater values, irrespective of the biomineralization pathways that led to the formation of the calcium carbonate. In particular, it will be critical to establish that carbonates faithfully record uranium values of seawater without fractionation. Here, we report uranium isotope values from a range of different marine algae from the Bahamas to further test the potential of calcium carbonates as a paleo redox proxy.

It has long been known that calcareous algae incorporate different amounts of uranium [2] and have different biomineralization pathways that affect the $\delta^{13}$C and $\delta^{18}$O of the precipitated carbonate [3]. These difference might also lead to a fractionation of uranium isotopes during the formation of calcium carbonate in different algae. However, our results indicate that fractionation of uranium during biomineralization of algae does in fact not take place. Despite difference in uranium concentrations (ranging from ~1ppm to ~2.5ppm) and different biomineralization pathways (*Acetabularia* sp., *Rhipocephalus* sp., *Halimeda incrassata*, and *Penicillus capitatus*), the uranium isotopic compositions in these various algae are the same and reflect seawater values ($\delta^{238}$U ~-0.40‰). The $\delta^{238}$U values of the calcareous green algae are indistinguishable from $\delta^{238}$U values of scleractinian corals (*Diploria* sp, *Siderastrea* sp., and *Porites* sp.) collected in the same area. Futhermore, $\delta^{234}$U values of all samples are indistinguishable from seawater values.

Our results show that biominearlization pathways do not lead to fractionation during the uptake of uranium isotopes and calcareous algae are thus excellent candidates to serve as paleo-redox proxies. Since calcareous algae are important producers of fine sediment in carbonate settings, the lack of fractionation within the algal material further implies that bulk sediment analyses of carbonate rocks dominated by algal material as a tracer of paleo-redox are possible.

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