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Environmental $^{129}$I: level, distribution and source in Northwestern China

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$^{129}$I is a long-lived radionuclide with a long half-life of 15.7 Ma. $^{129}$I in the present environment mainly comes from human activities including nuclear weapons testing, nuclear fuel reprocessing, nuclear accidents and operation of nuclear reactors. Iodine is a volatile element, and chemically active, the radioactive isotopes of iodine released can easily spread in the environment, migrate in the ecosystem and enrich in the thyroid gland in the human body. Due to long half-life, $^{129}$I can be used as a tracer to investigate nuclear environment safety, seawater exchange and transport, stable iodine geochemical cycle, etc. In this work, surface soil samples (0-5 cm) collected in Northwest China were analysed for $^{127}$I and $^{129}$I using an effective chemical separation combined with a high sensitivity AMS measurement, in order to investigate $^{129}$I level and distribution in Northwest China, explore its sources in this region. The data is also useful for establishment $^{129}$I environmental background in Northwest China, and investigation on the impact of early human nuclear activities on the environment in the region.

The collected soil samples were dried, ground and sieve through a 200 mesh sieve. About 5 g ground soil samples was taken to a quartz boat, 1.0 kBq $^{125}$I tracer was spiked for measurement of chemical yield. The boat with sample was put to a quartz working tube in a tube furnace for separation of iodine using combustion. The temperature of the furnace was gradually increased to 800°C and kept for 1.5 hours under oxygen gas flow. The off gas from the working tube passed through a bubbler filled with 0.5 M NaOH-0.02 M NaHSO$_3$, liberated iodine from the sample was trapped in the solution in the bubbler. The entire combustion took about 3 hours. 3 ml of trap solution was taken to a plastic tube and measured using a gamma detector for $^{125}$I, which was compared with the $^{125}$I standard (the same amount of $^{125}$I spike solution and diluted to 3 ml using the same trapping solution) for measurement of chemical yield of iodine during combustion. Chemical yield of more than 97% were obtained for soil samples. After measurement of $^{125}$I, the solution is combined to remained trap solution. 1.0 ml trapped solution was taken and diluted 10 times using deionized water for measurement of $^{127}$I using ICP-MS. To the remained solution, NaHSO$_3$ and 0.5 ml of $^{127}$I carrier solution with a concentration of 2.0 mg/ml (prepared from a $^{129}$I free iodine provided by Woodward company, USA, with a measured $^{129}$/127I ratio less than 5×10^{-14}) were added, and pH was adjusted to 1-2 using HNO$_3$. After mixed, 1 ml of 1.0 mol/L AgNO$_3$ was added for precipitate iodine as AgI, which was separated by centrifuge. After dried, AgI precipitated was ground and mixed with niobium powder in a mass ratio of 1:5, which was then pressed in copper target holder. $^{129}$/127I atomic ratio was measured using 3 MV accelerator mass spectrometry in Xi’an AMS Center. $^{5+}$ ion was selected for $^{129}$I measurement. Procedure blanks were prepared using the same procedure as samples, the measured $^{129}$/127I in the blanks are (1-2)×10^{-13}, which is 2-3 orders of magnitude lower than that in samples.

More than 200 surface soil samples was analysed, the concentrations of iodine isotope show a significant variation. The concentrations of $^{127}$I in Northwest China are 0.43 to 16.8µg/g (dry mass), with an average of 3.65µg/g, which agree with the literature values (0.5~40 µg/g); $^{129}$I concentration are 1.38×10^{6} to 2.62×10^{10} atmos/g, with an average of 4.29×10^{6}atmos/g. There is a hotspot, where high $^{129}$I level is observed, with $^{129}$I concentrations higher than 10^{9} atoms/g, this might be attributed to the early nuclear activities in this regional; $^{129}$/127I atomic ratios in all samples range from 9.1×10^{-11} to 7.38×10^{-6}, with an average of 7.47×10^{-8},
which is higher than pre-nuclear level of $10^{12}$ by 1-4 orders of magnitude. These data indicating that the surface environment in Northwest region was significantly influenced by human nuclear activities.

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