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IDMS ANALYSIS OF BLANK SWIPE SAMPLES FOR URANIUM QUANTITY AND ISOTOPIC COMPOSITION

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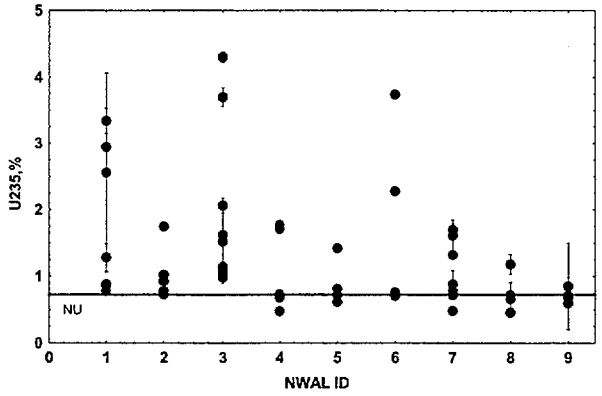
Since 1996 the IAEA has started routine implementation of environmental sampling. During the last 5 years more than 1700 swipe samples were collected and analyzed in the Network of Analytical Laboratories (NWAL).

One sensitive point of analyzing environmental samples is evidence of the presence of enriched U. The U content on swipes is extremely low and therefore there is a relatively high probability of a false positive, e.g. small contamination or a measurement bias. In order to avoid and/or control this the IAEA systematically sends to the laboratories blind blank QC samples. In particular more than 50 blank samples were analyzed during the last two years. A preliminary analysis of blank swipes showed the swipe material itself contains up to 10 ng of NU per swipe. However, about 50% of blind blank swipes analyzed show the presence of enriched uranium (see the Figure). A source of this bias has to be clarified and excluded.



This paper presents the results of modeling of IDMS analysis for quantity and isotopic composition of uranium in order to identify the possible contribution of different factors to the final measurement uncertainty. This modeling was carried out based on the IAEA Clean Laboratory measurement data and simulation technique.

As a rule the laboratories measured only a sample spiked with ^{233}U and isotope abundance of ^{234}U, ^{235}U, ^{236}U for the sample are calculated (stripped) from the isotopic ratios measured for the mixture spike plus sample. It is assumed that the sample does not contain ^{233}U. In this case, the magnitude of the spike to sample ratio may have a large contribution to the uncertainty of the stripped isotope abundances. Using the raw measurement data reported by the Clean Laboratory, it was shown that for spike to sample ratios higher than *ten* the relative uncertainties of stripped ^{234}U and ^{235}U abundance can reach hundreds percent. Data reported by some other laboratories show a similar tendency. However using a purer spike (with a lower content of ^{234}U and ^{235}U) leads to a decrease in the uncertainty for stripped ^{234}U and ^{235}U abundance in the sample. Using the raw isotopic ratios measured in the Clean Laboratory, the regression equations for SD versus the ratios measured were obtained. These equations were used for a simulation to identify the contribution of various factors to the final analysis uncertainty of U content and isotopics.

The RSD values of the simulated and experimental U mass are generally consistent and do not exceed 10% even when the spike to sample ratio equals to 1000. This means that any possible overestimation of U quantity in a blank sample is not a result of measurement uncertainties or a non-optimal spike to sample ratio.

A positive bias in the U mass can appear only if the ^{233}U mass in the spike has a negative bias. This is possible if equilibrium between the spike and the sample is not reached and the U yield for the sample is higher than for the spike.

The simulation results (as well as experimental data) show that the relative uncertainty of the stripped ^{235}U abundance goes up with increasing the spike to sample ratio. For ^{235}U this uncertainty becomes more than 50% (in case of CL measurements) if the spike to sample ratio is higher than 100. The comparison of simulated and experimental data shows the scattering of experimental data is significantly higher than simulation results. This allows concluding that some experimental results are positively or negatively biased due to "non-statistical" factors such as mass fractionation, isobaric interference, and/or small contamination. One can assume reducing the spike to sample ratio reduces the contribution of these "non-statistical" factors.

If the spike to sample ratio is higher than *one*, the relative uncertainty of the stripped ^{234}U abundance can reach 1000%. In this case, one can expect negative values of ^{234}U abundance. The simulated and experimental data are consistent and show that the scattering of the stripped ^{234}U abundance is mainly caused by statistical factors.

A bias in the stripped isotopic composition of the sample can only appear if the isotopic ratios measured in the mixture and/or certified in the spike are biased. This can be due to a small contamination with enriched uranium. Another source of a positive bias might be isobaric interference, also some form of contamination. The simulation was provided for various possible bias values (see the Figure). It was shown the contribution of this bias to the stripped ^{235}U abundance in the sample is a relatively large for the spike to sample ratio higher than 50 and decreases with decreasing the spike to sample ratio.