# Evaluation of Methyl-mercury production and decomposition by using $^{197}$Hg radiotracer produced out of mercury enriched in $^{196}$Hg isotope

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## i. Background

The project approved proposed the implementation of mercury (Hg) laboratory experiments to study the transformation of inorganic mercury ($Hg^{2+}$) to organic species, reproducing natural conditions. The main analytical tool proposed was the use of the short lived Hg isotope $^{197}$Hg ($T_{½}$ = 64.14 h) in tracing the transformation processes, being this radiotracer produced in the nuclear research reactors of the Jožef Stefan Institute, Ljubljana, Slovenia (TRIGA Mark II reactor 250 kW), and Centro Atómico Bariloche, Argentina (RA-6 MTR reactor, 500 kW), which showed an excellent performance in environmental studies developed in previous works.

Before the fellowship start, the chance to performed laboratory experiments in Hg retention in Flue Gas Desulphurisation (FGD) systems in coal power plants using the $^{197}$Hg radiotracer came out. The Hg emissions generated in the operation of coal power plants are the main anthropogenic atmospheric source at present, whereas the understanding of the chemical processes undergoing in the Hg retention in the FGD systems are key in the development of efficient retention systems in order to minimize the global Hg gaseous emissions. Therefore, the focus of the work to be developed in the project moved to this topic. Also, experiments in the preservation of Hg in natural waters were performed due to the relevance of this topic in environmental studies and the advantages provided by using the $^{197}$Hg radiotracer.

## ii. Research performed

a. The project started with preliminary experiments in the flue gases pilot plant at Institute Jozef Stefan, using the radiotracer $^{197}$Hg. The pilot plant at the Reaktor Centre, Institute Jožef Stefan, is an experimental arrange simulating chemical processes of the flue gasses purification systems, particularly Hg retention, consisting, basically, of a glass tube containing water with air bubbling. The air supply system includes a $Hg^0$ (vapour) source, by heating liquid $Hg^0$. The water circulates forced by a pump through a chemical cell, that allow pH control, addition of the reduction agent $SO_2$ and other chemicals, and measurement of water temperature and REDOX potential. After bubbling, the air flows out of the system through a plastic tube that allows different measurement on the out coming gasses that gives information about the chemical processes within the bubbling column.

The experiments tested the potential of the $^{197}Hg$ radiotracer produced at the TRIGA Mark II research reactor to assess the $Hg^{2+}$ reduction capacity of the system by adding $^{197}Hg^{2+}$ in the chemical cell and evaluating the tracer activity in the outgoing air flow and in the bubbling water, in time trend sequences.

b. A second stage of the project consisted in the evaluation of the $Hg^{2+}$ retention capacity of Gypsum (calcium sulfate dihydrate; $CaSO_4 \cdot 2H_2O$) small size particles in aqueous suspension by using the $^{197}Hg$ radiotracer, testing the retention dependence with the suspension pH. The Gypsum is a solid material used in the FGD systems which retains Hg together with the desulphurisation process.

c. The experimental series on Hg retention in FGD systems were completed by testing the $Hg^{2+}$ behaviour in an aqueous solution under different conditions in a chemical reactor providing gas bubbling in the solution, pH and REDOX control, and $Hg^0$ collection in a $KMnO_4$ trap. The $Hg^{2+}$ traced with $^{197}Hg$ is spiked in the solution, measuring the activity in the water solution, both in total and filtered by 0.45 μm pore size to evaluate $^{197}Hg$ association to any precipitate formed during the experiment, and in the $KMnO_4$ solution along the time evolution of the chemical system. The composition of the solution, the gas bubbling in the solution, pH and REDOX were changed in different experimental runs to study the behaviour of the Hg in this system under different conditions.

d. A complementary study testing the preservation of Hg in natural waters was the last experimental run performed. The experiments consisted in the evaluation of different conditions for the preservation of water samples to analyze Hg contents in natural levels. Water samples from different sources (lake, river, marine, coastal lagoon, and rain water) were spiked with $Hg^{2+}$ traced with $^{197}Hg$ and stored in different conditions, determining the time trend of the $^{197}Hg$ specific activity in order to evaluate $Hg^{2+}$ losses from the water solution. The traced $Hg^{2+}$ spiked was determined in order to obtain similar concentrations to those expected for the analysis of natural water, ranging in 10 ng $L^{-1}$.

### iii. Return phase

The return phase carried out the study of $Hg^{2+}$ transformations to organic compounds in lacustrine waters and further bioaccumulation in plankton, assessing the incidence of the dissolved organic carbon (DOC) in these processes. The waters studied belong to the oligotrophic lacustrine systems of the Patagonia. The experiments consisted in testing traced $Hg^{2+}$ transformations to organic Hg in waters from different lakes comprised in the Nahuel Huapi National Park, Patagonia, Argentina, with different DOC composition. The waters are incubated in time trends in laboratory reproducing natural conditions, also including different plankton cultures of native species to study the Hg bioaccumulation in the base of the lacustrine food web.