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Romanian-Swiss Research Programme:

Scientific reports of a Joint Research Projects

1. General Information

Project number: **IZERZ0 – 142228/1**

Project title: Mercury threat in industrially impacted surface water bodies in Romania - integrated approach (MERCURO)

Reporting period: 01.01.2014-31.12.2014

Swiss PI (name and affiliation):
Vera I. Slaveykova-Startcheva, University of Geneva

Romanian PI (name and affiliation):
Gheorghe Oaie, GeoEcoMar - National Institute for Research and Dev. of Marine Geology and Geoecology

Researchers financed by the project in Switzerland

Name and affiliation	Sex	Date of birth	Position (with employment percentage)	Other
Dranguet Perrine/ University of Geneva Institute Forel	F	18.04.1989	PhD student (100%)	

Researchers financed by the project in Romania

Name and affiliation	Sex	Date of birth	Position (with employment percentage)	Other
Oaie Gheorghe / GeoEcoMar	M	06.07.1956	General Director of GeoEcoMar / Senior researcher (Full time)	Sedimentology
Ungureanu Gh. Viorel / GeoEcoMar	M	09.04.1964	Senior researcher / Technical responsible (Part time)	Sedimentology
Secrieru Dan / GeoEcoMar	M	01.03.1950	Senior researcher (Full time)	Geochemistry
Voicaru Cristiana / GeoEcoMar	F	01.06.1968	Researcher / Administrativ responsible (Full time)	Hydrotechnical
Catianis Irina / GeoEcoMar	F	14.03.1969	Researcher (Full time)	Geology/ Ecotoxicology
Vasiliu Dan / GeoEcoMar	M	23.08.1970	Research (Full time)	Chemistry
Ungureanu Constantin / GeoEcoMar	M	01.10.1954	Research (Full time)	Sedimentology
Scrieciu Albert / GeoEcoMar	M	21.01.1986	Research (Full time)	Geology
Grosu Dumitru / GeoEcoMar	M	08.08.1959	Research (Full time)	Chemistry

2. Introduction

The main objective of the MERCURO project is to assess the Hg threat in industrially impacted surface water bodies in Romania. The project focuses on the one of the most strongly impacted surface water body in Romania - River Olt basin with specific emphasis on the following key issues:

- Performance of an Hg survey and estimation the pollution extent in water and sediments;
- Determination of the transport, dispersion and speciation of Hg in water column and sediments;
- Assessment of the bioaccumulation and effect of Hg to different organisms with emphasis on the primary producers in particular microalgae, periphyton and macrophytes;
- Evaluation of food chain transfer and possible risks for the human beings;

Strengthening the capacity, knowledge transfer, improving integration of scientists in the international network as well as developing “good practices” for impact assessment of mercury contamination are other expected outcomes of the project.

3. Scientific results

3.1. Transport and dispersion of Hg in the Olt River reservoirs (GeoEcoMar and UniGe)

3.1.1. Measurements of Hg and MMHg in surface sediments (GeoEcoMar and UniGe)

Grab sediment samples taken in 2013 were analyzed by a geochemist from Geoecomar in the laboratory of Institut F.-A. Forel during a research and training stage in November 2014. Data are still under evaluation, but a few conclusions can be drawn already:

- Although the industrial activity of Oltchim was diminished during the last 2 years, due to economic reasons, the sediment is still contaminated with Hg in Babeni reservoir (1.43-5.23 µg/g). The two tributaries - Bistrita and Topolog - build up fast growing deltas by bringing clean sediments, from the Hg standpoint, in the reservoir;
- Grab sediment samples from Ionesti reservoir, the next one in the Olt cascade of reservoirs, show lesser Hg contamination, with an average on 30 samples of 0.52 µg/g. This value is not very representative for the whole situation since many samples have been taken from the influence zone of the two tributaries that flow into the reservoir (Luncavat and Ursana) and show less or no Hg high concentrations. However, 20 samples present concentrations of Hg in the range 0.55 - 1.09 µg/g;
- Zavideni reservoir, situated downstream of Ionesti, is also contaminated with Hg. Four grab samples taken along a N-S transect show Hg concentrations of 0.51-1.33 µg/g, with an average of 0.87 µg/g. The higher contamination of Zavideni could be explained by the absence of clean tributaries;

It appears that suspensions rich in Hg are transported downstream over long distances creating potential threats to biota and humans.

3.1.2. Contamination history in the cascade reservoirs (GeoEcoMar and UniGe)

The cores retrieved during the 2013 field campaign have been analyzed for sedimentological parameters, radiometric dating (^{137}Cs), and total Hg contents. Results have been compared to cores retrieved in the 2007 field campaign to complement the data set (Garcia et al. 2009). The Figure 2 presents the downstream evolutions of Hg concentrations, specific flux and Hg specific stock in sediment deposited between 1986 and present. Results from the Valcea reservoir correspond to the reference situation, this reservoir being situated upstream the industrial platform Hg-contaminated discharges. The results demonstrated significant propagation of Hg contamination along the four cascade reservoirs. A strong contamination is still observed in the Dragasani reservoir, maximum and mean concentrations being 60 and 23 times higher in the latter than in the reference reservoir (Valcea) (Fig. 2A). The 1987 contamination event observed in Babeni is recorded in the downstream reservoirs, with a decrease of the maximum concentrations from Babeni (45 mg/kg) to Dragasani (7 mg/kg). However, the mean concentrations in the sediment of the period 1986-2013 show only a factor 2 decrease between these reservoirs (4.4 to 2.1 mg/kg, Fig. 1A). The specific fluxes of Hg show a factor 14 decrease from Babeni to Ionesti reservoirs, and again a factor 2 decrease in downstream reservoirs (Fig. 1B). The very high maximum flux at Babeni is the result of high sediment accumulation rate (few centimeters per year) and high Hg sediment contents. Finally, the specific stocks (µg/cm^2) of Hg deposited during the same period within the sediments decrease from 317 µg/cm^2 in Babeni reservoir to 37 µg/cm^2 in Dragasani reservoir. These results indicate that a significant portion of Hg is not retained in Babeni reservoir, and transported downstream. It is particularly worrying to observe that the mean Hg concentrations in the cascade reservoir sediment only slowly decrease with distance from the Hg source. In term of ecotoxicity, the Dragasani reservoir sediments, 30 km downstream the contaminated effluent mouth, present Hg concentrations twice the probable effect concentration (PEC). Cores of sediments have been sampled in each reservoir to link microbial diversity with Hg concentration and speciation. DNA and RNA have been extracted and the occurrence of the gene *hgcA* will be measured by illumina sequencing in collaboration with Dr A. Bravo (Upsalla University).

3.1.3. Numerically model sediment and Hg dispersion in the reservoir in various flow conditions (GeoEcoMar)

The first run of the hydrodynamical model has been completed. However, the model requires refinements with more flow data for Olt tributaries and for the water discharges that flow through the dams.

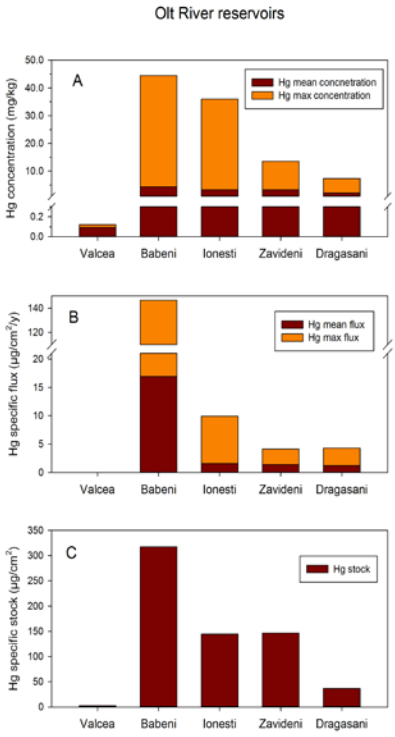

Figure 1: Downstream evolution of A) Hg concentrations, B) Hg specific fluxes, and C) Hg specific stocks in cascade reservoir sediments. There are no flux data in Valcea reservoir because of the lack of sediment core dating.

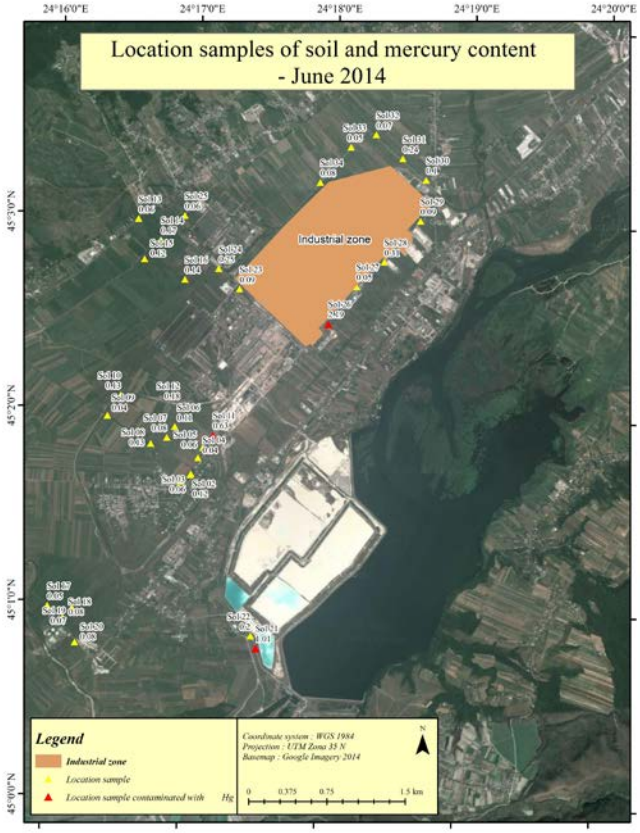

Figure 2: Location of different sites sampled during the campaign in June 2016 and the concentrations of total Hg in soil.

3.1.4. Soil sampling and Hg measurement

The main objective of the soil sampling is to identify potential Hg pollution around the industrial platform Rm. Valcea. Taking into account the long history of existence of the industrial platform and the almost complete lack of environmental concern in the past decades, especially during the communist times, some degree of soil Hg pollution is to be expected. A number of 33 soil samples have been analyzed and the results compared with two samples taken from the banks of the reference lake of Valcea. Since the access to the industrial complex was denied, all locations of soil samples are distributed around the platform at distances situated between 20 and 500 m from the fence. Other samples were taken in the vicinity of the Oltchim/USG treatement plant and near the USG settling ponds (Fig. 2). Total Hg has been determined in the laboratory of Institut F.-A. Forel using an AMA 254 Hg analyzer. The results have shown that only in three locations the Hg concentration is higher than the allowance of the Romanian Standard (0.3 µg/g). The highest value, of 2.19 µg/g was found just in front of Oltchim. Other 2 samples with high Hg were located in the SW end of the platform (0.63 mg/g) and near the USG settling ponds (1.01 mg/g).

3.2. Mercury partitioning and water speciation (UniGe)

During second year campaign surface waters (1 m depth) of Valcea (Vl), Govora, Babeni 1 and 2 (Bb1 and Bb2), Ionesti (Io), Zavideni (Zd) and Dragasani (Dg) reservoirs were collected in order to measure several physico-chemical parameters as well as dissolved Hg concentrations (Table 1 at the end of the document). Values obtained for pH varied between 7.6 and 10.7. That latter value was measured in

the Govora River and was much higher than pH measured in all the studied reservoirs. The measured dissolved organic carbon (DOC) concentration was also very high in the Govora River (58.4 ± 0.02 mg/L) compare to the values measured in the reservoirs (2.7 to 3.8 mg/L). Concentrations of Na^+ and Cl^- were also much higher in the Govora River compare to Valcea Water with an increase of about 13 and 288 times, respectively, and were observed to impact downstream reservoirs. Water quality is thus strongly impacted by the chemical platform despite the presence of a wastewater treatment plant. Beside anions and cations, dissolved Hg concentration (<0.45 µm) was also much higher (about 7 times) in the Govora River than in the reservoirs. However this latter was 10 times lower than the values measured during the first campaign in 2013, suggesting that the chloro-alkali plant may have decreased its Hg release in the water. The speciation of Hg in different reservoirs was modelled by using WHAM model taking into account the pH, concentrations of cations and anions, DOC, and dissolved Hg. For all sampling sites 99.9 % of the inorganic Hg was bound to humic and fulvic acids (Fig. 3). In all reservoirs, the fractions of Hg not bound to organic matter were $Hg(OH)_2^0$, $HgOHCl^0$ and $HgCl_2$ except in the Govora River, $Hg(OH)CO_3^-$ prevailed.

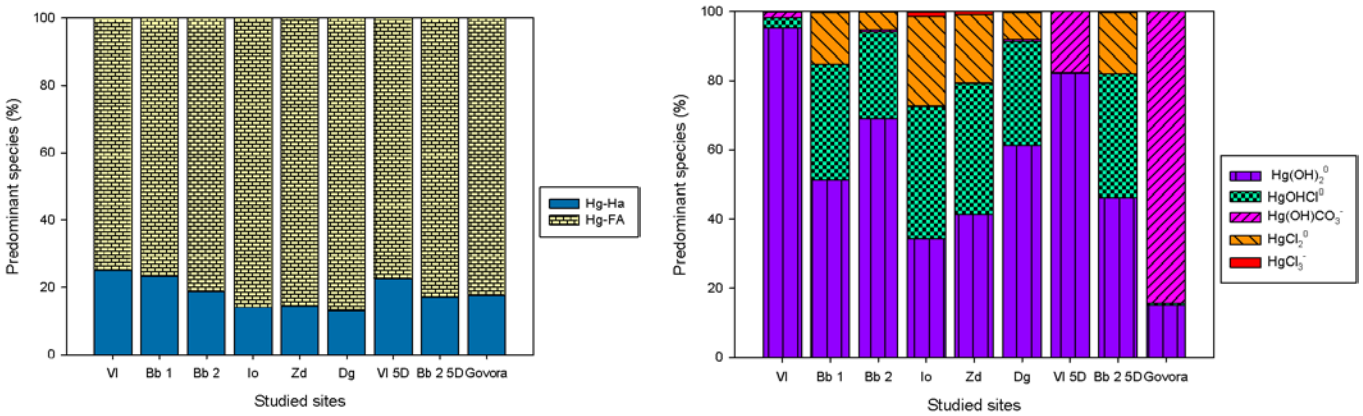

Figure 3: (A) Hg complexation by DOM, (B) inorganic Hg complexes.

3.3. Hg accumulation and effects to aquatic primary producers (UniGe)

3.3.1 Hg accumulation in algae and transcriptome analysis (UniGe)

Green alga *Chlamydomonas reinhardtii* grown in the laboratory were exposed using **S**tirred **U**nderwater **B**iouptake **S**ystem along the Olt River i.e. in control site (Valcea), in two Hg-impacted sites (Babeni 1 close to the mouth of the Govora River and Babeni 2 near the dam). Total and intracellular (e.g. cysteine washed) Hg was determined to assess Hg uptake by algae. No difference was observed between total and intracellular Hg concentration in algae, as well as between algae exposed to control site and sites impacted by Hg (Fig. 4). Nevertheless the transcriptomic analyses are underway in the Genomic platform of the University of Geneva to explore the link between the accumulation of Hg and transcriptomic response.

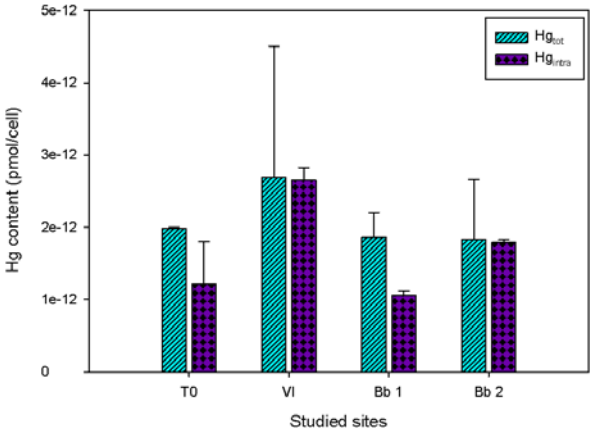

Figure 4: Total and intracellular Hg concentrations in *C. reinhardtii* exposed for 2 hours in Valcea (Vl) and Babeni (Ba).

3.3.2. Hg accumulation in periphyton (UniGe)

To complement the experiments with in-situ grown and translocated periphyton communities, during the second year of the project, short- and long-term exposure experiments under well controlled l conditions in microcosm were performed. To this end, periphyton cultured in Lake Geneva water was spiked at 200 pM Hg, a concentration close to the one measured in the water form Govora River. Periphyton was exposed to Hg during 10 min, 30 min, 1 h, 4 h, 6 h , 8 h and 24 h. At each step time, several points were examined such as Hg bioaccumulation, chlorophyll content and taxonomic composition. Moreover the experiment was performed with two communities, one aged of 85 d and the second one aged of 127 days. The results demonstrated that inorganic Hg contents in periphyton increased by about 2 times when exposed to Hg up to 10 h and no difference was observed between both communities (Fig. 5). However, after 24 h total Hg content of 886 ± 179 pmol/g dry weight for the younger periphyton and 1528 ± 73 pmol/g dry weight for the older periphyton were determined. However intracellular Hg content remained comparable in both periphyton. Proportion of Hg strongly attached to periphyton represented 48 % and 34 % for the biofilm aged of 85 days and of 127 days, respectively.

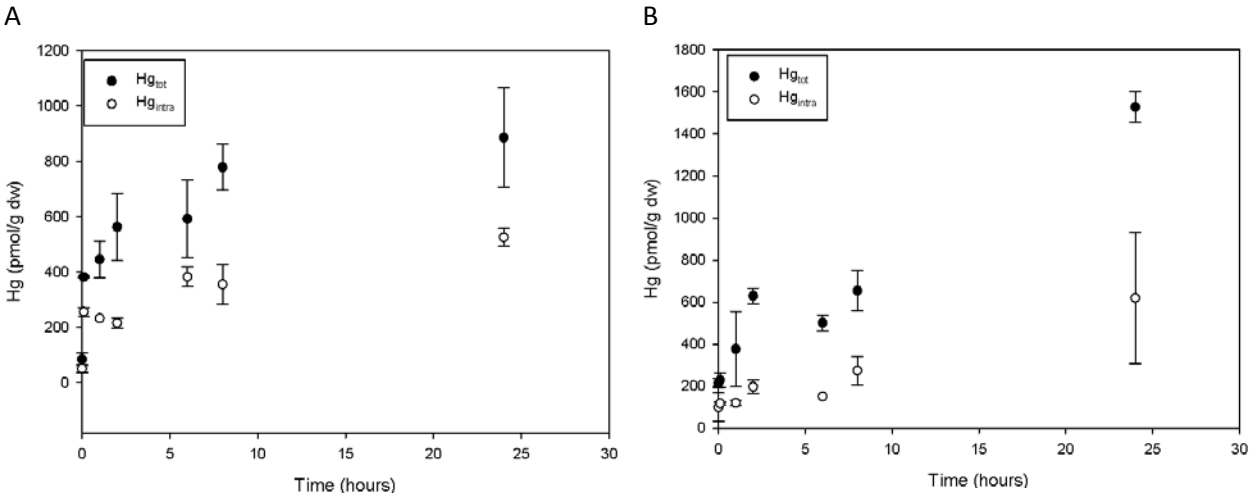

Figure 5: Concentration of total and intracellular inorganic Hg measured in periphyton exposed to 200 pM of Hg as function as exposure time (A) periphyton aged 85 days and (B) aged 127 days.

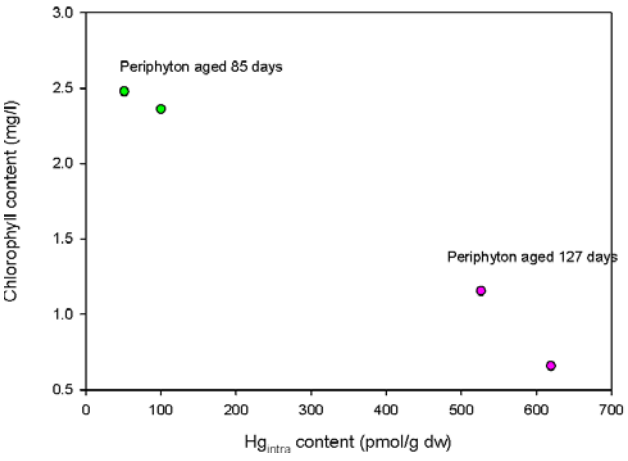

Figure 6: Chlorophyll content in periphyton before and after Hg exposure for both periphyton as function as intracellular Hg content.

Chlorophyll content decreased with the increase of Hg content in periphyton, suggesting a significant impact of Hg on the photosynthetic organisms (Fig. 6). Pyrosequencing method was used to study species composition of the studied periphytic communities before and after Hg exposure. To that end, three pairs of primers were used to distinguish bacteria, phototroph and fungi, which were 1053F/1319R, D512/D978 and ITS1/ITS4, respectively. For both periphyton, taxonomic composition of bacteria and algae changed with Hg exposure for 24 h (Fig. 7). No conclusion can be inferred for fungal species composition because low fungal hits were identified.

The proportion of phylum were similar for both periphyton with *Proteobacteria* as predominant bacterial phylum. However, cyanobacteria increased in the older periphyton exposed to Hg for 24 hours (Fig. 7). *Gemmatimonadetes* phylum was present in low proportion at the beginning of the exposure experiment but disappeared with Hg exposure, demonstrating very high sensitivity of this microorganism to mercury.

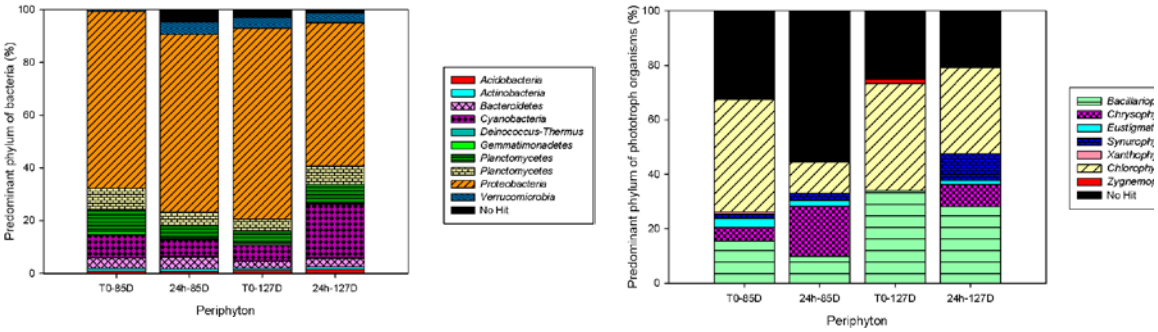

Figure 7: Predominant phylum of bacteria (A) and phototroph (B) determined by pyrosequencing of periphyton aged 85 and 127 days at the beginning of the experiment and after 24 hours exposure.

According to the results obtained with phototroph organisms, the younger periphyton was more heterogeneous than the old periphyton at the beginning of the exposure experiments (Figure 5). The two main phyla were *Bacillariophyta* and *Chlorophyta*. A lot of organisms were not identified, especially for the young periphyton at 24h in which only 40% of the phylum was known.
At the end of the experiment, taxonomic community changed with decrease of phyla such as *Bacillariophyta* and *Chlorophyta* and increase of other phyla such as *Synurophyceae* and *Chrysophyceae*. Mercury induced a modification of the periphytic community after only 24 h and the composition of phototroph organisms was degraded. These results are in agreement with the chlorophyll measurements.

3.3.4. Comparison with macrophytes exposed at the same sites (UniGe)

Shoots of *Elodea nuttallii* grown in the laboratory were exposed 2 hours in the field in similar sampling sites than *C. reinhardtii*. Additional shoots of Elodea were exposed for 5 days in order to assess if differences could be measured in Hg bioaccumulation. To that end, total and intracellular (e.g cysteine and EDTA washed) Hg were measured as well as methylmercury content and transcriptomic analysis were performed. Mercury bioaccumulation was found to not follow Hg concentrations measured in water. Indeed Hg concentration was higher in Babeni 2 (21.53 ± 0.27 pM) than in other reservoirs (1.28 ± 0.14 pM and 2.36 ± 0.23 pM in Valcea and Babeni site 1, respectively). Moreover bioaccumulation of total Hg after 5 days increased in Babeni 2 but decreased in Valcea.

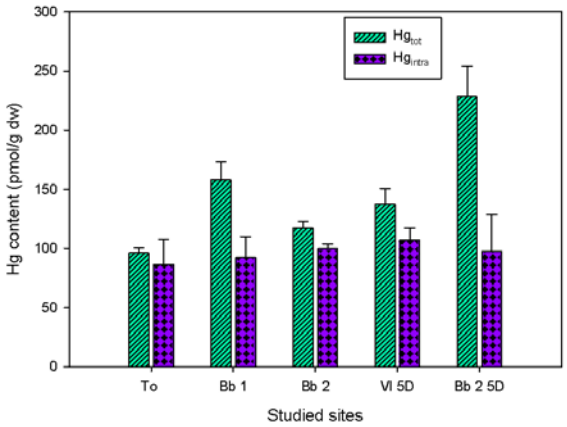

Figure 8: Total and intracellular Hg concentrations in shoots of *Elodea nuttallii* exposed at different sites during 2 hours in the Olt River (Romania).

C. reinhardtii, transcriptomic response are currently analysed by the genomic platform of the University of Geneva.

3.4. Ecotoxicological assessment of sediments by batteries of microbiotests (Unige and GeoEcoMar)

Two ecotoxicological tests were performed to evaluate the quality of the sediment reservoirs. These experiments were carried out by Irina Catianis, a researcher at GeoEcoMar in close collaboration with Dr Séverine Le Faucheur (UniGe) in the framework of a knowledge transfer in ecotoxicology. Sediments collected in Valcea, Babeni, Ionesti, Dragasani and Zavidani reservoirs in September 2014 were analyzed for their physical and chemical characteristics (pH, Loss on Ignition - LOI, grain size and Hg concentration) (Table 2). Sediment toxicity was evaluated using the insect larvae *Chironomus riparius* and the ostracod *Heterocypris incongruens* whereas Hg bioavailability was assessed by measuring Hg accumulation (after cysteine washing) by *C. riparius* at the end of the exposure experiment.

Studied sediments had similar pH and were mainly composed of silt (values ranging from 84.4% to 92.9%) (Table 2). However their organic matter content (expressed as percentage of total sediment dry weight) slightly differed with Valcea reservoir which had lower content (7.8 ± 0.2%) than the reservoirs located downstream the chemical platform (LOI values varying between 8.2 ± 0.2% and 9.9 ± 0.3%). Sediments collected in the control reservoir (Valcea - 0.13 ± 0.01 mg/kg) contained less Hg than sediments located downstream the Govora River (for example 0.51± 0.04 mg/kg in Babeni reservoir), suggesting a degradation of the sediment quality due to the industrial platform effluents release.

Table 2: Physicochemical characteristics (pH, size class, Loss on ignition – LOI) and Hg concentrations in sediments collected in the Olt River reservoirs in September 2014. * pH of the overlaying water

Reservoir	pH*	Size classes (% of particle volume)	LOI (% of sediment dry weight)	[Hg] (mg/kg)
Valcea	8.1	1.5 % clay; 92.9 % silt 5.6 % fine coarse sand	7.8 ± 0.2	0.13 ± 0.01
Babeni	8.3	3.3 % clay; 92.2% silt 4.5 % fine coarse sand	8.2 ± 0.2	0.51 ± 0.04
Ionesti	8.2	2.4 % clay; 84.4% silt 13.2 % fine coarse sand	8.5 ± 0.2	0.24 ± 0.01
Zavideni	8.1	6.1 % clay; 91.5% silt 2.3 % fine coarse sand	9.9 ± 0.3	0.46 ± 0.02
Dragasani	8.2	5 % clay; 90.1% silt 9.9 % fine coarse sand	8.7 ± 0.2	0.51 ± 0.02

Prior sediment exposure, the midge larvae were grown up to their fourth instar to reach a size of 0.50 ± 0.09 cm (n=102). This size is low compare to their expected size at that growing stage, which is probably due to the source of food. Indeed oatmeal was given to the chironomides instead fish powder to avoid Hg contamination from food and thus a possible bias in Hg accumulation data but oatmeal was probably not nutritive enough for the chironomides.

Larvae exposed for 7 days to the studied sediments were found to have similar size as well as to have comparable Hg content, suggesting that the higher Hg content in sediments of impacted reservoirs was not bioavailable for the chironomides (Figure 1). The growth of the ostracods *H. incongruens* was also not impacted by 6-day contact with the studied sediments (data not shown). Both ecotoxicological tests did not reveal any potential toxicity of the studied sediments for benthic invertebrates.

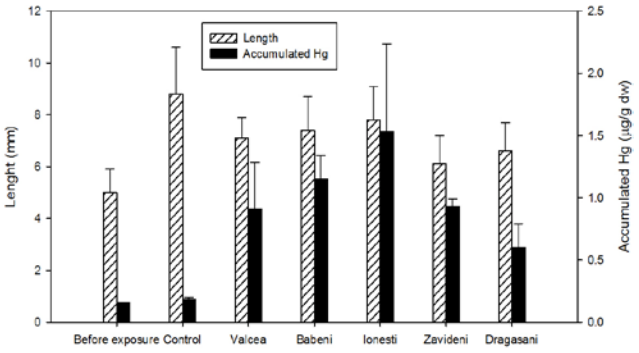

Figure 9: Length and Hg content in *Chironomus riparius* before and after exposure to sediments collected along the Olt river. Control represents exposure to OECD sediment.

3.4. Trophic transfer and potential impact to humans (GeoEcoMar)

This activity was postponed for 2015 due to the general weather of 2014. Last spring and summer were exceptional wet ones, with high precipitations and floods all along the Olt River basin. A new field campaign will be organized in April-May 2015 to gather the samples necessary to assess the trophic transfer.

4. Deviations from research plan

The comparison of the bathymetry measured in 2013 with the initial morphology has been started but it is not finalized since the access to historical data, owned by Hidroelectrica S.A. is restricted. As mentioned in §3.4 the trophic transfer and human impact related activities were postponed for 2015 due to the floods along the Olt River in 2014. Due to the fact that one of the nominated researchers (Costin Ungureanu) passed away in June 2014, he was replaced with Albert Scrieciu.

5. Collaboration aspects

General co-ordination of the project is under the responsibility of Prof. V. Slaveykova, Director of the Institute F. A. Forel and with active participation of Dr Loizeau. Dr Oaie, Director of GeoEcoMar and Prof. Ungureanu for Romanian side. During the second year collaboration aspects include participation in one meeting, two research stays of the GeoEcoMAr researchers to University of Geneva and two field campaigns. In addition, Skype conferences were organised at the regular base, as well as e –mail exchanges all over the year. To increase the visibility of the project and the cooperation between University of Geneva and GeoEcoMar, a web page dedicated on the project following the communication strategy for the Romanian-Swiss Research Programme was developed by Mme K. Loizeau (http://www.unige.ch/forel/fr/biogeochimie/projets1/mercuro/).

Prof. Ungureanu and Dr Loizeau participated in the mid-term meeting of the Romanian-Swiss Research Programme (RSRP) on 5 June 2015. They exposed a poster presenting the goals and first results of the MERCURO project to other project coordinators and Romanian scientific authorities.
Two visits of Romanian researchers to Geneva were carried out as planned. Mrs Irina Catianis participated in training program between the 18th of November and 14th December of 2014 and performed ecotoxicological tests in close collaboration with Séverine Le Faucheur. During that period, Ms Catianis was introduced to the relevant literature, got involved in the planning of the experiments, lab-trained to ecotoxicological assays performed with freshwater sediments and participated in the final scientific

discussions. The toxicity of the bottom sediments collected in five reservoirs along the Olt River in September 2014 was evaluated using two bioassays with the diptera larvae *Chironomus riparius*, and the ostracod *Heterocypris incongruens*. Both organisms were chosen based on their common use in sediment toxicity testing and their occurrence in a wide variety of aquatic environments including freshwater systems. Following this training period Mme Catianis will implement *Chironomus* test in the newly developed laboratory of ecotoxicology in GeoEcoMar.

Mr Dan Vasiliu participated in a training programme between the 4th to the 29th of November 2014. He was trained and supervised by Mrs Elena Gascon in the analysis of total and methyl-mercury in soil and lake sediment samples, on an AMA 254 total mercury analyser and MERX automated methylmercury system. The AMA 254 is available at Bucharest and the present training help Mr Vasiliu to master this instrument, and perform an inter-comparison between our two labs. A total of around 100 samples were prepared and analysed for Total Hg and 40 samples were analysed for methylmercury.

In the beginning of 2014 the two remaining pieces of equipment contracted in 2013 have been delivered: the cell counter and the growth chamber (incubator). Both of them are operational and contribute to the establishment of the ecotoxicology laboratory infrastructure of GeoEcoMar.

Three field campaigns were organized during this period. First one between 20 and 25th of April 2014 was dedicated to sediment sampling, while the second one between 14th and 17th of June 2014 to the soil sampling. These two campaigns were performed by the research stuff of GeoEcoMar. The third campaign between the 9th and 20th of September 2014 was executed jointly with University of Geneva. V. Ungureanu and D. Grosu (GeoEcoMar), and C. Cosio, P. Dranguet, A. Freiburghaus and J.-L. Loizeau (UniGe) actively participated in this campaign. Sediment cores and waters samples in the four cascading reservoirs were collected; in-situ exposure and transplantation experiments with aquatic plants were performed by the UniGe team. Surface sediment samples in Dragasani reservoir were retrieved for total mercury analysis and benthic community determination by GeoEcoMar team. No major difficulties to mention during the organisation and performance of the field campaign.

6. Research output

Research papers

Dranguet, P.; Fluck, R.; Regier, N.; Cosio, C.; Le Faucheur, S.; Slaveykova, V. I., Towards Mechanistic Understanding of Mercury Availability and Toxicity to Aquatic Primary Producers. *Chimia 68*, (11), 799-805 (2014).
C. Cosio, R. Flück, N. Regier, V.I. Slaveykova "Effects of macrophytes on the fate of mercury in aquatic systems" *Environ. Toxicol. Chem.* Special issue Global mercury partnership 33, 1225-1237 (2014).
S. Le Faucheur, P. G.C. Campbell, C. Fortin and V.I. Slaveykova "Interactions between mercury and phytoplankton: Speciation, bioavailability and internal handling" *Environ. Toxicol. Chem*. Special issue Global mercury partnership 33, 1211-1224 (2014).

Poster presentations

Dranguet P., Le Faucheur S., Cosio C., Slaveykova V.I. (2014) Effect of mercury on periphyton collected in Romanian reservoirs impacted by industrial activities. 24th annual Europe meeting of the Society of Environmental Toxicology and Chemistry (SETAC) (Basel, Switzerland).
Dranguet P., Le Faucheur S., Cosio C., Slaveykova V.I. (2014) Periphyton communities in mercury impacted surface water body Goldschmidt Conference, 8-13 June, 2014 Sacramento, California, Goldschmidt Abstracts p. 596.

Table 1: Measured pH, mean dissolved organic carbon (± s.d., n=3), total dissolved concentrations of major cations and anions (± s.d., n=2) as well as total metal concentrations (± s.d., n=3) in the studied reservoirs along the Olt River and Govora River (Romania) in September 2013. N.D. indicates not determined. Charges omitted for simplicity.

Measured	Valcea	Babeni 1	Babeni 2	Ionesti	Zavideni	Dragasani	Govora	Valcea 5 days	Babeni 2 5 days
pH	8.48	7.68	8.14	7.94	7.95	8.15	10.7	9.5	7.6
DOC ($mg.L^{-1}$)	3.3 ± 0.1	3.8 ± 0.4	3 ± 0.04	3 ± 0.03	2.7 ± 0.4	3.5 ± 0.2	58.4 ± 0.2	3.6 ± 0.2	3± 0.3
Na ($mg.L^{-1}$)	16.51 ± 0.07	21 ± 3.5	32.43 ± 0.05	64 ± 1	54 ± 1	47.96 ± 0.05	227± 1	16.51 ± 0.07	32.43 ± 0.05
K ($mg.L^{-1}$)	3.58 ± 0.04	3.80± 0.13	1.27 ± 0.2	2.80 ± 0.07	1.77 ± 0.02	3.64 ± 0.07	1789 ± 24	3.58 ± 0.04	1.27 ± 0.2
Mg ($mg.L^{-1}$)	3.94 ± 0.02	4.2 ± 0.12	<0.01	2.40 ± 0.08	5.36 ± 0.02	4.23 ± 0.76	420 ± 17	3.94 ± 0.02	<0.01
Ca $mg.L^{-1}$)	84 ± 2	110 ± 1	31 ± 6	29.75 ± 0.05	44.08 ± 0.05	96 ± 1	57367 ± 1183	84 ± 2	31.11 ± 6.35
F ($\mu g.L^{-1}$)	65 ± 2	30.5 ± 0.50	33.5 ± 0.50	71 ± 8.00	82.5 ± 1.5	83.5 ± 1.5	5500 ± 500	65 ± 2	33.5 ± 0.5
Cl ($mg.L^{-1}$)	20.37 ± 0.05	73± 1	113± 1	223 ± 1	186 ± 1	158 ± 0.04	5881 ± 20	20.37 ± 0.05	112.70 ± 0.65
Br ($\mu g.L^{-1}$)	22.5 ± 1.5	13.5 ± 1.5	12.5 ± 0.5	17.5 ± 4.5	28 ± 3	31 ± 3	<0.01	22.5 ± 1.5	12.5 ± 0.5
SO_4 ($mg.L^{-1}$)	15.88 ± 0.02	8.9 ± 0.01	10.32 ± 0.44	22.19 ± 0.2	20.78 ± 0.05	19.36 ± 0.13	66.5 ± 1.5	15.88 ± 0.02	10.32 ± 0.44
NO_3 ($mg.L^{-1}$)	3.05 ± 0.06	1.78 ± 0.26	2.02 ± 0.21	3.38 ± 0.02	3.38 ± 0.17	3.0 ± 0.01	281 ± 131	3.05 ± 0.06	2.02 ± 0.21
Cr ($\mu g.L^{-1}$)	11.66 ± 0.01	7.72 ± 0.01	3.32 ± 0.01	1.25 ± 0.01	1.27 ± 0.01	0.76 ± 0.01	N.D	11.66 ± 0.01	3.32 ± 0.01
Cd (ng.L-1)	27± 1	29± 0.6	29± 1	26.6± 0.6	27± 0.9	27± 0.7	N.D	36.5± 0.6	27.83± 0.01
Ni (µg.L-1)	0.87 ± 0.04	1.03 ± 0.07	1.17 ± 0.1	1.02 ± 0.05	1.00 ± 0.02	0.95 ± 0.03	N.D	0.72± 0.01	1.00± 0.03
Zn (µg.L-1)	0.46 ± 0.28	1.36 ± 1.14	3.42 ± 0.88	0	0	0	N.D	0.63 ± 0.01	4.52 ± 0.01
Pb (µg.L-1)	0.23 ± 0.01	0.49 ± 0.03	0.55 ± 0.03	0.49 ± 0.02	0.42 ± 0.01	0.56 ± 0.24	N.D	0.21 ± 0.01	0.52 ± 0.02
Cu ($\mu g.L^{-1}$)	0.88 ± 0.01	1.01 ± 0.08	1.01 ± 0.04	0.87 ± 0.03	0.95 ± 0.02	0.92 ± 0.03	N.D	0.94 ± 0.02	0.90 ± 0.03
Hg ($ng.L^{-1}$)	1.53 ± 0.27	1.28± 0.14	2.36 ± 0.23	0.90 ± 0.06	0.70 ± 0.13	0.52± 0.07	9.98 ± 0.16	1.87 ± 0.18	1.45 ± 0.01