Jožef Stefan
Institute
Department of Environmental Sciences

GMOS
TRAIN

GMOS-Train Mid-Term Check

POSTER BOOKLET

Bibliographical Information

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Authors (in alphabetical order)	Aleksandra Lobnik, Aleš Lapanje, Alina Kleindienst, Aljoša Košak, Alkuin M. Koenig, Allwin Mabes Raj, Aurélien Dommergue, Bastien Thomas, Cecile Cathalot, Charikleia Gournia, Charlotte Haugk, Corinna Schrum, David Amouroux, David Amptmeijer, David Kocman, Dmitrii Deev, Elsie Sunderland, Emmanuel Rinnert, Francesco De Simone, Geoflamme shipboard scientific party, Ian M. Hedgecock, Iaroslav Rybkin, Igor Živković, Isabel Garcia Arevalo, Jan Gačnik, Jeroen E. Sonke, Joël Knoery, Johannes Bieser, Jože Kotnik, Katarina Gårdfeldt, Koketso Molepo,, Lars-Eric Heimbürger-Boavida, Marta Jagodic Hudobivnik, Milena Horvat, Nicola Pirrone, Noelle E. Selin, Olivier Magand, Radojko Jaćimović, Ralf Ebinghaus, Rémy Guyoneaud. Saeed Waqar Ali, Sergio Ribeiro Guevara, Sofi Jonsson, Sonja Gindorf, Sreekanth Vijayakumaran Nair, Teodor D. Andron, Tomaž Rijavec, Volker Matthias, Warren T. Corns
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Contact info	Jožef Stefan Institute Jamova 39 SI - 1000 Ljubljana Slovenia Phone: +386 1 588 54 15 e-mail: info@gmos-train.eu
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Foreword

The GMOS-Train Mid-Term meeting was an opportunity for the GMOS-Train Early Stage Researchers (ESRs) and supervisors to meet face-to-face for the first time. The meeting offered an opportunity to present the main achievements of their research work and exchange ideas for further work. The high interdependence of research work between the EFF is an opportunity but also a challenge that requires additional effort from all researchers involved. Therefore, it was decided to prepare posters that will offer a platform to present the work of individual ESRs after the first year of their participation in the GMOS-Train project. In addition, additional presentations of doctoral student research at JSI were presented. Although the research work is in its infancy, the results obtained are very promising and the foundations for a good continuation have been established. The present publication summarizes all the posters presented at the interim GMOS-Train meeting, which took place from 6 to 8 October 2021 at the Jožef Stefan Institute in Ljubljana, Slovenia. Congratulations to all participants!

Prof Milena Horvat
GMOS-Train Coordinator

GMOS-Train Mid-Term Check Meeting Participants

List of Posters

1. Alina Kleindienst, Rémy Guyoneaud, Corinna Schrum, David Amouroux: ***Monomethyl-mercury (MMHg) sources in coastal ecosystem: new molecular and isotopic experimental speciation approaches***

2. Alkuin M. Koenig, Aurélien Dommergue, Olivier Magand, Jeroen E. Sonke: ***El-Niño driven mercury increase in tropical South America linked to high-altitude air masses from the tropical Pacific***

3. Allwin Mabes Raj, Aleksandra Lobnik, Aljoša Košak, Aleš Lapanje: ***Removal of Hg^{2+} ions from aqueous solutions using amino-functionalized SiO_2 particles***

4. Charikleia Gournia, Nicola Pirrone, Noelle E. Selin, Volker Matthias, Ian M. Hedgecock, Francesco De Simone: ***An analysis of Mercury Deposition based on four different Anthropogenic Mercury Emission Inventories***

5. Charlotte Haugk, Sofi Jonsson, David Kocman, Katarina Gårdfeldt: ***Release of Mercury from Thawing Permafrost***

6. David Amptmeijer, Johannes Bieser, Joël Knoery, Corinna Schrum: ***Modelling the importance of the ecoystem in marine Hg cycling***

7. Dmitrii Deev, Iaroslav Rybkin, Tomaz Rijavec, Ales Lapanje: ***Methylation of mercury by SRB can be an aerobic process***

8. Isabel Garcia Arevalo, Joël Knoery, Bastien Thomas, Cecile Cathalot, Emmanuel Rinnert, Geoflamme shipboard scientific party: ***Preliminar results of volcanic exhalations influence on mercury levels from a newlyformed submarine volcano and a seepage site in the Western Indian Ocean***

9. Jan Gačnik, Igor Živković, Sergio Ribeiro Guevara, Radojko Jaćimović, Jože Kotnik, Milena Horvat: ***Use of ^{197}Hg radiotracer in Hg research***

10. Koketso Molepo, Ralf Ebinghaus, Aurélien Dommergue, Noelle Selin: ***Exploration of long term observational datasets to examine ocean atmosphere exchange processes of Hg***

11. Saeed Waqar Ali, David Kocman, Marta Jagodic Hudobivnik, Milena Horvat: ***Evaluation of sample preparation methodologies for multi-elemental analysis in foliage samples***

12. Sonja Gindorf, Sofi Jonsson, Lars-Eric Heimbürger-Boavida, David Amouroux, Elsie Sunderland: ***The Role of Dissolved Organic Matter in the Coastal Cycling of Mercury***

13. Sreekanth Vijayakumaran Nair, Jan Gačnik, Igor Živković, Milena Horvat: ***Hg^0 Plasma oxidation experiments, analysis of Hg^{II} species using Quadrupole Mass Spectrometer, and KCl denuder specificity stability tests***

14. Teodor D. Andron, Milena Horvat, Warren T. Corns, Igor Živković, Jože Kotnik: ***Tackling the uncertainty of using gas calibrators for GEM that use the Dumarey equation***

GMOS TRAIN

UNIVERSITÉ DE PAU ET DES PAYS DE L'ADOUR

Monomethyl-mercury (MMHg) sources in coastal ecosystem: new molecular and isotopic experimental speciation approaches

Alina Kleindienst[1], Rémy Guyoneaud[1], Corinna Schrum[3] and David Amouroux[1]

[1]Universite de Pau et des Pays de L'Adour, E2S UPPA, CNRS, IPREM Institut des Sciences Analytiques et des Physico-chimie pour L'Environement et Les Matéiaux, Pau, France, [2]Institute of Coastal Systems, Helmholtz-Zentrum Hereon, Max-Planck-Strasse 1, 21502 Geesthacht, Germany

Introduction

Mercury (Hg) in marine systems has been extensively studied over the past decades but knowledge gaps on Hg-transformation dynamics and underlying (a-)biotic mechanisms remain [1].

Coastal areas and ocean margins are of great ecological and economical importance, receiving significant riverine Hg and nutrient inputs thus potentially favoring MMHg production and bioaccumulation in fish [2]. Novel coupled biogeochemical models are being developed to evaluate the effectiveness of the implementation of the Minamata Convention.
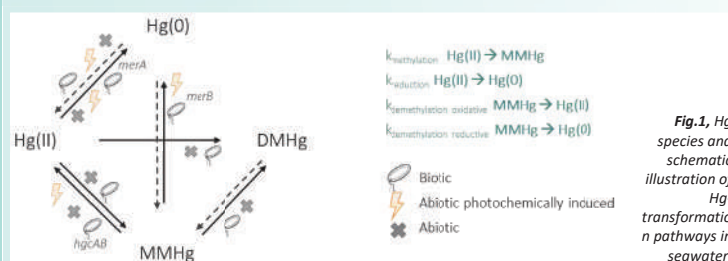

Fig.1, Hg species and schematic illustration of Hg-transformation pathways in seawater.

However, substantial differences in transformation rates due to methodological differences may introduce large uncertainties in model calculations. **To study Hg-transformation dynamics in coastal environments, a methodological critically evaluated experimental approach is needed.**

Materials and Methods

- Seawater incubation experiments in different conditions (Fig. 2) applying enriched stable isotopic tracers adapting established protocols[3]
- Preservation of samples (triplicates) at chosen timepoints and trapping of volatile Hg species
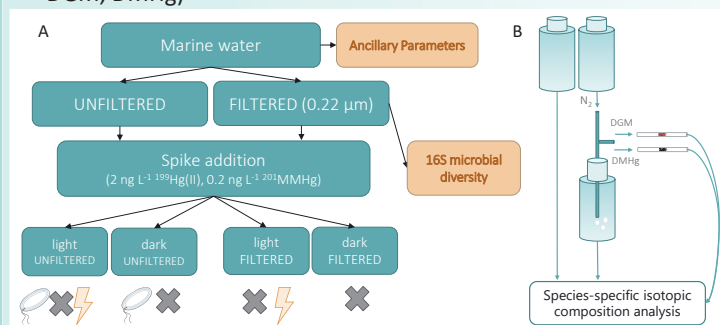- Species-specific isotopic composition analysis (MMHg, Hg(II), DGM, DMHg)[4]

Fig.2, A General overview for incubation experiments under different conditions; B Incubations are conducted in sextuplicate, triplicates are immediately acidified and Hg(0) displaced onto gold (DGM) or carbo (DMHg) traps for remaining samples

Incubations experiments were conducted in the

- Western Mediterranean Sea: on surface waters from a coastal (C), shelf (S) and a deeper margin station (O), fall 2020 and spring 2021
- Bay of Biscay (Atlantic Ocean): on coastal (C) and estuarine-influenced (E) waters, spring 2021
- Baltic Sea: on coastal (C) and deep open station (O), summer 2021

Preliminary Results and Discussion

Herein we present preliminary results on NW and BB incubations. Mass balance calculation on discrete Hg isotopes indicate good recoveries (Fig.3 A,B).

Methylation was not observed, however, photochemically induced demethylation in bulk waters (as linear best fit ln (MMHg)) was in the same order of magnitude for MS and BB waters (3.6-$9.6 \times 10^{-3} h^{-1}$; all $R^2 > 0.85$). In the dark, demethylation was significantly different for MS waters in fall (C: $3.5 \times 10^{-4} h^{-1}$, S: $4.6 \times 10^{-5} h^{-1}$; all $R^2 > 0.97$) but comparable for BB waters in spring (4.7-$6.5 \times 10^{-4} h^{-1}$; all $R^2 > 0.96$). Hg(0) produced from ^{201}MMHg was detectable in unfiltered but not in filtered waters (spring MS, BB), indicating a microbiologically mediated degradation pathway.

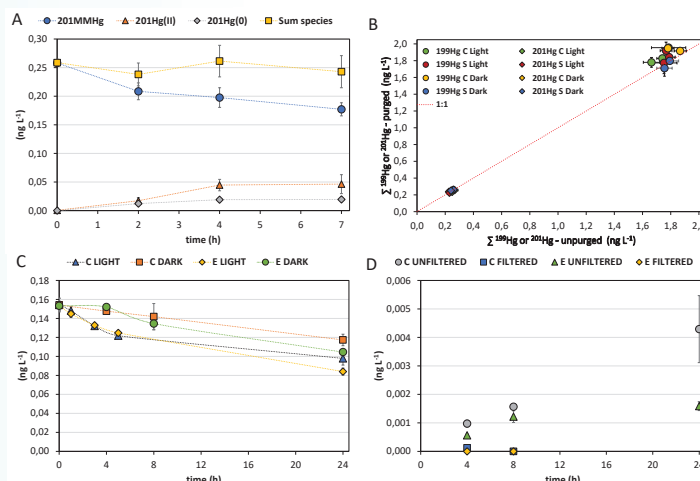Fig.3, Error bars for A-D represent 1SD for triplicate incubations, A MMHg, Hg(II) and Hg(0) on ^{201}Hg isotope for NW coastal station (C) in fall 2020 under natural light conditions; B Sum of averages Hg species on ^{199}Hg and ^{201}Hg for all incubations from NW fall 2020 comparison between unpurged and purged incubations; C demethylation in BB incubations from spring 2021 in UNFILTERED samples under light and dark conditions, please note lines are not a kinetic fit; D ^{201}Hg(0) in unfiltered and filtered samples from BB coastal (C) and estuarine-influenced (E) waters

Preliminary Conclusions

- Validation of Incubation approach based on mass balance calculations (^{199}Hg, ^{201}MMHg)
- Observable differences in spatially close waters in Hg-transformation dynamics
- Importance of photochemically and (micro-)biological induced MMHg degradation

Outlook

- **Field campaigns fall 2021 (BB, NW)**
- **Incubation experiments along a deep profile (NW, BS)**
- **Extraction and sequencing - 16S rDNA (field campaigns 2021)**
- **MMHg stable isotope fractionation – development of experimental set-up degradation experiments**

References
[1] Bowman et al. (2020), Sci. Total Environ **710**, 136166.
[2] Liu et al. (2021), Nat. Geosci. **14**, 672–677.
[3] Monperrus et al. (2007), Mar. Chem. **107**, 49–63.
[4] Rodríguez et al. (2004), Mar. Chem. **90**, 107-123.

GMOS TRAIN

UGA — Université Grenoble Alpes

El-Niño driven mercury increase in tropical South America linked to high-altitude air masses from the tropical Pacific

Alkuin M. Koenig[1], Aurélien Dommergue[1], Olivier Magand[1], Jeroen E. Sonke[2]

[1] Institut des Géosciences de l'Environnement, Université Grenoble Alpes, CNRS, IRD, Grenoble INP, Grenoble, France
[2] Géosciences Environnement Toulouse, CNRS/IRD/Université Paul Sabatier Toulouse 3, France

Context & Introduction

- We measured atmospheric mercury (Hg) at Chacaltaya mountain (5240 m asl.) in tropical South America[1].

- A strong Hg increase (+ ≈ 50%) was observed during the 2015-2016 „Godzilla" El Niño.

- The question: **What caused this Hg increase?**

Methods

- Hg was measured in the form of total gaseous mercury (TGM) with a Tekran 2537A Analyzer[1].

- Air mass origin was characterized with HYSPLIT back trajectories[2] (meteo input: GDAS1).

- The first derivative of experimental data (dHg/dt) was estimated with smoothing spline functions[3].

Results and discussion

1) Transport anomaly during the 2015-2016 El Niño

- Air mass origin shifted strongly westwards.

- The anomaly was most pronounced for westerly mid-to-upper tropospheric (4 km – 13 km) air masses.

  → We defined this air mass origin as:
  „Upper Tropospheric Pacific Channel" (UTPC)

2) A box model approach to link Hg data with transport

- We assumed that Hg at Chacaltaya is representative of a well-mixed box of arbitrary extent.

- Then, the inward transport (through the UTPC) should relate not directly to Hg concentrations within the box, but to their rate of change (dHg/dt):

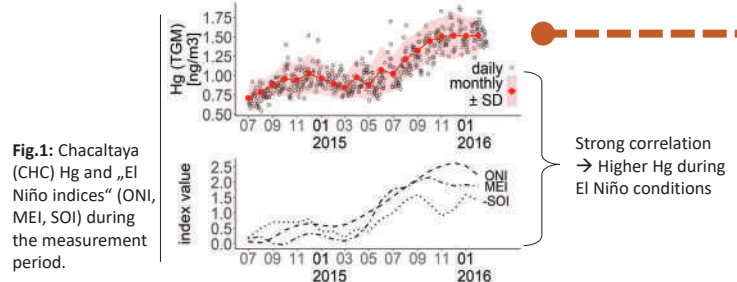$$dHg/dt = a \cdot UTPC(t) + b + f(t) \qquad \text{(Eq.1)}$$

→ Based on Eq.1, we found a strong relationship between **westerly transport (UTPC)** and **Chacaltaya dHg/dt**.
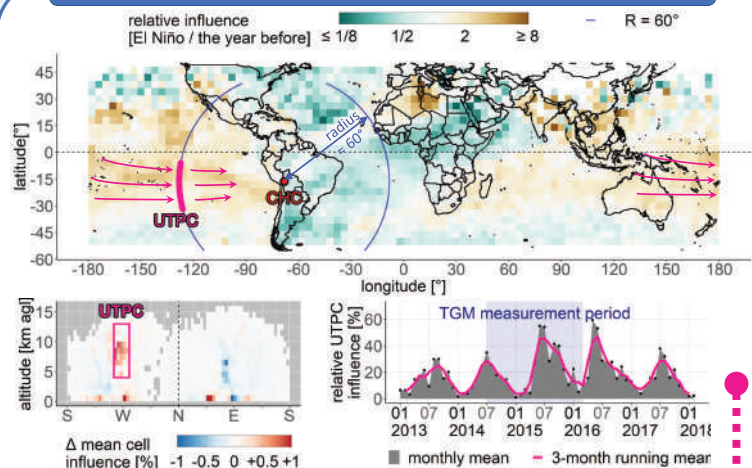
Conclusions

- The Hg increase at Chacaltaya is linked to an increased influence of westerly air masses originating from the tropical pacific ocean.

- These are not surface air masses, but come from the mid-to-upper troposphere (4 km – 13 km agl).

- The results suggest enhanced Hg concentrations in the upper troposphere of the tropical Pacific (UTPC).

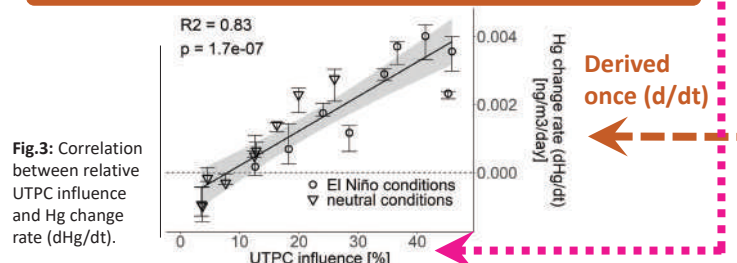**Why high Hg in the UTPC? Our hypothesis: It is due to interhemispheric exchange.**

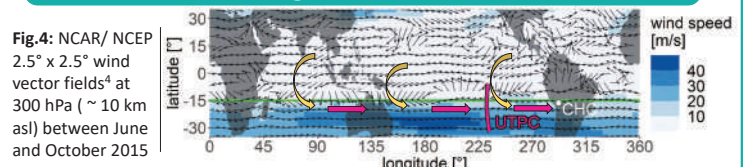### Atmospheric mercury (Hg) increase during El Niño

Fig.1: Chacaltaya (CHC) Hg and „El Niño indices" (ONI, MEI, SOI) during the measurement period.

Strong correlation → Higher Hg during El Niño conditions

Transport anomaly during El Niño (UTPC enhanced)

Fig.2: HYSPLIT-derived transport anomalies during El Niño. **Top:** Relative influence of source regions during El Niño conditions (05.2015 to 05.2016) compared to neutral conditions (05.2014 to 05.2015). **Left:** Difference in air mass origin between El Niño and neutral conditions for a cross-section at a radius of 60° from Chacaltaya. **Right:** Time evolution of the air mass origin defined as „UTPC".

Hg increase linked to enhanced UTPC influence

Derived once (d/dt)

Fig.3: Correlation between relative UTPC influence and Hg change rate (dHg/dt).

Hypothesis: interhemispheric exchange causes high Hg in the UTPC

Fig.4: NCAR/ NCEP 2.5° x 2.5° wind vector fields[4] at 300 hPa (~ 10 km asl) between June and October 2015

References:
[1] Koenig et al. (2021), ACP, DOI: 10.5194/acp-21-3447-2021
[2] Stein et al. (2015), BAMS, DOI: 10.1175/BAMS-D-14-00110.1
[3] Berghaus and Cannon (1973), EM, DOI: 10.1007/BF02319311
[4] From NOAA: https://psl.noaa.gov/cgi-bin/data/composites/printpage.pl [last access: 20/09/2021]

Removal of Hg^{2+} ions from aqueous solutions using amino-functionalized SiO$_2$ particles

A. F. P. Allwin Mabes Raj [1], Aleksandra Lobnik [1], Aljoša Košak[1], Aleš Lapanje[2]

1 IOS, Institute of Environmental Protection and Sensors, Ltd., Beloruska 7, SI-2000 Maribor, Slovenia

2 Department of Environmental Sciences, Jožef Stefan Institute, Jamova 39, 1000 Ljubljana, Slovenia

Introduction

Amino-functionalized SiO$_2$ particles (NH$_2$@SiO$_2$) based on the Stöber method involving the reaction of hydrolysis and condensation of alkoxide precursors tetraethoxysilane (TEOS) and (3-aminopropyl) trimethoxysilane (APTMS) for specific and selective removal of Hg^{2+}. The prepared silica and amino-functionalized silica particles were characterized by FT-IR spectroscopy, thermogravimetric analysis (TGA), specific surface area (BET), transmission electron microscopy (TEM), zeta potential measurements and potential titration measurements. We studied the adsorption efficiency of the prepared adsorbent materials toward Hg^{2+} in model salt solutions. The adsorption process was evaluated in terms of adsorption efficiency, adsorption capacity, adsorption isotherms, kinetics and thermodynamic based on the result from inductively coupled plasma optical emission spectrometer (ICP-OES) measurements for Hg^{2+}. Desorption of Hg^{2+} from nanomaterials were performed using 1.5 molar citric acid solution.

Experimental Work
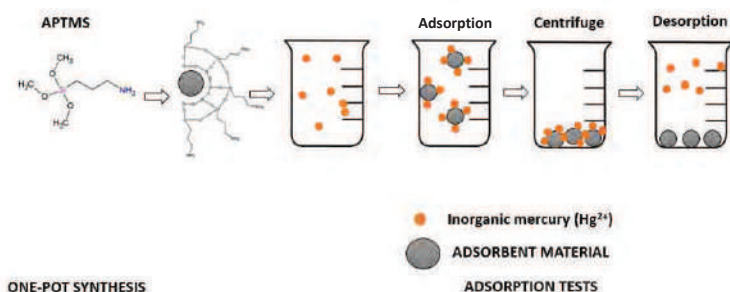
Fig. 1 Methodology of adsorption & desorption

BET spectra surface area and porosity analysis

Potentiometric titration

The specific surface area measured using Brunaer – Emer – Teller (BET) **(Fig.5)** Electro- kinetics shows successful surface binding **(Fig. 6)**.

Fig. 5 BET hysteresis curves & **Fig. 6** Zeta potential curve

Results and Discussion

Transmission Electron Microscopy Analysis

TEM images of the silica nanoparticles SiO$_2$ showed that they are monodispersed spherical in shape with an average particles of size 500 nm **(Fig. 2)**.
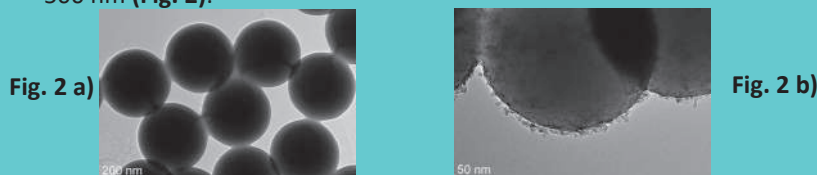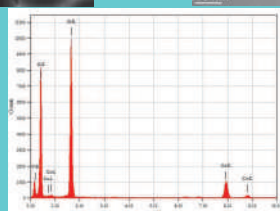

Fig. 2 TEM images of **a)** silica (SiO$_2$) nanoparticles **b)** (SiO$_2$) nanoparticles coated with amino groups & **c)** EDXS patterns with characteristics of Si-O-Si bond in SiO$_2$ structure

Removal/Adsorption efficiency: The adsorption percentage of Hg^{2+} ions was calculated by the difference of initial and final concentration using the equation:

$$\text{adsorption efficiency } (\%) = \frac{C_0 - C_t}{C_0} * 100$$

Where C_0 is initial concentration of Hg^{2+} in solution and C_t (mg/L) is the concentration of Hg^{2+} after adsorption

Removal efficiency

Removal efficiency plot for Hg^{2+} in varied time points by amino functionalized SiO$_2$ particles **(Fig. 7)**.

Removal efficiency :88%
Desorption 100%

Fig. 7 Removal efficiency of amino functionalized SiO$_2$ particles on Hg^{2+}

Thermogravimetric analysis

FT-IR Spectroscopy

TGA results of SiO$_2$ nanoparticles coated with NH$_2$ **(Fig. 3)**

FTIR measurements for amino coated SiO$_2$ particles **(Fig. 4)**

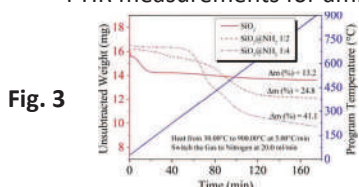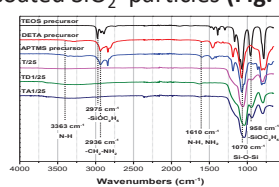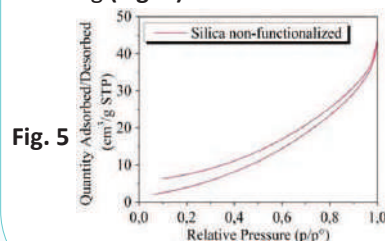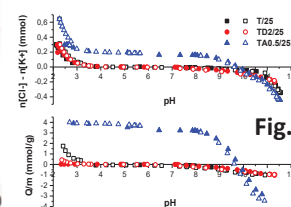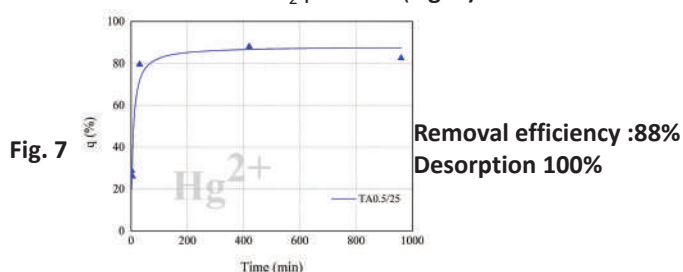**Fig. 3** TG analysis of NH$_2$ coated SiO$_2$ nanoparticles

Fig. 4 FTIR spectra of silica nanoparticles with NH$_2$ group

Conclusion

Highly efficient adsorption system-Biosensing application – Future work – Gold nano particle based sensing.

References

1. Ding, Yu & Zhu, Weiping & Xu, Yufang & Qian, Xuhong. (2015). A small molecular fluorescent sensor functionalized silica microsphere for detection and removal of mercury, cadmium, and lead ions in aqueous solutions. Sensors and Actuators B: Chemical. 220. 762-771. 10.1016/j.snb.2015.05.113.
2. Chunrong Ren, Xingeng Ding, Wenqi Li, Huating Wu, Hui Yang(2017). Highly Efficient Adsorption of Heavy Metals onto Novel Magnetic Porous Composites Modified with Amino Groups. J. Chem. Eng. Data 2017, 62, 6, 1865–1875.

An analysis of Mercury Deposition based on four different Anthropogenic Mercury Emission Inventories

Charikleia Gournia[1], Nicola Pirrone[1], Noelle E. Selin[2,3], Volker Matthias[4], Ian M. Hedgecock[1], Francesco De Simone[1]

[1]National Research Council—Institute of Atmospheric Pollution Research c/o University of Calabria, 87036 Rende, Italy
[2]Institute for Data, Systems, and Society, Massachusetts Institute of Technology
[3]Department of Earth, Atmospheric, and Planetary Sciences, Massachusetts Institute of Technology
[4]Institute of Coastal Research, Helmholtz-Zentrum Hereon, Max-Planck-Straße 1, 21502 Geesthacht, Germany

Introduction

Global inventories are a key component of global Hg modeling, used as input data sets for the transport and chemical reaction calculations. A deeper comprehension of the inconsistencies between inventories could deliver information on the accuracy and precision of the employed methods and identify areas for further study. This study performs an analysis of Hg deposition patterns using a global-scale transport model and several numerical experiments driven by four different anthropogenic emission inventories (AMAP/UNEP, EDGAR,STREETS,WHET).

Materials and methods

We used the GEOS-Chem version 13.2 to simulate elemental mercury, divalent mercury, and primary particulate mercury atmospheric mercury species. Our simulations are conducted for a 6-year period with the first 5 years used for initialization and 2015 as a production year. GEOS FP, the current operational met data product from NASA/GMAO, are used for the simulations. The horizontal resolution is degraded here to $2°×2.5°$ for input to GEOS-Chem. We also investigate the effect of the oxidation processes of atmospheric Hg using an O3/OH oxidation scheme and an alternative Br based oxidation mechanism.

Fig. 1 Geos-Chem model configuration files for mercury simulation.
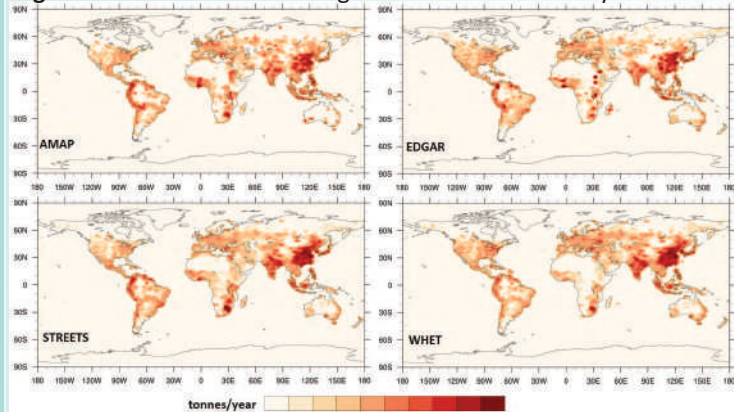

Fig. 2 Geographical distribution of annual Hg anthropogenic emissions as estimated by AMAP-2010, EDGAR, STREETS and WHET.
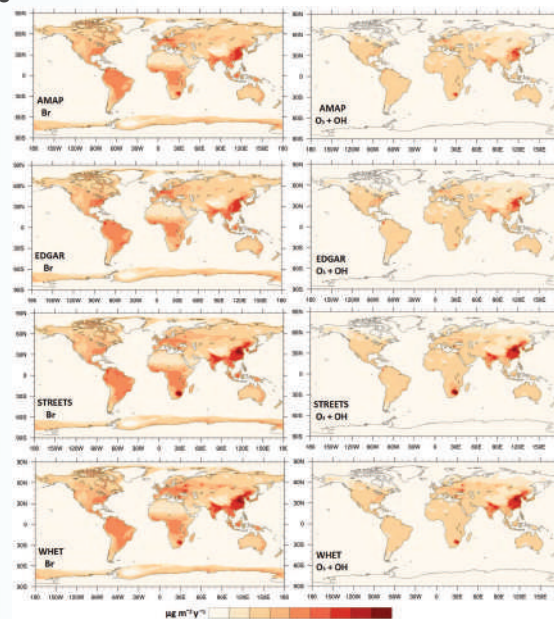
Results

Fig. 3 Geographical distribution of the dry deposition for the year 2015 resulting from model runs adopting Bromine and O_3/OH oxidation for the different inventories.
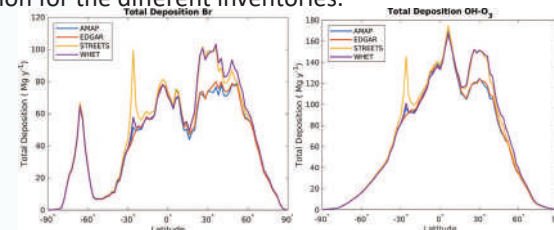

Fig. 4 Latitudinal profiles of mercury total deposition for the year 2015 resulting from model runs adopting Bromine and O_3/OH oxidation for the different inventories.
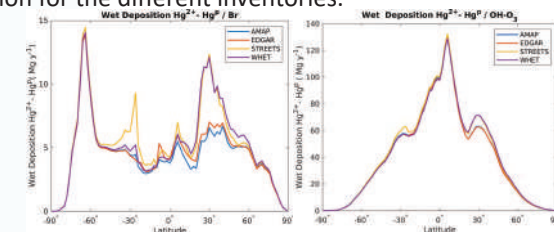

Fig. 5 Latitudinal profiles of Hg^{2+} and Hg^p wet deposition for the year 2015 resulting from model runs adopting Bromine and O_3/OH oxidation for the different inventories.

Conclusion

The analysis highlighted minor differences in the geographical distribution of Hg deposition when using inventories that rely on different approaches and levels of detail. A more precise understanding of the chemical processes involved in atmospheric Hg redox reactions would allow models to better simulate Hg deposition and deepen our understanding of where and how Hg circulates worldwide.

GMOS TRAIN

Release of Mercury from Thawing Permafrost

Charlotte Haugk[1], Sofi Jonsson[1], David Kocman[2], Katarina Gårdfeldt[3]

[1]Stockholm University, Department of Environmental Science (ACES)
[2]Józef Stefan Institute, Department of Environmental Sciences
[3]Polarforskningssekretariat, Swedish Polar Research Secretariat

1. AIM

To understand how a warming climate can perturb the biogeochemical cycle of mercury (Hg) in Arctic environments.

2. APPROACH

- THg & MeHg in soil, water & biota
- Lab-based incubation experiments
- Determination of Hg (de)methylation rates
- Data implementation into global Hg model

3. ACCOMPLISHMENTS

Permafrost thaw gradients

Analytical techniques for THg/MeHg

✓ Data on **Hg (de)methylation** in permafrost thaw gradients as well as THg & MeHg stocks

✓ Training in **analytical techniques for THg and MeHg** using DMA & Tekran 2700 MeHg analyzer coupled to IPCMS

✓ Sampling of **long-term geoengineering earthworm experiment**

✓ **Field Campaigns**

Abisko Field Campaign 2020

Earthworms in Arctic Soil mesocosms

Tavvavuoma Field Campaign 2021

4. WHATS NEXT

- **Incubation experiments** investigating timeframes of Hg (de)methylation & remobilization
- Climate Scenario Field Experiment

Charlotte.Haugk@aces.su.se

Modelling the importance of the ecoystem in marine Hg cycling

David Amptmeijer[1], Dr. Johannes Bieser[1], Dr. Joël Knœry[2] and Prof. Dr. Corinna Schrum[1]

1 Institute of Coastal Research, Helmholtz-Zentrum Hereon, Max-Planck-Str. 1, 21502 Geesthacht, Germany

2 IFREMER, RBE/BE- LBCM, BP 21105, 44311 Nantes Cedex 3, France

Introduction

Mercury (Hg) speciation and bioaccumulation in coastal seas is an essential link between Hg pollution and methylmercury (CH_3Hg^+) exposure to humans. Yet, the dynamics and interactions between the ecosystem and marine Hg cycling are still poorly understood. We designed a bioaccumulation model in order to investigate the role of the ecosystem in marine Hg cycling.

Results and Discussion

Comparison to observations

We found good agreement with existing observations for dissolved Hg and CH_3Hg^+ and their accumulation in phyto- and zooplankton (Fig. 1). Results differ notably between runs with and without accumulation of Hg to Biota and organic material (Fig. 2) with more CH_3Hg^+ formation when bioaccumulation and binding to Organic material occurs. (Fig. 3)

CH_3Hg^+ build up in the ecosystem

Because the current model focuses on low trophic levels the difference in total Hg content appears to be limited. It is however telling when we solely look at the accumulation of CH_3Hg^+ and see a dramatic increase with each trophic level (Fig.4)

POC/DOC

Our model is greatly affected by the particulate (POC) and dissolved organic carbon (DOC) content of the water. POC and DOC decrease phytoplankton uptake by scavenging mercury from the watercolumn but increase the concentration in zooplankton that feed on the polluted POC.

Atmospheric exchange

Mercury can enter or leave the ocean via atmospheric exchange of elemental Hg^0. Uptake by bioaccumulation or association to organic material lowers surface Hg levels allowing for a higher net Hg transport into the ocean (Fig. 5). Results indicate the without bioaccumalation evaporation increases by 3% whereas removing all organic material leads to an increase of 31%.

Conclusion

The ecosystem plays an essential role in Hg speciation and cannot be ignored when modeling marine Hg cycling.

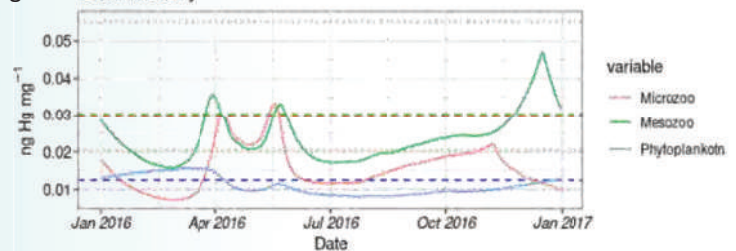## Acknowledgements

Fig. 1

Total mercury

Fig. 2

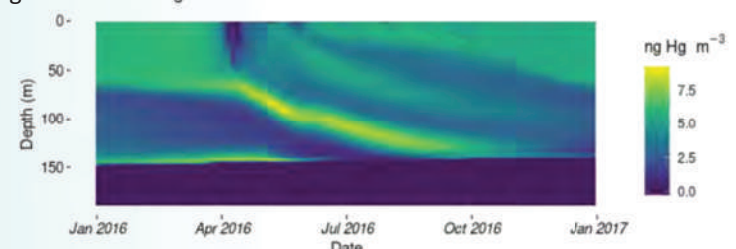Dissolved Hg with - without bioaccumulation and TOC

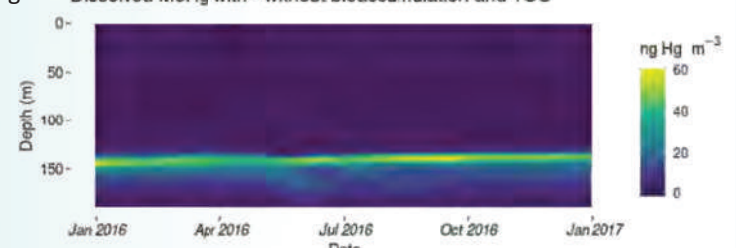Fig. 3

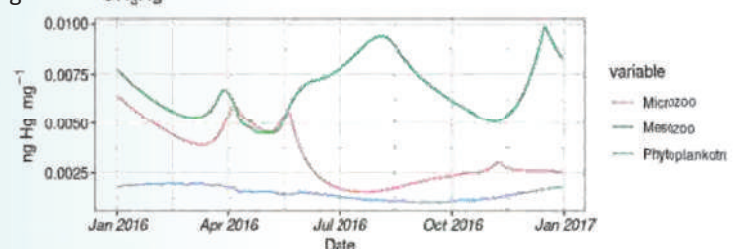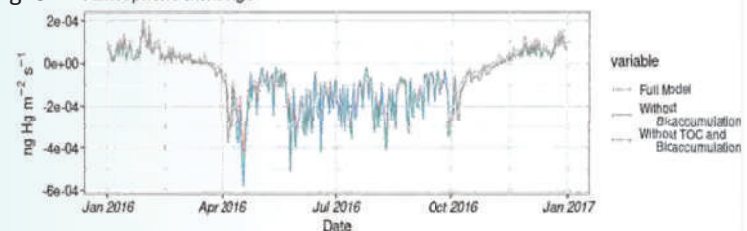Dissolved MeHg with - without bioaccumulation and TOC

Fig. 4

CH_3Hg

Fig. 5

Atmospheric exchange

Jožef Stefan Institute

# Methylation of mercury by SRB can be an aerobic process

Dmitrii Deev[1,2], Iaroslav Rybkin[1,2,3], Tomaz Rijavec[1], Ales Lapanje[1]

[1]Department of Environmental Sciences, Josef Stefan Institute, Jamova cesta 39, Ljubljana, Slovenia
[2]Jožef Stefan International Postgraduate School, Jamova cesta 39, Ljubljana, Slovenia
[3] Helmholtz-Zentrum Dresden-Rossendorf, Research Site Leipzig, Institute of Resource Ecology, Permoser Str. 15, 04318 Leipzig, Germany

Background:
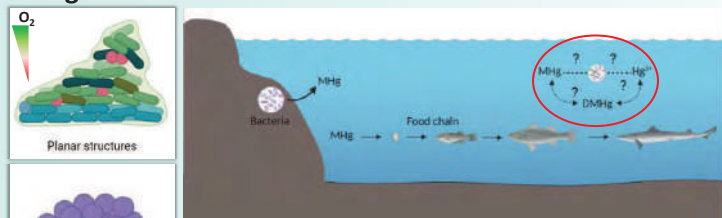
> Microbes play a crucial role in MeHg production.
> Bacterial communities may form anaerobic niches in an aerated environment.
> Possibly, this is the reason for the anomaly concerning the presence of MeHg in the seawater column.

Motivation:

To construct anaerobic niches **artificially** and to test the hypothesis that **mercury methylation can occur in an aerated environment**.
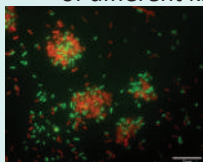
Methods:

Specific electrostatic aggregation of species
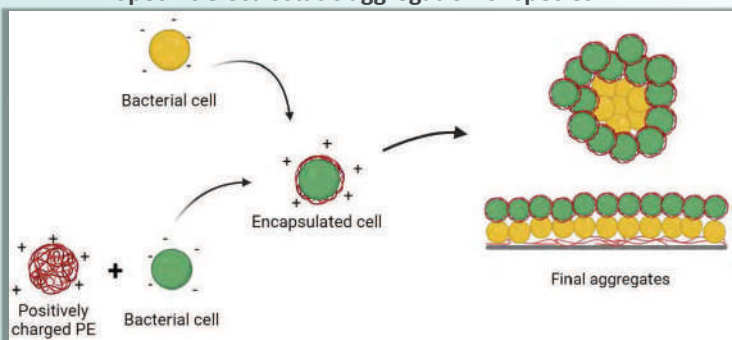
Surface charge of cells can be modified by polyelectrolyte, causing electrostatic attachment, including aggregation

Results:

Working protocol for the preparation of different kind of aggregates
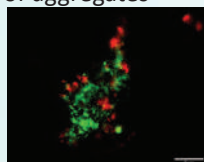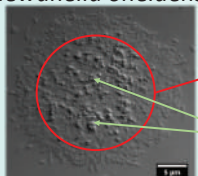

from single-species...   ...to multispecies
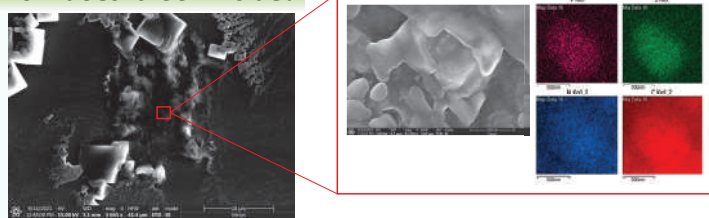
Proof of concept:

Anaerobic niche establishment for *Shewanella oneidensis*

Oxygen depletion triggers the anaerobic metabolism of facultative anaerobes.
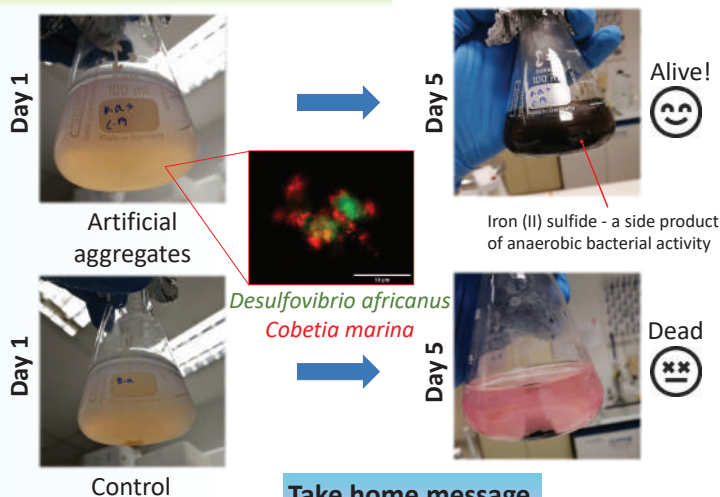

Anaerobic zone

Solid particles of metals – result of anaerobic activity

How does it look inside?

The formation of inorganic solid particles of vanadium was confirmed by SEM-EDS

Will it work for strict anaerobes?

Day 1   Day 5   Alive!

Artificial aggregates

Iron (II) sulfide - a side product of anaerobic bacterial activity

Desulfovibrio africanus
Cobetia marina

Day 1   Day 5   Dead

Control

Take home message

> We can grow strict anaerobic bacteria in aerobic conditions. It simplifies many complicated anaerobic biotech processes.
> With this method, we can determine environmental conditions where mercury methylation is most intensive.
> Cells don't tend to escape from the aggregates.

Future prospects:

> Studies of **methylating activity** of artificial aggregates both *in vitro* and *in situ*
> Characterization of aggregated bacterial communities

Acknowledgments:

References:

1. Deev D, Rybkin I, Rijavec T and Lapanje A (2021) When Beneficial Biofilm on Materials Is Needed: Electrostatic Attachment of Living Bacterial Cells Induces Biofilm Formation. *Front. Mater.* 8:624631. doi: 10.3389/fmats.2021.624631
2. Rybkin, I., Gorin, D., Sukhorukov, G., and Lapanje, A. (2019). Thickness of Polyelectrolyte Layers of Separately Confined Bacteria Alters Key Physiological Parameters on a Single Cell Level. *Front. Bioeng. Biotechnol.* 7, 378. doi:10.3389/fbioe.2019.00378
3. Rijavec, T., Zrimec, J., Spanning, R., and Lapanje, A. (2019). Natural Microbial Communities Can Be Manipulated by Artificially Constructed Biofilms. *Adv. Sci.* 6 (22), 1901408. doi:10.1002/advs.201901408
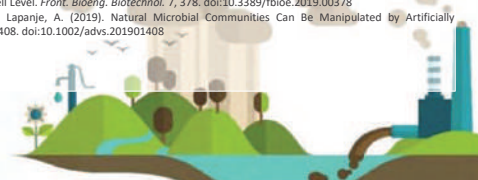
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Ifremer

Preliminar results of volcanic exhalations influence on mercury levels from a newly-formed submarine volcano and a seepage site in the Western Indian Ocean

Isabel Garcia Arevalo[1], Joël Knoery[1], Bastien Thomas[1], Cecile Cathalot[2], Emmanuel Rinnert[2], Geoflamme shipboard scientific party

1 Ifremer, Laboratoire de Biogéochimie des Contaminants Métalliques (RBE/BE/LBCM), Nantes, France.
2 Ifremer, Laboratoire Cycles Géochimiques et ressources (REM/GM/LCG), Plouzané, France

Introduction

Mercury (Hg) emissions from primary natural sources such as volcanic activity are considered an important but poorly understood source of Hg to the environment. Understandably, operational and analytical difficulties conspire to make Hg emission data difficult to obtain. However, there is a big knowledge gap concerning Hg emissions from submarine volcanism, specially regarding chemical speciation and environmental fate of Hg emitted from different volcanic systems (Edwards et al., 2021). In the present study, we investigated the concentration of Hg species in the volcanic exhalations and their distribution in the overlying water column near one the largest submarine eruption ever observed, off the island of Mayotte.

Materials and methods

The GEOFLAMME oceanographic cruise was carried out from April to May of 2021 board R/V Pourquoi pas? where water samples were collected in the vicinity of the volcano. Preliminary results are issued from real-time, shipboard determinations of dissolved total mercury species (HgT), and Dissolved Gaseous Mercury (DGM).

Fig 1. Station map

Results and Discussion

The results from this study include several depth-profiles over the full water column, both in areas under and without direct volcanic influence.
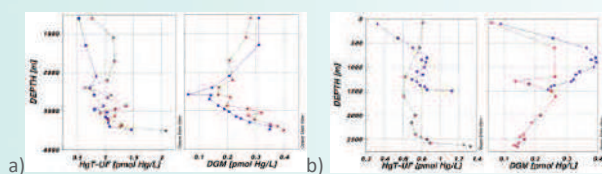

Fig 2. A) Reference (northern stations) and b) plume-influenced stations (red=volcano, blue=seepage site) mercury levels profiles in the water column

Hg concentrations show that volcanic degassing is associated with positive anomalies of Hg in deep levels.

Levels of Hg are likely impacted by sorption onto other plume components. This complicates the direct quantification of Hg inventory from submarine volcanic exhalations since minerals are readily resuspended from the seafloor.

These preliminary results will be completed by Hg determinations on particulate phases and on hot and cold fluids collected on the volcano.

Fig 3. HgT and DGM distribution in sections across a) newly-formed volcano and b) Horseshoe seepage site

Conclusion

Submarine volcanism contributes to hg global cycle. Nevertheless, oceanic processes conspire to contain emissions into the aquatic realm.
This study provided new constrains on the abundance and the chemical speciation of mercury in the vicinity of a submarine. Likewike, it provides insights on Hg dynamics associated with volcanic and pelagic processes through dissolved mercury speciation analyses. However, to more accurately assess volcanic Hg fluxes monitoring programs should be established and maintained in time.

Acknowledgement

References

Edwards, B. A., Kushner, D. S., Outridge, P. M., & Wang, F. (2021). Fifty years of volcanic mercury emission research: Knowledge gaps and future directions. Science of the Total Environment, 757, 143800. https://doi.org/10.1016/j.scitotenv.2020.143800
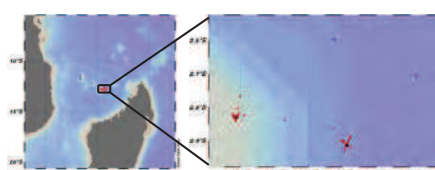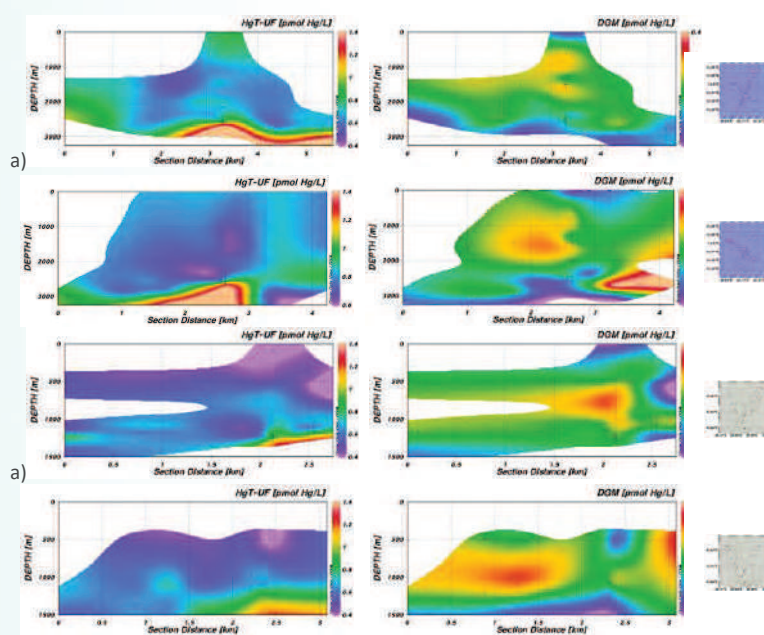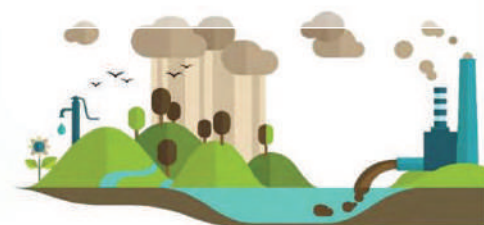
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Use of [197]Hg radiotracer in Hg research

Jan Gačnik,[1,2] Igor Živković,[2] Sergio Ribeiro Guevara,[3] Radojko Jaćimović,[2] Jože Kotnik,[1,2] and Milena Horvat[1,2]

[1]Jožef Stefan International Postgraduate School, Jamova cesta 39, 1000 Ljubljana, Slovenia
[2]Jožef Stefan Institute, Department of Environmental Sciences, Jamova cesta 39, 1000 Ljubljana, Slovenia
[3]Laboratorio de Análisis por Activación Neutrónica, Centro Atómico Bariloche, Av. Bustillo km 9.5, 8400 Bariloche, Argentina

Introduction

- How can we trust worldwide mercury measurements? – Validation of analytical procedures (sampling, preconcentrate-ion, calibration, etc.)
  - Challenges: low ambient concentrations, high blanks, reactive nature of Hg species, tedious procedures and cleaning protocols
  - Solution: [197]Hg radiotracer – high specificity, no blanks, low detection limits, relatively simple detection
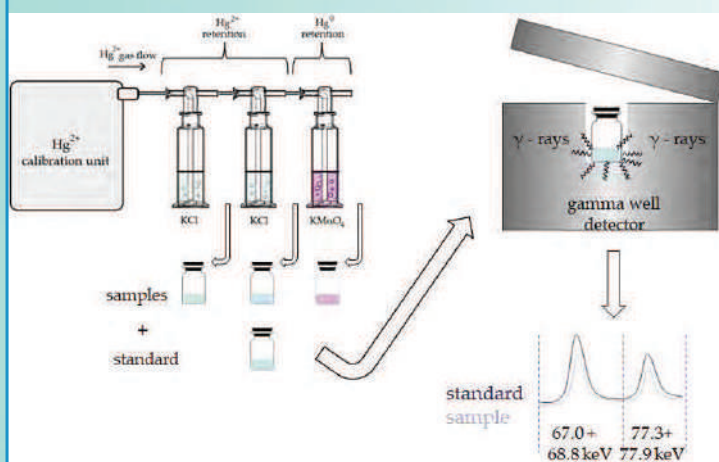
Methods

- Solution of Hg enriched in [196]Hg isotope is irradiated with neutrons in the central channel of the reactor (JSI, Ljubljana)
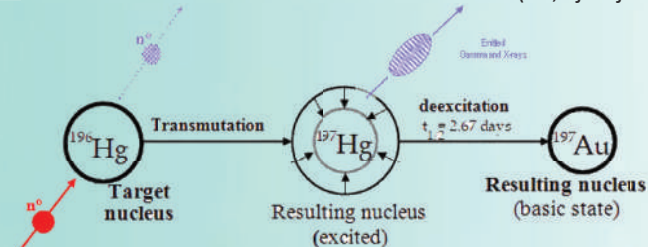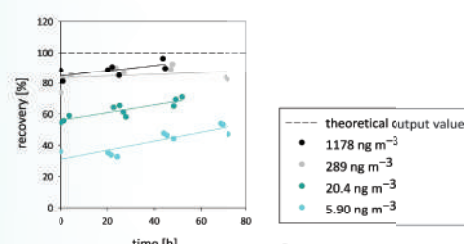

- [197]Hg in solution can be used for studies of all major Hg cycle parts (aqueous, atmospheric and terrestrial Hg)
- Detection of γ- and X-rays is done using HPGe gamma detector (well-type or coaxial-type)
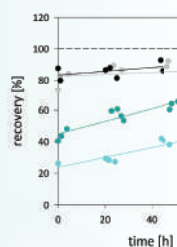
Results and conclusions

- Validating an evaporative calibrator for gaseous oxidized mercury (GOM)
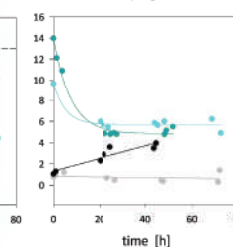

- Conclusions: i) concentration-dependent output, ii) time-dependent output, iii) suitable for ↑conc., not suitable for ↓conc. GOM calibration

- Validating sampling methods for GOM
  - Conclusions: i) KCl sorbent traps: GOM captured is stable, losses can occur at low ambient conc., trap specificity is great, ii) KCl trapping solutions: not suitable for ambient GOM conc

- Validating magnetic nanomaterial for aqueous Hg[2+] analysis
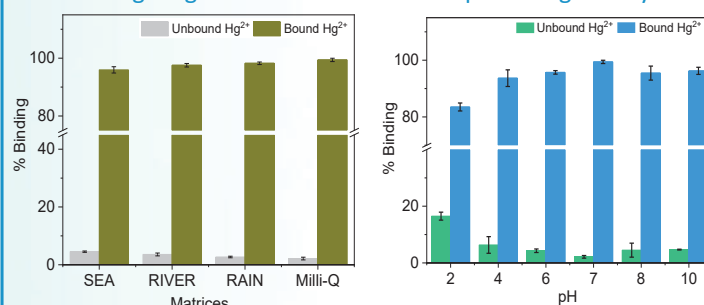
- Conclusions: magnetic nanomaterial suitable for aqueous Hg[2+] analysis, no interferences, optimal working pH value 7

Take-aways

- [197]Hg radiotracer can be used for validation of analytical procedures and for Hg transformation studies in atmospheric, aquatic and terrestrial environments
- In presented work, we successfully applied the said radiotracer for validation GOM measurements and for validation of a newly developed material for aqueous Hg[2+] analysis

Funding

GMOS TRAIN

Exploration of long-term observational datasets to examine ocean-atmosphere exchange processes of Hg

Koketso Molepo[1], Ralf Ebinghaus[1], Aurélien Dommergue[2], Noelle Selin[3]

[1] Helmholtz-Zentrum Hereon, Max-Planck-Str. 1, 21502 Geesthacht, Germany

[2] Université Grenoble Alpes, CNRS, IRD, Grenoble INP, 38400 Grenoble, France

[3] Massachusetts Institute of Technology, Boston, Massachusetts 02139 United States

1. Introduction

Exchange of mercury (Hg) between oceans and the atmosphere is one of the most important drivers of the concentration as well as composition of atmospheric Hg. Yet, the influence of ocean-atmosphere exchange on atmospheric Hg is still poorly understood, largely due to lack of sufficient data. Here, we analyse available long-term gaseous elementary mercury (GEM) observations to investigate influence of ocean-atmosphere exchange on atmospheric Hg.

2. Methodology

We analyzed GEM concentrations from 9 monitoring stations across the globe, as well hourly ^{222}Rn from Cape Point (Fig. 1). For each station, 96-hourly air mass backward trajectories were computed using HYSPLIT for every hour at 50, 200, and 500 m above model ground level. Each air mass was then designated as of marine (>= 90 % of trajectory segments over the ocean), continental (>= 90 % of trajectory segments over continental or island landmass) or mixed origin (the rest) (Fig. 2).

3. Validation of air mass separation method

Comparison of continental-linked ^{222}Rn to marine-linked ^{222}Rn at Cape Point shows generally higher concentrations for the continental-linked ^{222}Rn, with mean concentration of the continental-linked ^{222}Rn (3.10 Bq m^{-3}) ~10 times larger than that of the marine-linked ^{222}Rn (0.36 Bq m^{-3}) (Fig. 3).

4. Results and discussion

The (full) GEM datasets for costal stations Amsterdam Island, Dumont D'Urville, Dome C and Zeppelin show diurnal patterns expected for sites influenced largely by marine-originating GEM (Fig. 4). The pattern is described by a distinct midday peak in GEM, likely linked to sunlight-induced reduction of Hg(II) (and subsequent emission of GEM) from the ocean surface. At the remaining coastal sites, Cape Point, Cape Verde and Mace Head, the pattern is not as well-defined .

Looking at Amsterdam Island (Fig. 5a) and Cape Point (Fig. 5b) as examples, for both sites comparison between the full and marine-linked GEM shows little differences. For Amsterdam Island, this is unsurprising, due to the island's isolation from other major GEM sources (i.e., ocean-atmosphere exchange may be the most important source here). For Cape Point, the results are more difficult to explain; however, these results may also indicate lack of a marine GEM signal at Cape Point.

5. Conclusions

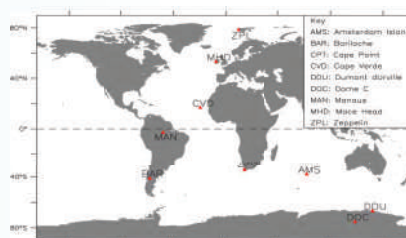(analyses and discussion ongoing)

Acknowledgements

Fig. 1: Locations of the GEM monitoring stations used in this study.
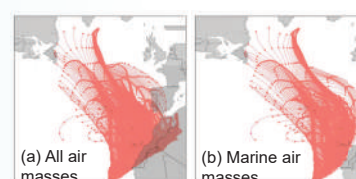
Fig. 2: Illustration of air mass distinguishing method at Cape Verde station for Jan – Feb 2014.
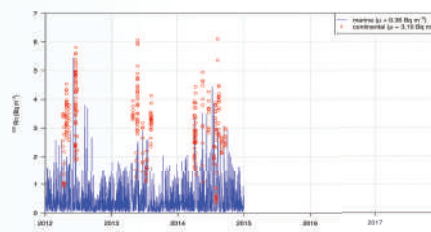
(a) All air masses

(b) Marine air masses

Fig. 3: Hourly concentrations of ^{222}Rn (Bq m^{-3}) for continental- and marine-linked air masses at Cape Point.
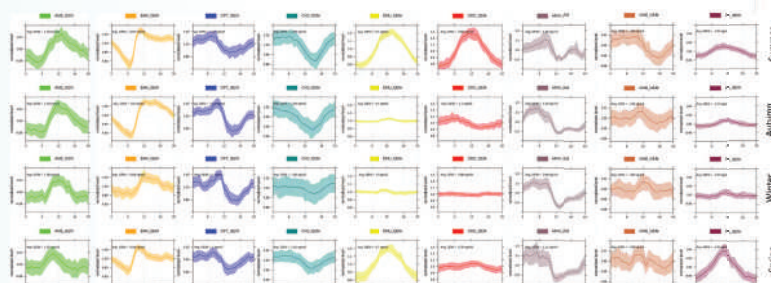
Fig. 4: Diurnal variations of GEM at the nine stations during different seasons, for the full datasets. Note that y-axis is similar across the seasons for each station but different across the stations.

All data   Marine-linked   All data   Marine-linked
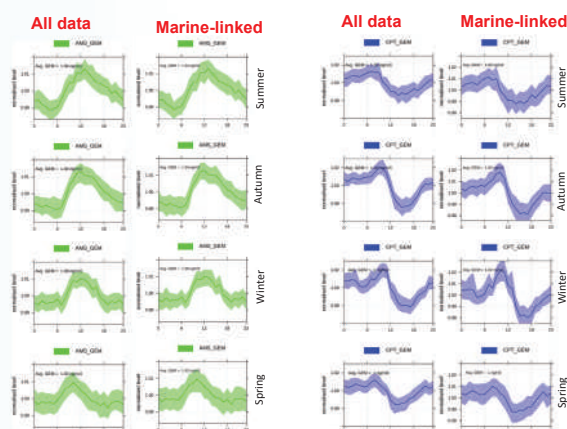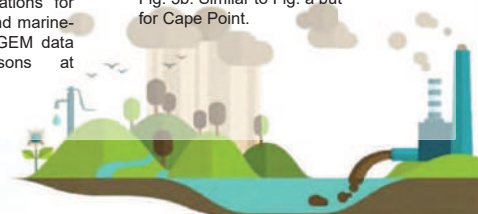
Fig. 5a: Diurnal variations for the full (left panels) and marine-linked (right panels) GEM data for different seasons at Amsterdam Island.

Fig. 5b: Similar to Fig. a but for Cape Point.

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Evaluation of sample preparation methodologies for multi-elemental analysis in foliage samples

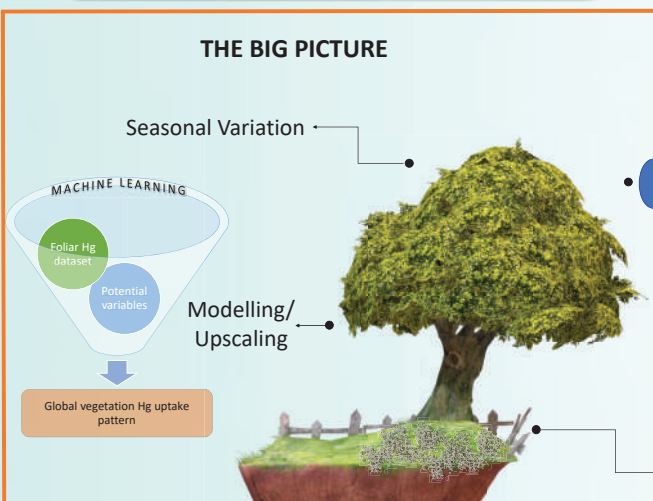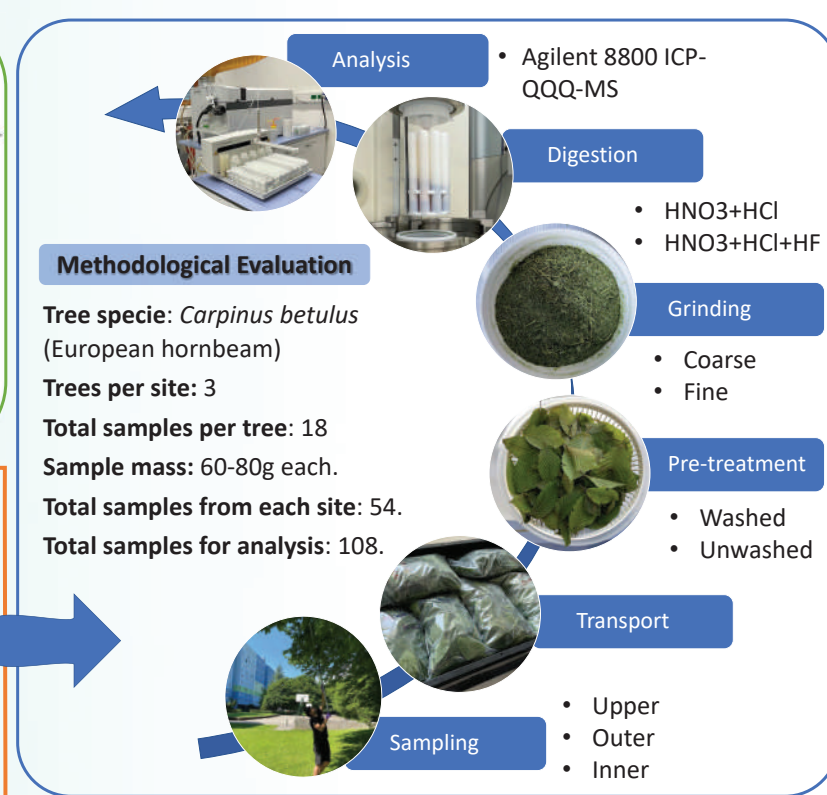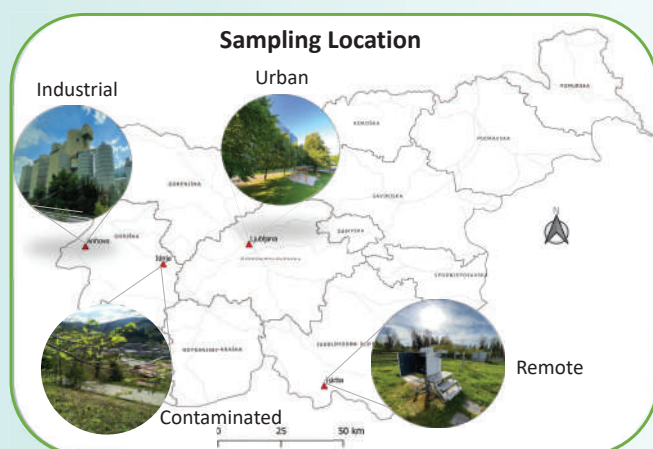Saeed Waqar Ali[§], David Kocman[§], Marta Jagodic Hudobivnik[§], Milena Horvat[§]

[§]Jožef Stefan Institute, Department of Environmental Sciences, Jamova cesta 39, 1000 Ljubljana, Slovenia

Introduction

- Several approaches exist in literature for sample collection, preparation, and quantification of elemental concentration in foliage samples.
- Comparability of results from studies with varying methodological approaches critical for accurate estimation of vegetation control on Hg intercompartmental exchanges.
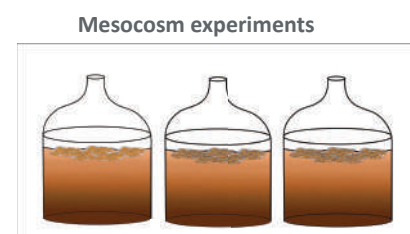
Approach:
- Testing different pre-treatment methods.
- Several combination of digestion and dilution methods are also assessed for their suitability to achieve desirable analytical outcomes.

Sampling Location

Industrial

Urban

Remote

Contaminated

THE BIG PICTURE

Seasonal Variation

MACHINE LEARNING

Foliar Hg dataset

Potential variables

Global vegetation Hg uptake pattern

Modelling/ Upscaling

Litterfall Degradation

Environmental Matrices: Foliage, Soil, Precipitation, Atmosphere measurements.
Parameters: Mercury speciation and fractionation, Hg-isotopic signatures, Multi-elemental analysis.

Methodological Evaluation

Analysis
- Agilent 8800 ICP-QQQ-MS

Digestion
- HNO3+HCl
- HNO3+HCl+HF

Grinding
- Coarse
- Fine

Pre-treatment
- Washed
- Unwashed

Transport

Sampling
- Upper
- Outer
- Inner

Tree specie: *Carpinus betulus* (European hornbeam)
Trees per site: 3
Total samples per tree: 18
Sample mass: 60-80g each.
Total samples from each site: 54.
Total samples for analysis: 108.

Mesocosm experiments

Expected outcomes:
Specific: The extent of variability in analytical outcomes from contrasting methodological approaches.
Overall:
- How changes in foliar mercury content from emergence to senescence in contrasting environmental conditions relate to mercury accumulation in litterfall.
- Improved understanding of mercury kinetics during litterfall degradation.
- Spatial pattern of mercury uptake by vegetation predicted by potential explanatory variability.

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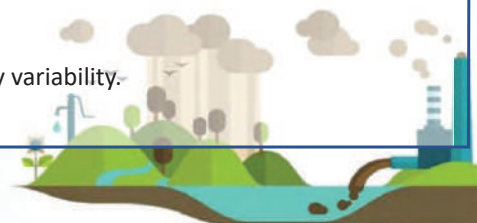
The Role of Dissolved Organic Matter in the Coastal Cycling of Mercury

Sonja Gindorf[a], Sofi Jonsson[a], Lars-Eric Heimbürger-Boavida[b], David Amouroux[c], Elsie Sunderland[d]

[a] Stockholm University, Department of Environmental Science
[b] Aix Marseille Université, Mediterranean Institute of Oceanography
[c] Institut des Sciences Analytiques et de Physico-chimie pour L'Environnement et LesMatériaux, Université de Pau et des Pays de L'Adour
[d] Harvard University, Department of Environmental Health

1 Introduction

Rivers are the most important source of mercury (Hg) for coastal oceans[1]. Dissolved organic matter (DOM) is an important vector for the transport & reactivity of Hg from land to sea. The composition & concentration of DOM have been shown to control Hg bioavailability[2], desorption rates[3], rates of photolytic methyl-Hg (MeHg) degradation & bacterial Hg methylation[4]. Only loosely bound or dissolved Hg compounds will contribute to the Hg pool available for microbial methylation and subsequent biological uptake. We want to answer the question:

What role plays the origin of DOM for coastal Hg transformations?

ELECTRA

2 Study Area
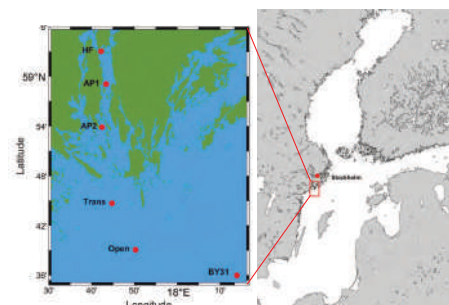
In June, 2021 the first sampling campaign was conducted: a transect from Himmerfjärden Bay (HF) and Stockholm Archipelago (AP1, AP2) towards the open (Trans, Open) and the deepest point in the Baltic Sea Landsort Deep (BY31).

3 Methods

© HORIBA

25 L

30 mL

Waste

DOM Extractions

We characterize DOM using fluorescence spectroscopy for emission excitation matrices to identify terrestrial vs. marine compounds. Additionally, solid phase extractions of DOM along the transect were carried out.

Hg

Next step: Incubation Experiments

1. Add DOM & Hg tracer to water
2. Expose it to sunlight or add biota
3. Measure the Hg species at different time points.

This will help us to determine the influence of DOM composition on the rates of

Water + DOM+ Tracer

Photodegradation
Biological Uptake
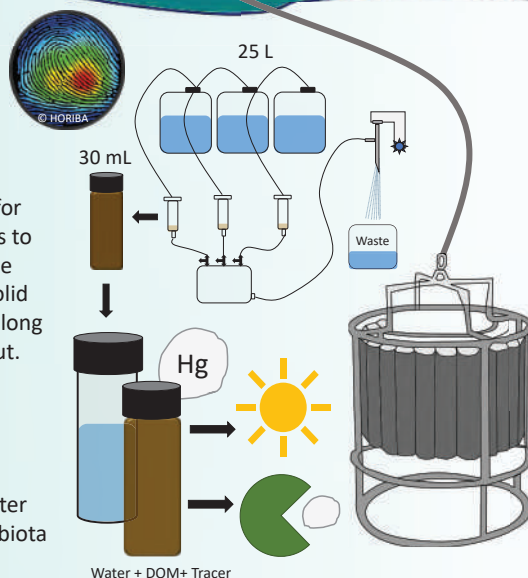
A strong salinity gradient causes a stratified water column and anoxia below the halocline favoring Hg methylation
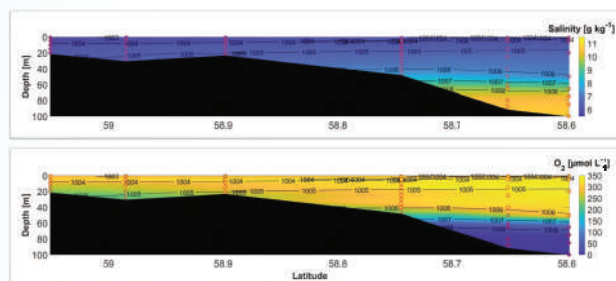
References

[1] Liu, M., Zhang, Q., Maavara, T. et al. Rivers as the largest source of mercury to coastal oceans worldwide. Nat. Geosci. 14, 672–677 (2021). https://doi.org/10.1038/s41561-021-00793-2

[2] Jonsson, S., Skyllberg, U., Nilsson, M. B., Lundberg, E., Andersson, A., & Björn, E. (2014). Differentiated availability of geochemical mercury pools controls methylmercury levels in estuarine sediment and biota. Nature Communications, 5. https://doi.org/10.1038/ncomms5624

[3] Jordan, R. N., Yonge, D. R., & Hathhom ', W. E. (1997). Enhanced mobility of Pb in the presence of dissolved natural organic matter. In Journal of Contaminant Hydrology (Vol. 29).

[4] Jędruch, A., & Bełdowska, M. (2020). Mercury forms in the benthic food web of a temperate coastal lagoon (southern Baltic Sea). Marine Pollution Bulletin, 153. https://doi.org/10.1016/j.marpolbul.2020.110968

Sonja.Gindorf@aces.su.se

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Hg^0 Plasma oxidation experiments, analysis of Hg^{II} species using Quadrupole Mass Spectrometer, and KCl denuder specificity-stability tests

Sreekanth Vijayakumaran Nair[a,b], Jan Gačnik[a,b], Igor Živković[a] and Milena Horvat[a,b]

[a] Jožef Stefan Institute, Department of Environmental Sciences, Jamova cesta 39, 1000 Ljubljana, Slovenia
[b] Jožef Stefan International Postgraduate School, Jamova cesta 39, 1000 Ljubljana, Slovenia

Introduction

Atmospheric Hg-redox reactions play an important role in the global mercury cycle. In general, atmospheric Hg exists as operationally defined GEM, GOM and PBM. Identification of the exact species that comprise GOM in ambient air is challenging due to ultra trace concentration levels at environmentally relevant conditions. We present here the application of ^{197}Hg radiotracer and cold plasma in conducting laboratory Hg^0 oxidation experiments at ultra trace levels and the identification of different Hg^{II} species using Quadrupole mass spectrometer (QMS). Additionally, we evaluated the performance of KCl-coated denuders (used as a standard method for sampling GOM) for better estimation of biases in real-time Hg air speciation measurements, since biases can lead to under/over-estimation of Hg species in the ambient air.

Materials and Method

### Plasma oxidation of Hg^0 with electrolytically produced chlorine

QMS
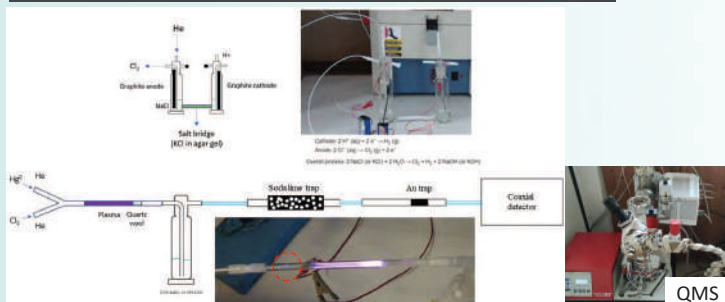
$^{197}Hg^{2+}$ reduced to $^{197}Hg^0$ in presence of $SnCl_2$ → allowed to react with chlorine gas in cold plasma → mass balance checked → qualitative analysis of Hg^{2+} using QMS (compared with thermogram of standards and previous studies). Similar experiment was carried for Hg^0 plasma oxidation with bromine generated by electrolysis of KBr solution.

KCl denuder specificity test
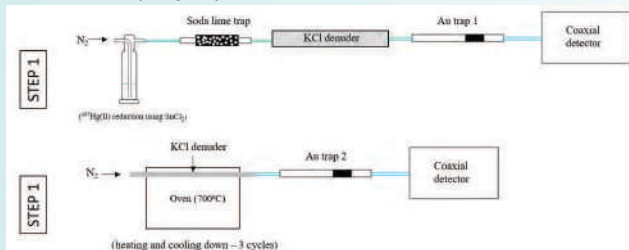
$^{197}Hg^{2+}$ reduced to $^{197}Hg^0$ in the presence of $SnCl_2$ → allowed to pass through KCl denuder and breakthrough was checked. KCl denuder was then heated to check if any Hg^0 was retained in it. Mass balance was created in the end.

### KCl denuder specificity test
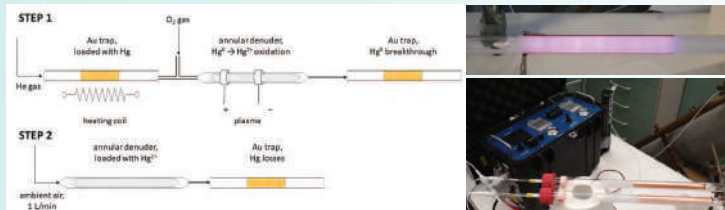
In step 1, Plasma oxidation of Hg^0 with O_2 gas was done and the breakthrough was tested. In step 2, GOM losses from the denuder was recorded in 30 min intervals of exposure to the airflow; losses measured after each exposure interval.

Results and Discussion
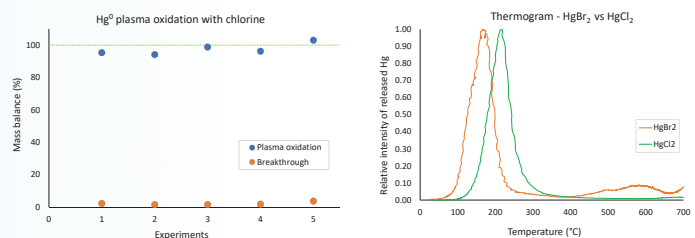
Average:
Plasma oxidation = 97.5 ± 4,0 %
Breakthrough = 2.2 ± 1,0 %
(± shows repeatability SD)

$HgCl_2$ → max peak b/w 217-232°C
$HgBr_2$ → max peak at 164°C
Decomposition: $HgBr_2 < Hg_2Cl_2 = HgCl_2$
Peak range depends on sorbent matrix
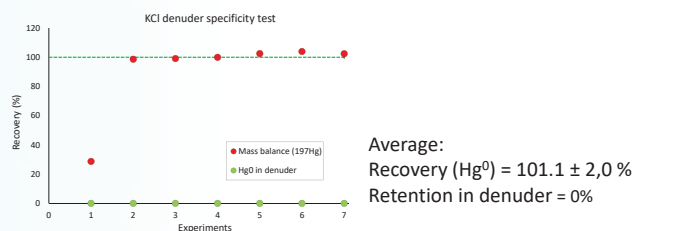
Average:
Recovery (Hg^0) = 101.1 ± 2,0 %
Retention in denuder = 0%

Stability of Hg^{2+} plasma loading on denuder: N_2 flow vs synthetic air vs ambient air flow

Experiment		Denuder number	Hg^{2+} loss in time interval [%] (1L/min flow)		
			0,5 h	1,0 h	2,0 h
Under N_2 flow		1	1,45	1,68	2,63
Synthetic air		4	15,7	16,5	15,8
		5	3,72	2,30	3,74
Ambient air		1	15,6	22,4	32,4
		2	22,0	32,7	41,8
Ambient airflow (Lab 1)	65% RH, 20.8 °C	1	16,8	26,2	32,6
		2	32,2	44,4	54,3
	70% RH, 19.9°C	1	22,8	46,9	79,4
		2	36,4	56,3	66,9
	71% RH, 23,2°C	1	33,6	44,2	53,1
		2	36,6	47,1	59,1
	70% RH, 21,5°C	3	16,8	28,1	33,6
		4	16,9	23,3	29,4
Ambient airflow (Lab 2)	65% RH, 20,8 °C	2	1,82	3,85	9,50
		5	4,30	7,53	10,9
	68% RH, 21,0°C	4	8,26	11,0	17,2
		5	14,8	15,3	20,0
Ambient airflow (Outside)	43% RH, 25,1 °C	2	69,3	74,4	77,3
		5	41,9	46,3	49,1
	41% RH, 24,3°C	2	77,4	78,8	79,7
		5	34,2	33,8	35,6
Ambient air (Outside + PM filter)	49% RH, 20,0°C	4	55,4	66,4	71,0
		5	63,6	73,4*	63,8

- Losses are different when exposed to ambient air, synthetic air and N_2 flow.
- Losses are higher in the first two cycles (1h).
- Cumulative losses in 2h are between 29-80 % for ambient air.
- Much higher losses in outside lab-air.

Take away points

The use of ^{197}Hg radiotracer is highly specific and efficient in conducting studies at trace levels. QMS can be used for qualitative identification of laboratory generated oxidized Hg species. Present laboratory tests show KCl denuder is specific to Hg^{II} species and does not retain Hg^0. However, the efficiency of KCl denuder in sampling GOM species is questionable since Hg^{2+} losses are higher when exposed to ambient air that leads to speciation biases (GOM biased low) and losses were very low when exposed to synthetic air and N_2 flow. Moreover, losses were different for different denuders and varies for the same denuders depending upon whether they are freshly prepared or not. Reusing KCl denuders for longer periods reduces their efficiency at retaining Hg^{2+}. It was observed that losses from KCl denuders are mostly in the form of Hg^0. This implies that Hg^{2+} loaded in the denuder is unstable when exposed to ambient air, and complex Hg transformations occur in the denuder after sampling under constant flow. This could be due to the presence of ozone and water vapour in ambient air leading to Hg^{2+} reduction as Hg^0 which has to be further evaluated. A similar study will be performed for ion exchange membranes.

GMOS TRAIN

Jožef Stefan Institute
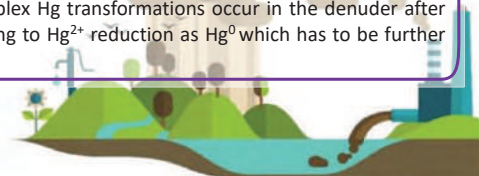
Tackling the uncertainty of using gas calibrators for GEM that use the Dumarey equation

ESR 10: Teodor Daniel Andron; Supervisor: Prof. Dr. Milena Horvat; Co-supervisor: Dr. Warren Corns

˝Jožef Štefan˝ Institute, Ljubljana, Slovenia

P S Analytical, Orpington, England

Teodor D. Andron, Milena Horvat, Warren T. Corns, Igor Živković, Jože Kotnik

Introduction

Calibration is the procedure through which we can make our results traceable. Through traceability we can improve the comparability of our measurements and their uncertainties. One way to do this is by using a gas standard calibrator unit which is based on the Dumarey equation, which calculates the saturation of mercury in air at different temperatures. It has been proven that the equation can be used for higher masses (>10 ng of Hg^0) with a relative uncertainty of 2% (Dumarey et al., 2010), but it has been reported in the literature that there might be some discrepancies between the theoretical Hg^0 that the Dumarey equation shows, and the actual mass that these gas standard calibrators give (De Krom et al., 2021; Quétel et al., 2016). Through this work, we wanted to verify this difference by tracing the amount of mercury from the gas calibrator (SI units) to the NIST 3133 CRM (international standard).
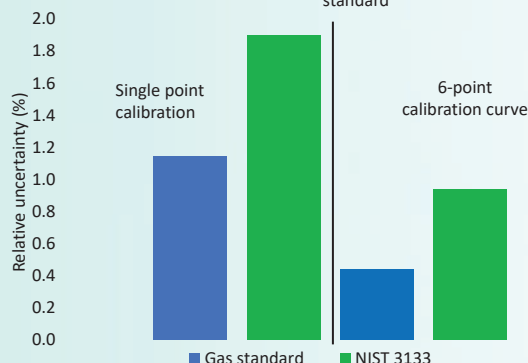
Materials and methods
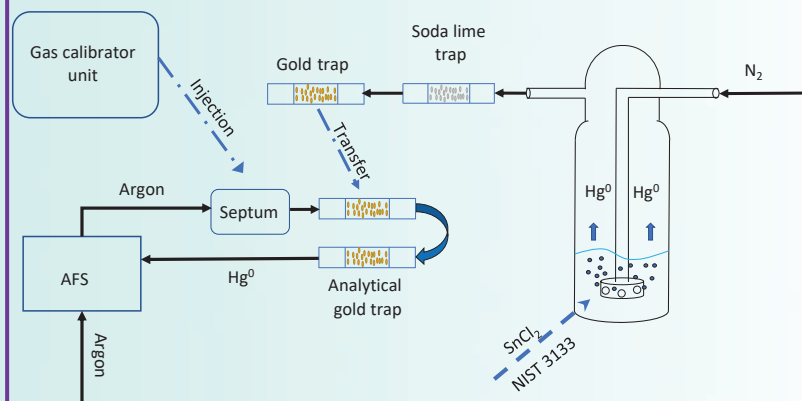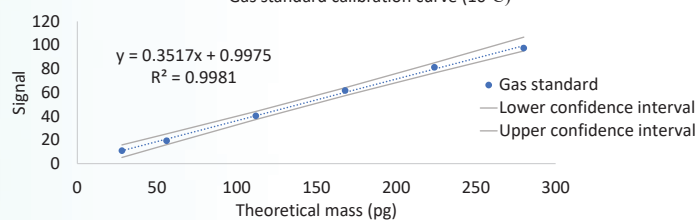

Results and discussions

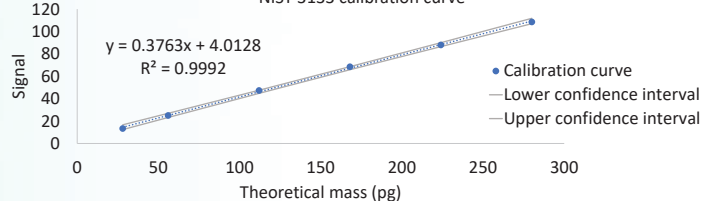Hg in the gas standard at different temperatures determined by NIST 3133

■ Mass of Hg in the gas standard   ── Combined uncertainty range
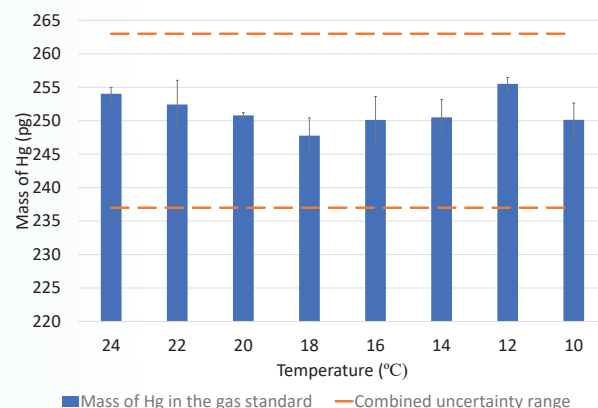
Relative uncertainty of the gas standard and the NIST 3133 standard

■ Gas standard   ■ NIST 3133

Gas standard calibration curve (10°C)

$y = 0.3517x + 0.9975$
$R^2 = 0.9981$

● Gas standard
── Lower confidence interval
── Upper confidence interval

NIST 3133 calibration curve

$y = 0.3763x + 4.0128$
$R^2 = 0.9992$

● Calibration curve
── Lower confidence interval
── Upper confidence interval

Conclusions

Based on our results we conclude that calibration at ambient concentration levels using gas standards based on Dumarey equation and NIST 3133 provide comparable results.

References

Dumarey, R., Brown, R. J. C., Corns, W. T., Brown, A. S., & Stockwell, P. B. (2010). Elemental mercury vapour in air: the origins and validation of the "Dumarey equation" describing the mass concentration at saturation. Accreditation and Quality Assurance, 15(7), 409–414. doi:10.1007/s00769-010-0645-1

Quétel, C. R., Zampella, M., & Brown, R. J. C. (2016). Temperature dependence of Hg vapour mass concentration at saturation in air: New SI traceable results between 15 and 30°C. TrAC Trends in Analytical Chemistry, 85, 81–88. doi:10.1016/j.trac.2015.12.010

De Krom, I., Bavius, W., Ziel, R., Efremov, E., van Meer, D., van Otterloo, P., … Ent, H. (2021). Primary mercury gas standard for the calibration of mercury measurements. Measurement, 169, 108351. doi:10.1016/j.measurement.2020.10