

Speciation and Net Methyl Mercury Formation in Mercury Contaminated Sediments

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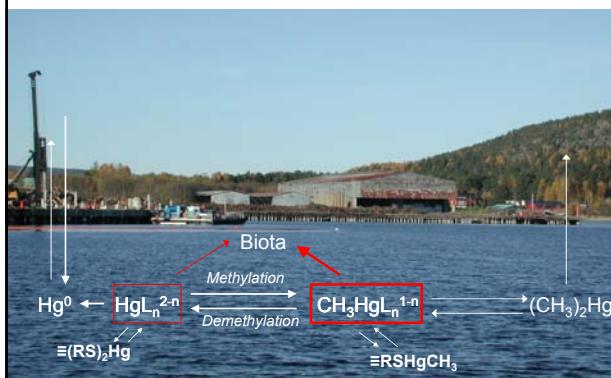


Mercury contaminated sediments



- Phenylmercury acetate, pulp industry -1941-1967
- Elemental mercury (Hg^0), chlor-alkali industry -2010

Mercury biogeochemistry



Sites



Site characteristics

Site and Date	C	C/N	C/S	Fe	Total Hg	Total MeHg	Cl
	(%)	(%)	(%)	(nmoles /g dw)	(pmoles /g dw)	(mM)	
Köpmaholmen Oct 2004	3.7-23	30-1286	6-55	0.3-2.6	1.0-1143	4.4-387	81-88
Köpmaholmen Sept 2005	1.7-29	14-450	3-84	0.5-1.9	8.0-1104	19-328	80-86
Skutskär Dec 2004	7.3-19	24-59	4-13	1.1-4.9	15-404	8.3-297	24-93
Skutskär June 2005	13-20	24-59	6-7	3.7-4.4	72-233	338-575	80-85
Karshäll March 2005	29-39	36-68	47-59	1.3-2.3	14-17	13-106	0.53-2.0
Karshäll Sept 2005	29-44	37-74	49-74	1.0-2.3	3.9-36	27-300	0.19-0.55
Nötöfjärden June 2005	22-39	20-57	12-24	0.8-2.1	5.1-9.6	33-258	0.36-0.83
Marnästjärn June 2005	15-17	9-10	7-9	4.0-4.2	85-138	216-476	0.43-0.72
Turingen June 2005	5.0-6.0	10-12	14-17	3.2-4.4	5.9-23	16-26	0.58-0.65
Svartsjöarna June 2005	28-30	16-17	22-30	2.8-4.5	2.7-5.2	66-183	0.21-0.24

Methylation: measurement and theory

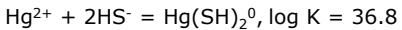
Methylation determination:
Addition of $^{201}\text{Hg}^{2+}$ and incubation for 48 h in a glove-box (N_2).
Methylated $^{201}\text{Hg}^{2+}$ measured.

- Sulfate Reducing Bacteria (SRB) are the main methylators in sediments (e.g. Compeau and Bartha, Appl. Env. Microbiol., 1985;50)
 - stimulated by SO_4^{2-} , fresh organic C (e-**donor**), temperature, need access to Hg^{2+}
 - Hg^0 controls uptake in laboratory experiments with methylating bacteria (e.g. Benoit et al., ES&T, 2001;35)
 - accidental side reaction, enzyme-mediated donation of a methyl group from methylcobalamin: passive uptake over the cell membrane

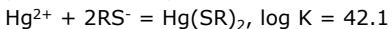
Speciation: models A, B and C



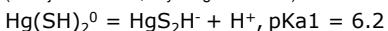
(Dyrssén and Wedborg 1991)



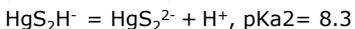
(Smith and Martell 1976)



(Khwaia et. al. 2006, Skjellberg et. al. 2000)



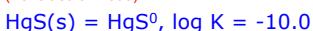
(Smith and Martell 1976)



(Smith and Martell 1976)



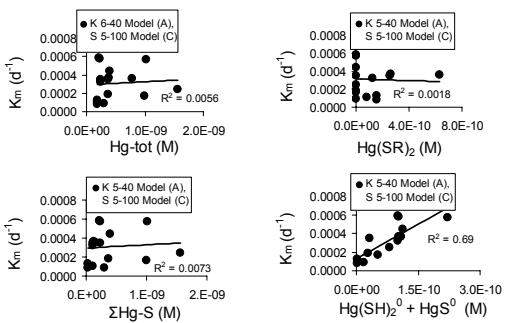
(Benoit et. al. 1999)



(Dyrssén and Wedborg 1991)

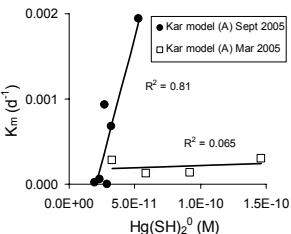
Speciation and methylation

Köpmannholmen and Skutskär:



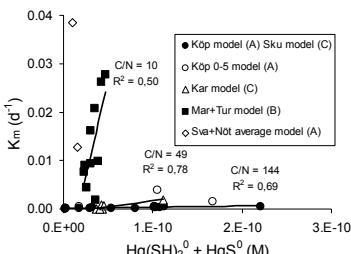
Speciation and methylation

Karlshäll September (circles) and March (squares) 2005:



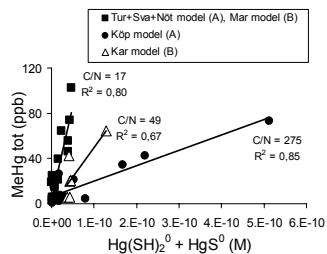
Speciation and methylation

Köpmaholmen and Skutskär (circles), Karlshäll (September, triangles), Marnästjärn and Turingen (squares), Svartsjöarna and Nötöfjärden (diamonds):

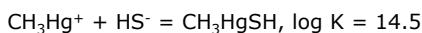


Speciation and total methyl mercury

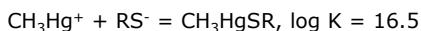
Turingen, Ö. Svartsjön, Nötöfjärden and Marnästjärn (squares), Karlshäll (triangles, September), Köpmaholmen (circles):



Methyl mercury speciation model



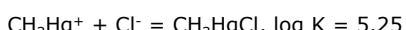
(Dyrssen and Wedborg 1991)



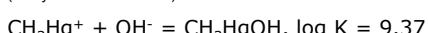
(Karlsson and Skyllberg 2003)



(Dyrssen and Wedborg 1991)

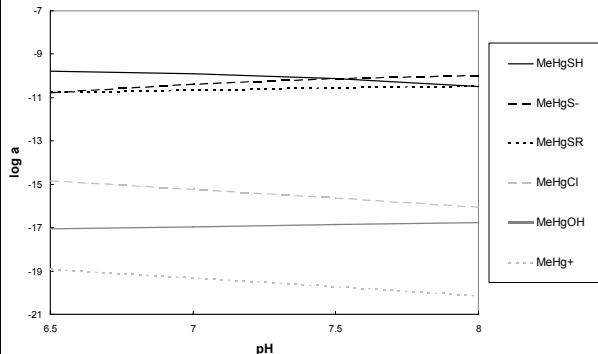


(Carty and Malone 1979)



(Smith and Martell 1976)

Methyl mercury speciation



Major conclusions

- Passive uptake of Hg^{2+} as neutral species in methylating bacteria is supported
- Availability of Hg^{2+} (as neutral species) and fresh organic carbon (e-donor) are the main factors controlling methylation rates in contaminated sediments
- Methylation rates largely determine the concentration of MeHg in contaminated sediments
- The solubility of MeHg in anoxic sediments is mainly determined by the concentrations of inorganic sulphides and DOC in the sediment pore water

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