EXPERIMENTAL STUDY OF DIFFUSIONAL AND FILTRATIONAL MIGRATION OF MERCURY MOBILE FORMS AND COLD EMISSION ENDOGENIC FLOW

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Different series of experiments aimed to study migratory properties of mercury in liquid and gaseous phases through natural samples with known composition were set. There were dynamic experiments with gaseous and liquid phases and static experiments studying loam and sand adsorbing capacity. In these series we used standard parameters of rock mass with mercury concentration both closed to anthropogenic and also with partial pressure of saturated mercury vapour as a boundary condition. We obtained a number of kinetic parameters and adsorbing properties for sands and orgillaceous soils as main natural barriers for mercury transport to surface. The mercury migration and adsorption did not take in consideration for humusic soil as it situated in permanent dynamical change with atmospheric air. All experiments conducted with subsoil rocks with depth more than 1 m with very low humus concentrations as sorbent for exception emission – reemission flows from upper soil layers. It was determined that the main mechanism of sorption in solution (both for static and dynamic experiments) is chemical sorption, caused by reactions of precipitation oxidational mercury forms at the phase interface. It was found that the main transport mechanism for Hg^0 vapor in dry soil samples is gaseous diffusion, but in wet soils the limiting stage of adsorption is diffusion through the liquid membrane to the grain surface (fig. 1).

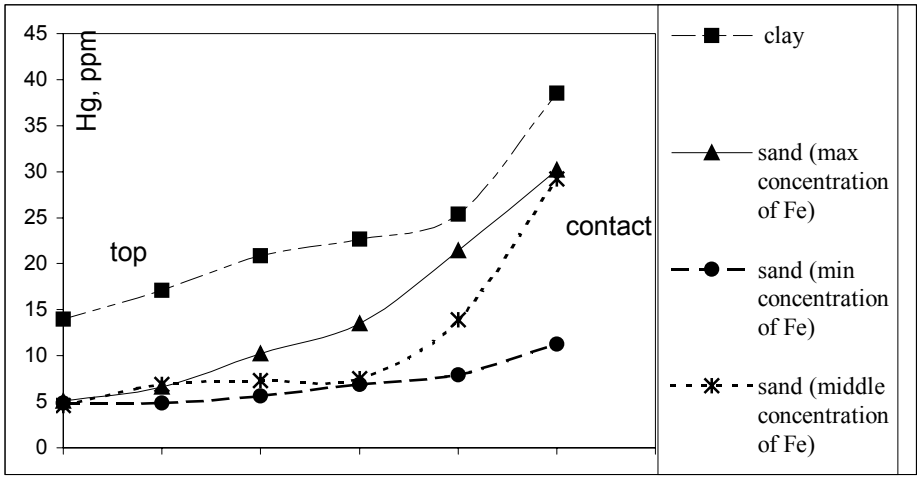

Fig.1. Mercury diffusion concentration profiles for samples with liquid phase

After level-by-level analysis of experimental samples all concentration profiles allowed us to determine limiting stage and mechanism of proceeding process. At this time the character of concentration profiles developed in porous transport medium is extremely specific. It depends on the main transport mechanism and also on the limiting stage of migration and adsorbing capacity of the transport medium, what influences on effective diffusion coefficient (D_{ef}) value. This coefficient was calculated by using the equation for density of mercury flow $J = (D_{ef} * \mathrm{grad}C_{Hg}$, where $\mathrm{grad}C_{Hg} = \Delta C/l = \Delta C/(\xi * h)$, (where l - length of a way in view of the factor of tortuosity ξ at height of a column h). At the same time we pioneer to evolve the method on determination of local partial Hg vapour pressure in void volume of rock mass. For this purposes we compare initial amount of adsorbing Hg (defined by stepped thermo evacuation) with amount of Hg in samples which were in adsorption equilibrium with the saturated Hg vapour.

Besides this we set several series of atmospheric and pore air pumping from drawned-out wells in Baikal rift zone and Baikal steppes, and also from the rocks from Russian platform (tab. 1). This pumping was conducted with Hg amalgam precipitating at the specific sorbent we synthesized.

Table 1.

Results of atmospheric and pore air pumping

№	Place and period of pumping	Air	Concentration of Hg, ng/m^3
1	Lake Baikal, shore of the bay M.Orso, august 2002y.	Atm.	17.5
2	Lake Baikal, bay M.Orso, break zone, august 2002y.	Well	150
3	Lake Baikal, steppe in Priolhonie, august 2002y.	Well	5.7
4	Lake Baikal, bay M.Orso, near break zone, august 2002y.	Well	38
5	t. Usolie-Sibirskoe, over liquidated mercury electrolise department of chemical factory, august 2002y.	Atm.	12060
6	t. Usolie-Sibirskoe, forest, 1.5 km far from chemical factory, august 2002y.	Atm.	21.8
7	t. Usolie-Sibirskoe, forest, 1.5 km far from chemical factory, august 2002y.	Well	234
8	Bratsk reservoir, from t.Balagansk to r.Belaya, august 2002y.	Atm.	22,5
9	t. Balagansk, Irkutsk region, august 2002y.	Well	6,0
10	c.Irkutsk, at height 10m, august 2002y.	Atm.	32,1
11	c. Moscow, MSU, at height 70m, June 2002y.	Atm.	58,4
12	c. Moscow, MSU, at height 0.5m, June 2002y.	Atm.	300
13	c. Moscow, MSU, at height 0.5m, April 2002y.	Atm.	1,8
14	Pumping in moving at automobile highway c. Moscow – t. Petushki, December 2002 y. (without aerosol)	Atm.	2,9
15	Pumping in moving at automobile highway c. Moscow – t. Petushki, December 2002 y. (with aerosol)	Atm.	35,3
16	MSU Main building lab, April 2003y.	Atm.	1,4-3,6
17	Bladimir region, Meschera, July 2002y.	Atm.	5,2
18	Bladimir region, Meschera, July 2002y.	Atm.	6,5
19	Bladimir region, Meschera, July 2002y.	Well	15,5
20	Bladimir region, Meschera, September 2002y.	Atm.	3,15
21	Bladimir region, Meschera, December 2002y.	Atm.	1,8
22	Bladimir region, Meschera, December 2002y.	Atm.	1,3
23	Bladimir region, Meschera, February 2003y.	Atm.	4,8
24	Bladimir region, Meschera, June 2003y.	Atm.	2,1
25	Bladimir region, Meschera, June 2003y.	Well	5,75
26	Bladimir region, Meschera, June 2003y.	Well	7,8
27	Italy, t. Bari, March 2003y.	Atm.	2,6

Data of table demonstrate, that regional natural background is stable for definite time of the year and weather condition. For dry smoke summer atmosphere of 2002 year for the Baikal region was stable background at 17,5; 21,8; 22,5 ng/m^3 and 5.2-6.5 for lowland of Meschera. At this background we can see pollution of city territory (Irkutsk – 32.1, Moscow – 58.4-300 with vertical distribution). At the same time for humid period of the year (autumn – spring) 2002-2003 years for Meschera and Moscow the concentration of Hg at atmospheric air is near concentration of Europe atmospheric air (1.3-3.2 ng/m^3). At March 2003 in Bary, Italy, on our data, concentration of atmospheric air Hg was 2.6 ng/m^3.

Local anthropogenic sources of pollution are not influence practically on regional background (tab. 1, row 5, 6). At the linear sources of pollution (automobile highway) principal quantity of the Hg are in the aerosol (tab. 1, row 14, 15).

The pore air from drawned-out wells in Baikal rift zone contain more Hg than other wells of this region, that endogenic emission process demonstrate (tab. 1, row 2). For the platform territory (Russian platform, Meschera) pore air concentration more, than atmospheric background at 2.5-3 times.

The comparisons of experimental vapour pressure data with the data, obtained from the pumping of pore and atmospheric air allow us (for some regions) not only estimate man-made contribution and also determine the flux of natural endogenous gaseous emission. As a result, it was estimated a molal stream of mercury (only cold endogenic emission) through a surface of globe: 3, $7*10^7$ mol/year. In mass expression - 7, 4 thousand tons mercury per year, that is much higher than possible world (global) annual industrial extraction of mercury. At the same time this estimation is much higher, than that of Nriagu (2,5 thousand tons/year), and comes nearer to estimations of Tornton (18 thousand tons /year) and Rasmussen (35 thousand tons /year), which other flows taken in consideration (volcanic and antropogenic emission).

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