Supplementary Material

"Environmental Partitioning, Spatial Distribution, and Transport of Atmospheric Mercury (Hg) Originating from a Site of Former Chlor-alkali Plant"

S1. Detailed description of input parameters for fugacity models

For the Level I model, the input included chemical properties of Hg (same input was used for QWASI: molecular weight, Log K_{ow}, melting point, water solubility, vapor pressure, water temperature); environmental parameters, i.e., volumes and densities of all compartments - air, aerosol, water, suspended particulate matter (SPM), fish, soil and sediments; organic carbon (OC) in soil, sediments, SPM; fish lipid content and input Hg mass (in kg). The lake depth and area were taken from [1], fish lipid content was calculated from the real fish types reported in [2]; for everything else, the values were adapted from [3], and the volumes of the segments were adjusted to be proportional to the lake water volume. Since the Level I model is the only to take into account the soil compartment, the total Hg input was taken as 1000 t (total Hg mass discharged into the environment from the plant during the whole operating period [1, 4]), whereas in other models only direct input of Hg to the lake was considered - 135,400 kg [5]. The outputs of models can be also found in Table 2 of the manuscript.

For the QWASI, the variables were similar to Level 1 in terms of chemical parameters of Hg (and degradation half-lives in water and sediments); however, environmental parameters were different since this model does not include soil and sediments, but rather focuses on the lake parameters - surface area, mean depth, volume, active sediment layer depth, density and concentration of the solids in lake and inflow water, aerosol and sediment, and OC fraction in those. For deposition, burial, and resuspension rates of the sediments, atmospheric deposition parameters, and mass transfer coefficients (MTC), HERMES default values were used as input.

Modified input variables in the HERMES model for Balkyldak simulation were: water temperature, lake size parameters (i.e., surface area, mean depth, and volume), precipitation rate, the concentration of Hg in air, water inflow rate, and its Hg concentration, as well some densities and organic carbon fractions in the compartments. Other parameters were assumed default since the data from the real site was not available.

Finally, some of the input parameters were selected based on the assumptions, but in order to obtain more reliable results, additional runs with modified parameters for all models were performed. Sensitivity analysis might be helpful in understanding which factors and/or parameters affect the fugacity the most.

- The concentration of SPM in the water: the default value of 0.463 mg/L in the HERMES was adapted from the measurement in Harp Lake in Ontario, Canada [6]. While in the generic environment described in [3], SPM concentration in the water was approximately 5 mg/L, so additional models were simulated with this value in all three programs.
- According to [5], Hg concentrations in the near-earth air on the territory of the chemical plant measured in 2006 were in the range of 100-1600 $ng/m^{3,}$ and the value of 300 ng/m^3 was exceeded in 7 out of 16 points; but during the field trips in 2018-2019, the average Hg concentration in air around lake Balkyldak was 17 ng/m^3. So, QWASI and HERMES models were additionally simulated with 300 ng/m^3 Hg air concentration.
- Since Balkyldak is a settling lagoon formed specifically for industrial waste, there is no inflow from any river, only wastewater discharge from the PCP. The average wastewater discharge rate during the years of operation was approximately 20–25 m^3/h, and Hg concentrations in the outflow water usually were in the range of 15-40 mg/L [1]. In QWASI and HERMES models, non-zero (river) inflow rate was taken as 22.5 m^3/h; however, since current wastewater discharge is unknown, additional runs were performed with a very low (assumed) value of 0.001 m^3/h.

Table S1. Semivariogram parameters and prediction error statistics

Parameters	Campaign 1	Campaign 2	Campaign 3	Campaign 4
Semivariogram parameters				
Model parameter	1.95078	2	2	0.2
Nugget	0	0.01131	0.06480	0.13323
Major range	0.01828	0.03000	0.04922	0.01431
Partial sill	0.12014	0.00737	0.11708	0.10014
Lag size	0.00229	0.00375	0.00615	0.00179
Number of lags	12	12	12	12
Cross validation				
Regression function	0.698 * x + 3.65	0.104 * x + 6.96	0.321 * x + 2.82	0.196 * x + 3.08
Mean	-0.20055	-0.01658	-0.00457	0.08829
Root-mean-square	3.74285	0.87899	1.57385	1.90173
Mean standardized	-0.00788	-0.01308	-0.02072	0.01868
Root-mean-squared standardized	0.88109	0.91889	1.06798	0.66235
Average standard error	4.32538	1.00121	1.41896	2.60418

Table S2. Field measurement results of ambient TGM in Pavlodar.

No.	Campaign 1: 23.07.2019				Campaign 2: 24.07.2019				Campaign 3: 25.07.2019				Campaign 4: 26.07.2019			
	Latitude	Longitude	Local Time	TGM, ng/m^3	Latitude	Longitude	Local Time	TGM, ng/m^3	Latitude	Longitude	Local Time	TGM, ng/m^3	Latitude	Longitude	Local Time	TGM, ng/m^3
1	52.3928	76.96555	11:36	22	52.40811	76.86362	10:12	9	52.28409	76.95283	21:38	5	52.30914	76.94105	11:36	4
2	52.3938	76.9155833	11:46	22	52.44476	76.86619	10:18	8	52.29155	76.95177	21:41	5	52.32988	76.92821	11:39	5
3	52.39785	76.9135	11:55	16	52.44735	76.88436	10:21	8	52.29844	76.94952	21:44	6	52.34356	76.92787	11:42	5
4	52.4130167	76.9118833	12:18	15	52.4494	76.89952	10:23	8	52.30469	76.92876	21:52	3	52.36762	76.92739	11:44	3
5	52.4307833	76.9118833	12:37	15	52.45277	76.90614	10:26	9	52.32397	76.92836	21:57	4	52.38339	76.92701	11:47	5
6	52.4303667	76.9110333	12:53	15	52.4498	76.86163	10:35	9	52.34492	76.92784	22:00	4	52.38359	76.94883	11:49	6
7	52.4384	76.9118167	12:58	14	52.46082	76.87165	10:37	8	52.36765	76.92736	22:04	3	52.39096	76.96494	11:52	6
8	52.4409333	76.9169	13:01	13	52.46382	76.88918	10:40	7	52.38343	76.92834	22:07	6	52.38322	76.90128	11:59	6
9	52.4444667	76.9299833	13:24	30	52.46751	76.95386	10:55	7	52.38367	76.95243	22:11	8	52.38299	76.87693	12:02	5
10	52.4431667	76.9323	13:44	24	52.47342	76.95298	11:18	9	52.389	76.96503	22:14	6	52.3671	76.88286	12:07	2
11	52.4355	76.9628833	14:20	22	52.47609	76.96676	11:25	8	52.39013	76.96493	22:16	12	52.34594	76.89311	12:09	3
12	52.4407667	76.9724833	14:29	26	52.4768	76.9929	11:40	7	52.38323	76.90302	22:24	6	52.32518	76.90469	12:11	4
13	52.4400333	76.9854833	14:34	27	52.47844	76.99763	11:53	7	52.38297	76.87589	22:32	1	52.30982	76.92755	12:15	2
14	52.4391	76.9981167	14:52	17.5	52.46622	76.99795	12:07	8	52.36642	76.88319	22:35	3	52.30808	76.96002	12:18	2
15	52.4365	77.0068333	15:23	32	52.46369	76.99763	12:20	7	52.34496	76.89363	22:37	2	52.31859	76.96252	12:19	2

16	52.4288167	77.0063167	15:34	37	52.4637	76.99246	12:32	9	52.32357	76.90673	22:40	2	52.32161	76.96251	12:21	3
17	52.42155	76.9973833	15:46	15	52.46276	76.97564	12:55	9	52.3098	76.92864	22:44	2	52.29777	76.96494	12:31	4
18	52.4166333	76.9982833	15:58	12	52.46191	76.96518	13:10	9	52.30812	76.9613	22:47	2	52.29877	76.97247	12:33	5
19	52.4107	77.0133	16:10	13	52.46498	76.9519	13:22	7	52.3209	76.96254	22:50	2	52.29801	76.98605	12:35	3
20	52.4042	77.0181667	16:17	11	52.45961	76.96199	13:33	7	52.32743	76.96239	22:52	3	52.29871	77.00236	12:37	2
21	52.40025	77.02465	16:22	13	52.46143	76.97348	13:44	7	52.30126	76.96271	23:06	3	52.30165	77.02464	12:40	3
22	52.3863333	77.0234833	16:32	9	52.43721	76.99004	14:10	6	52.29675	76.95117	23:06	3	52.29223	77.02632	12:41	2
23	52.3841833	77.00785	16:34	9	52.43743	76.98412	14:30	6	52.29404	76.93995	23:10	4	52.28936	77.00776	12:43	3
24	52.3738667	77.0280167	16:40	9	52.43786	76.97653	14:46	6	52.29008	76.94066	23:13	3	52.28765	76.98788	12:46	2
25	52.37555	77.04275	16:43	8	52.43726	76.94865	15:21	8	52.29123	76.96252	23:16	4	52.28406	76.96762	12:50	3
26	52.37425	77.0506833	16:44	9	52.43827	76.93742	15:30	8	52.29147	76.97183	23:18	5	52.28133	76.95644	12:52	3
27	52.3692667	77.0516667	16:46	11	52.4212	76.91217	16:10	8	52.29857	76.97353	23:20	5	52.28086	76.94207	12:55	3
28	52.35995	77.0553167	16:48	14					52.29812	76.98652	23:22	5	52.27536	76.94192	12:57	3
29	52.3474	77.0521833	16:51	13					52.29887	77.00354	23:24	5	52.27549	76.95825	13:00	4
30	52.3289833	77.0333167	16:53	7					52.30177	77.02505	23:26	5	52.27615	76.97656	13:04	4
31	52.3219333	77.0259667	16:54	5					52.29224	77.02632	23:27	5	52.27139	76.98376	13:06	2
32	52.31285	77.0204667	16:55	4					52.28922	77.00665	23:29	6	52.26525	76.98315	13:08	3
33	52.3023833	77.02495	16:56	4					52.28762	76.98553	23:32	6	52.26449	76.96522	13:10	3
34	52.2911667	77.0218	16:58	6					52.28722	76.97272	23:35	5	52.28653	76.96458	15:26	5

35	52.2881833	76.9989167	17:00	5					52.2841	76.96877	23:38	5	52.28619	76.93777	15:40	4
36	52.28765	76.9878667	17:02	9					52.28128	76.95541	23:40	5				
37	52.28045	76.9875833	17:04	10					52.28082	76.94165	23:43	5				
38	52.2763	76.9769333	17:07	8					52.27536	76.94199	23:44	4				
39									52.27557	76.95911	23:48	3				
40									52.2765	76.98156	23:51	3				
41									52.2704	76.98392	23:52	3				
42									52.26527	76.98246	23:54	3				
43									52.26447	76.96342	23:57	2				

Table S3. Model compartment dimensions for Level I fugacity modeling

Medium	Volume (m^3)	Notes
Air	1.00E+11	100 km^2 x 1 km [3]
Aerosol	2.00E+00	V_{air} x 2 x 10^{-11} [3]
Water	6.75E+07	Lake area x mean depth [1]
Suspended particles	3.38E+02	0.463 ppm [6]
Fish	6.75E+01	1 ppm in water [3]
Soil	6.43E+05	Water/soil area = 70:30 [7]
Sediments	1.50E+05	1 cm [3] x lake area

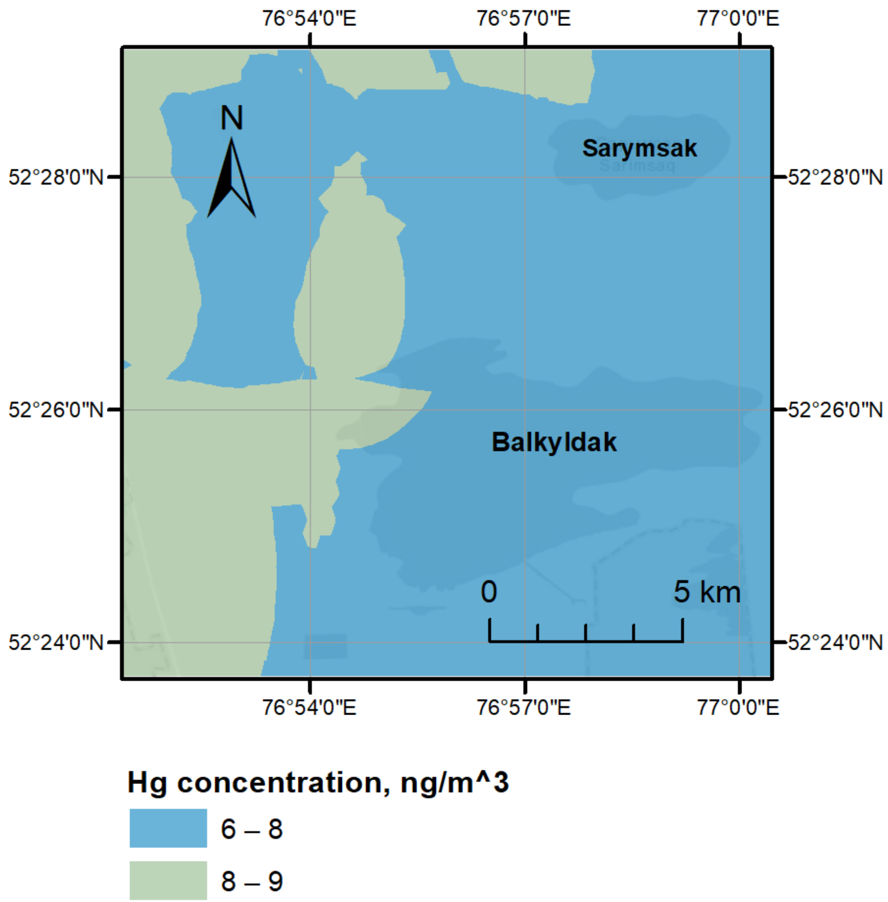
Table S4. Input parameters for QWASI model

Lake surface area (m^2)	1.50E+07 [1]
Water volume (m^3)	6.75E+07 [1]
Sediment active layer depth (m)	1.00E-02 [3]
Concentration of solids in water (mg/L)	4.63E-01 [6]
Concentration of solids in inflow (mg/L)	4.63E-01 [6]
Concentration of solids in aerosol (μg/m^3)	4.00E+01 [6]
Concentration of solids in sediment (m^3/m^3)	9.00E-02 [6]
ϱ Suspended particles (kg/m^3)	1.50E+03 [3]
ϱ Sediments (kg/m^3)	2.40E+03 [3]
ϱ Aerosol (kg/m^3)	2.00E+03 [3]

Table S5. Input parameters for HERMES model

Water surface area (m2)	1.50E+07
Water volume (m3)	6.75E+07
Mean water depth (m)	4.50E+00
Sediment active layer depth (m)	1.00E-02
Precipitation rate (m/yr)	2.50E-01
Settling rate of solids (g/m2 day)	3.04E-01
Resuspension rate of solids (g/m2 day)	2.43E-01
Burial rate of solids (g/m2 day) *	6.08E-02
Total suspended solids in water (mg/L)	4.63E-01
Total suspended solids in inflow water (mg/L)	4.63E-01
Concentration of particles in air (ug/m3)	4.00E+01
Aerosol dry deposition velocity (m/h)	1.08E+01
Volume fraction particles in surface sediment	9.00E-02
Density of water particles (kg/m3)	1.50E+03
Density of sediment particles (kg/m3)	2.40E+03
Density of air particles (kg/m3)	2.00E+03
Fraction OC in water column particles	2.00E-01
Fraction OC in sediment solids (SS)	4.00E-02
Fraction OC in resuspended SS	2.00E-01
Fraction OC in inflow suspended SS	4.00E-02

Figure S1. Atmospheric Hg dispersion over region around Lake Balkyldak during Campaign 2



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