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Persistent organic pollutants (PCBs and OCP) in air and soil from Ulaanbaatar and the Lake Hovsgol region, Mongolia

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Abstract: The investigations of POPs in soil and air in three urban and rural sites of the Mongolia are presented. The POPs distribution in air repeats the POPs distribution in soil on the area investigated. The POPs levels in soil and air are lower than maximum permissible concentrations (MPC) and preliminary permissible concentrations (PPC) of PCBs and OCP accepted in Russia. POPs levels in Mongolian soil obtained in the investigation are comparable with those from background areas of the world. POPs levels in Mongolian air are in the frame of concentrations found in the world. The PCB homological pattern in soil near electric power station in Ulaanbaatar is close to homological pattern in PCB technical mixture (Sovol or Arochlor 1254). The homological patterns in soil from other sites changed due to the redistribution of PCB congeners in the environment. The ratio of DDT and its metabolites indicates fresh entrance of DDT in the environment of Mongolia due to the atmospheric transboundary transport from countries using DDT (China, India) or from local agricultural sources. Hazard indexes in result from human exposure with POPs in soil and air are lower by 2-4 orders than 1 that denotes the possible default of disturbances in target organ and system. CR under the same scenario corresponds to the first diapason that is taken by population as negligible risk, not differ from usual everyday risks. Such risks don't require additional measures for the reducing of risks and their levels are a subject of periodical control. The necessity of additional investigation of POPs distribution and the fate in Mongolian environment is indicated.

Introduction

Persistent organic pollutants (POPs) is the group of dangerous compounds with common properties. They are persistent in environment; bioaccumulative along food chain; toxic for human and other alive organisms (AMAP, 1998; Sunden, 1998). POPs bring remote effects including cancer, disturbance of development, dysfunction of reproductive, endocrine and immune systems and other health problems. POPs are able to long range transport. POPs can be found in remote areas, far from primary sources of formation and application (AMAP, 1998). The measures protecting human health and the environment form negative effects of the compounds are taken on the national, regional and international levels. The measures provide elimination and/or reduce the emission, release of POPs into environment and, if it is necessary, the cessation of production and application. Stockholm Convention on the persistent organic pollutants was adopted on the Conference of the Parties and opened for the signing 22 May 2001. More then 150 countries have signed the convention. The Convention entered into force on 17 May 2004 (http://www.chm.pops.int).

The list of compounds in the Stockholm Convention consists originally of 12 individual substances and group of compounds. The list A (the ban of production and application) of Stockholm Convention in version of 2001 consists of aldrin, chlordane, dieldrin, endrin, heptachlor, hexachlorobenzene (HCB), mirex, toxaphen, polychlorinated biphenyls (PCBs). DDT is included in the list B (the limitation of production and application). Polychlorinated dibenzo-para-dioxins (PCDDs) and dibenzofurances (PCDFs), PCBs and HCB are included in the list C (the reduction and liquidation of emissions in result from unpremeditated production). The possibility and the procedure of addition of new compounds are provided by the Stockholm Convention. Nine additional compounds including α-hexachlorocyclohexane (α-HCH), β-HCH and lindan (γ-HCH) were included in the list of the Stockholm Convention in 2009 by the Conference of the Parties of the Convention. The amendments entered into force on 26 August 2010 (http://www.chm.pops.int).

Mongolia signed the Stockholm Convention during the Conference of Plenipotentiaries in 2002, 17 May. State Great Khural of Mongolia ratified the Stockholm Convention 2004, 30 April (http://www.chm.pops.int). National plan on POPs was heard and approved at the session of Government of Mongolia in 2006, May 3 (National Implementation Plan, 2006).

HCB, PCBs and organochlorine pesticides included in the list of Stockholm convention have never been produced in Mongolia (National implementation plan, 2006). Pesticides were purchased in other countries including former USSR. Aldrin, chlordane, dieldrin, heptachlor, HCB and HCH were used since 1950s to control internal parasite of livestock, treatment of fences and grasslands against grasshoppers. DDT, endrin, mirex and toxaphene have never been used on the territory of Mongolia (National implementation plan, 2006). Ministerial Decree № 75 from 14 May 1997 banned the usage of aldrin, dieldrin, chlordane, DDT, endrin, HCB, heptachlor and toxaphen in Mongolia.

PCBs have been used previously and they being used at present days in industrial transformers. The inventory of 2004-2005 found that there are 4637 transformers of 35 types in Mongolia at that time (National implementation plan, 2006). 96 percents of the transformers were produced in Russia in 1968-1980s. The rest transformers were produced in China (2.1 %), Japan (0.7 %), Germany (0.7 %), Bulgaria, Romania, Korea, Czechoslovakia and Hungary (0.5 %). The investigation of 557 samples of transformer oil indicates that 12.4 percents of the samples don't contents PCBs, 7.7 percents of the samples content PCBs more than 50 ppm. 81.3 percents of transformers are used or kept in the central district of Ulaanbaatar and other large cities. Inadequate conditions of application, storage and utilization of obsolete transformers and pesticides can result in pollution of the environment and increasing of health risk for population due to the exposure with PCBs and OCPs. For example, the Erdenet Mining Corporation used 2 tones of waste of transformer oil on the roads in order to reduce road slippery in winter time. The analyses of soil samples found the PCBs concentrations more than 50 ppm (National implementation plan, 2006). The investigation of POPs in air in the area of settlement of Khankh and sediments and zoolplankton from Lake Hovsgol indicate that the compounds arrive to the region as a result of atmospheric transport (Goreglyad et al., 2007; Mamontova et al., 2009a). However the impact of small local sources was found also (Mamontova et al., 2009a).

The aim of the paper is to present the first data on PCB and organochlorine pesticides levels in air and soil samples from Ulaanbaatar, its residential suburbia Yargayat-bogino and in Tarialan somon Hovsgol aimag.

Experimental

Samples collection

Passive air samplers (PAS) have been obtained by using of polyurethane foam disks (PUF-disks) and used to investigate POPs levels in air. The construction of PAS used in the study was similar to those described in Pozo et al. (2009). The theory of the passive sampling using similar devices was described by Shoeib and Harner (2002). PUF-disks were precleaned by Soxhlet extraction for 10 h using dichloromethane (DCM). PAS were installed in the zone of influence of electric power station III in Ulaanbaatar, in Yargayt-bogino in 18 km to the north-east from Ulaanbaatar and in rural area in the south-west from Lake Hovsgol (Tarialan somon Hovsgol aimag). The position data and time of the installation of PAS are presented in Table 1 and Fig. 1.

In addition soil was sampled in the PAS installation sites in summer of 2009. Soil samples were collected using tube metal sampler with diameter of 5 cm from five sites by the method of envelope. Soil samples were dried under a room temperature up to air-dry condition. The fraction of 2 mm was taken for the analysis.

Soil samples and PUF-disks were stored at –30^0C prior to the analysis in laboratory of the Institute of Geochemistry, Siberian Branch of RAS in Irkutsk, Russia

Table 1. The description of air sampling sites in Mongolia

	Time of air sampling	Longitude	Latitude	Elevation under sea level, m
Ulaanbaatar, electric power station III	24.12.2008 - 15.04.2009	$47^0$53'38"	$106^0$52'28"	1278
Yargayt-bogino	25.12.2008 - 15.04.2009	$48^0$02'12"	$106^0$53'40"	1504
Tarialan somon Hovsgol aimag	06.01.2009 - 11.07.2009	$49^0$25'22"	$101^0$55'20"	1054

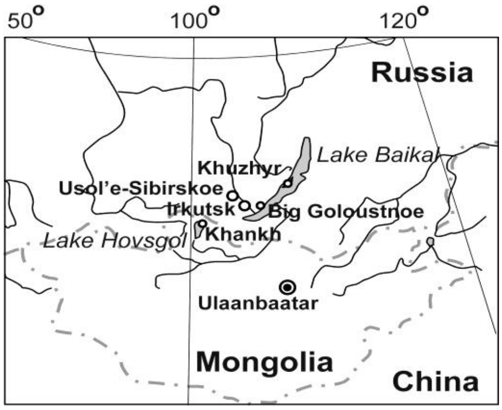

Fig. 1. The scheme of the region investigated.

Sample extraction and analysis

Samples were analyzed for 28 PCBs, including indicator PCBs (28, 52, 101/90, 138, 153, 180), *p,p'*-DDT, *p,p'*-DDD, *p,p'*-DDE, α-HCH, γ-HCH, *trans*-chlordane (TC), *cis*-chlordane (CC), *trans*-nonachlore (TNCL) and *cis*-nonachlore (CNCL) at the laboratories of the Institute of Geochemistry in Irkutsk (Russia).

PUF disks were Soxhlet extracted using DCM. Surrogate standards (PCB 14 and PCB 65) were added in solvent prior to the extraction. The extract was purified on an aluminum oxide / silica gel / silica gel+H_2SO_4 column.

Soil were Soxhlet extracted using acetone/hexane (1:1). Surrogate standards (PCB 14 and PCB 65) were added in solvent prior to the extraction. Large molecules in the extract were separated by gel permiation chromatography (GPC) on Bio-Beads S-X3. This was followed by a column consisting of 3 g Al_2O_3 (activated at the 900^0C), 3 g silica gel (activated at the 450^0C) and 3 g Na_2SO_4. Published analytical methods were used (Mamontov et al., 2004; Mamontova et al., 2009).

The POPs were analyzed using gas chromatography with a ^{63}Ni electron-capture detector (GC-ECD). The Hewlett – Packard 5890 series II GC was equipped with a 0.25 μm $\times$ 60 m DB-5 capillary column (J&W Scientific). The amounts of PCBs and OCPs in air in sampling time were obtained assuming that sampling rates were 3.5 m^3 in a day.

Human health risk assessment

The carcinogenic and noncarcinogenic human health risks were calculated under the "Guidance on risk assessment…" (R 2.3.10.1920-04). The data on the inhalation rate for adult (20 m^3 per a day), the index of accidental ingestion of soil particles by adult human (50 mg/kg per a day), adult body mass (70 kg), the rate of exposure per year (365 days) and the lifelong scenario of exposure (70 years) (R 2.3.10.1920-04) were used for calculations.

Results and Discussion

Results are presented in Tables 2-3 and Figures 2-3.

Soil. The levels of POPs in soil from Mongolia were equal to 3.97 – 13.2 ng 28 PCB congeners/g dry weight including 1.43 – 5.94 ng 6 indicator PCBs/g, 0.06 – 4.28 ng total DDTs/g, 0.02 – 0.81 ng α+γ-HCHs/g, and 0.0045 – 0.03 ng HCB/g. These concentrations are lower than maximum permissible concentrations (MPC) and preliminary permissible concentrations (PPC) of PCBs and OCP accepted in Russia (The control chemical and biological parameters of the environment, 1998 and GN 1.2.1323-03).

PCB levels obtained in Mongolian soil are comparable with levels in soil from background regions of the world (5.41 (0.026 – 96.9) ng/g) (Meijer et al., 2003), rural, background and suburb areas of the Lake Baikal region (0.45 – 32 ng/g) (Mamontov et al., 2004) and lower than those in industrial areas. For example PCB levels in soil from the town of Usol'e-Sibirskoe in the Lake Baikal Region comes to 530 ng/g (Mamontov et al., 2004).

The HCB levels in soil from Mongolia are comparable or lower than in soil from background regions of the world (0.68 (0.01 – 5.21) ng/g) (Meijer et al., 2003) and soil of the Lake Baikal region (0.02 – 72 ng/g) (Mamontov et al., 2004). DDTs and HCH concentrations in soil are comparable or lower than in Lake Baikal Region (Mamontov et al., 2004), in forest soil of background areas of Austria (nd-22 ng/g and 0.6 – 6.6 ng/g, accordingly) (Weiss et al., 2000), in mountain areas of Europe (1.7 – 13 and 0.08 – 0.49 ng/g, accordingly) (Grimalt et al., 2004), and considerably lower than in rural soil from China (Guangzhou) (3.58 – 830 and 0.19 – 42 ng/g, accordingly) (Chen et al., 2005).

The highest accumulation levels (concentrations expressed on the square) of PCB and OCP were found in soil from the zone of the atmospheric influence of electric power station III in Ulaanbaatar (Table 2). In Yargayt-bogino, residential suburbia of Ulaanbaatar, the PCBs, DDTs and HCHs levels are 2-14 times lower and amount to 22, 3.12 and 0.25 $ng/cm^{2,}$ correspondingly. The HCB levels in soil from Yargayt-bogino were insignificantly higher than in Ulaanbaatar. The lowest accumulation levels were found in soil from rural area in the south-west from Lake Hovsgol (Tarialan somon Hovsgol aimag).

The ratio of individual compounds in sum of POPs group is also different. For example the homological composition of PCB in soil sampled nearby the electric power station in Ulaanbaatar is different from the PCB composition in soil from Yargayt-bogino and Tarialan somon Hovsgol aimag (Fig. 2). The homological composition of PCB in soil nearby electric power station is similar to those in soil from Usol'e-Sibirskoe (the Lake Baikal Region, Russia) and in technical mixture of PCB (Sovol or Arochlor 1254) (Fig. 2). Whereas the homological compositions of PCB in the rest Mongolian soil samples changed due to the redistribution of PCB congeners in the environment because of different physical-chemical properties of the individual PCB congeners (Wania, Mackay, 1996).

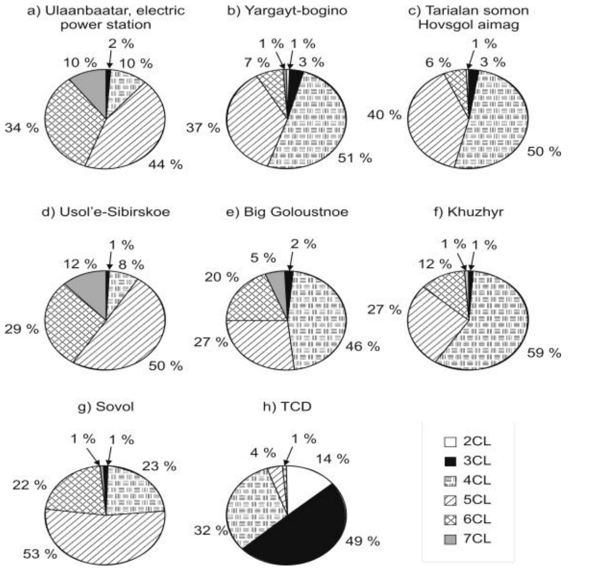

Fig. 2. The PCB homological pattern in soil from Mongolia (Ulaanbaatar (a), Yargayt-bogino (b) and Tarialan somon Hovsgol aimag (c) (present investigation), the Lake Baikal region (the settlements of Usol'e-Sibirskoe (d), Big Goloustnoe (e), Khuzhyr (f)) (Mamontova et al.,2009) and PCB technical mixtures produced in former USSR (Sovol (g) and TCD (h)) (Ivanov, Sendell, 1991) (%).

The cluster analysis of PCB congener patterns in soil samples from Mongolia and some soil samples from the Lake Baikal region allow to distinguish two groups (Fig. 3). The first group consists of soil samples from Yargayt-bogini and Tarialan somon Hovsgol aimag (Mongolia) and settlements Khuzhyr and Big Goloustnoe located on Lake Baikal in the zone of atmospheric impact from industrial centers of the Irkutsk Region. The second group unites the soil samples from the cities of Ulaanbaatar and Usol'e-Sibirskoe and technical mixture of PCB (Sovol or Arochlor 1254). It should be noted that the PCB congener pattern in both groups differ from the PCB congener patterns in TCD (trichlordiphenil) being another technical mixture produced in former USSR (Ivanov, Sendell, 1991).

Table 2. The POPs accumulation levels in soil and concentrations in air from Mongolia

	Ulaanbaatar		Yargayt-bogino		Tarialan somon Hovsgol aimag	
	soil	air	Soil	air	soil	air
	ng/sm^2	pg/m^3	ng/sm^2	pg/m^3	ng/sm^2	pg/m^3
HCB	0.14	20	0.21	20	0.009	7,6
Total HCHs	3.56	78*	0.25	21.5*	0.04	15.5*
Total DDTs	18.8	335	4.12	185	0.11	237
Total PCBs	58	2640	21.6	1650	7.7	1730
Total indica tor PCBs	26	965	8.4	610	2.7	625

* - only α-HCH

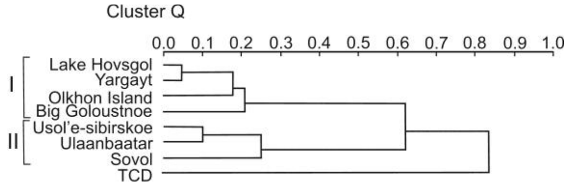

Fig. 3. The grouping of PCB congener patterns in soil from Mongolia and the Lake Baikal region with the cluster method.

Thus, the homological and congener pattern of PCB in soil nearby electric power station assumed the presence of source. It is possible that the transformers containing PCB technical mixture (Sovol or Arochlor 1254) can be the source in this case. These transformers can be functioned or/and stored on the electric power station.

The ratio of α- and γ-isomers of HCH are different in samples investigated. The ratio α/γ-HCHs are equal to 6.7 in Ulaanbaatar, 2.5 in Yargayt-bogino and 1.05 in Tarialan somon Hovsgol aimag. These values of α/γ-HCHs are evidence of the preferred application of technical mixture of HCH in the Ulaanbaatar region and additional using of lindan in the rural areas of Hovsgol aimag. The technical mixture of HCH consists of 53 – 70 percents of α-HCH and 11 – 18 percents of γ-HCH. While γ-HCH is the main component of lindan (90 percents) (Mel'nikov, 1987).

The ratio of DDT/DDE above 1 indicates the recent using of the pesticide. The ratios of DDT and DDE were higher 1 in all soil samples from Mongolia (13 in Ulaanbaatar and 1.36 – 1.85 in Yargayt-bogino and Hovsgol aimag).

Atmospheric air. The data on the levels of POPs in air of Mongolia are presented in Table 2. The distribution of POPs in air repeats the distribution of POPs in soil on the area investigated. Data obtained in Mongolian air were lower than MPC and PPC of PCBs and OCP accepted in Russia (1 μg/m^3 for PCB, 0.001 mg/m^3 for HCH and DDTs and 0.013 mg/m^3 for HCB) (The control chemical and biological parameters of the environment, 1998 and GN 1.2.1323-03).

The values of total PCB in three areas of Mongolia in the period from January-April till July 2009 were higher than the data obtained for the settlement of Khankh located on the northern coast (May-June 2008 (96 pg 28 PCBs/m^3)) (Mamontova et al., 2009a). The phenomena can be result from different season of air sampling. The mean temperature in January in the Lake Hovsgol Region and the city of Ulaanbaatar come to 24 and about 20-22 ^oC below zero, accordingly (absolute minimum are 45.3 and 49 ^oC below zero) (Atlas of Mongolia, 2004). In addition to the Siberian anticyclone places in the region in winter time and can be the reason of difficulties of the emission scattering from industrial and domestic sources in the season. It is possible that long time of air sampling is one more reason of elevated levels obtained. It is probably that the long time of passive air sampling can give rise to nonlinear accumulation of PCBs investigated in PUF-disks and give a mistake in data obtained (Shoeib, Harner, 2002). The additional investigations are needed to specify levels and the distribution of PCBs in atmospheric air in Mongolia. However, PCBs levels obtained in air of Mongolia are in the frame of data found in the result of GAPS study (Global Atmospheric Passive Sampling) (0.1 – 2830 pg 48 PCBs/ m^3) in December 2004 (Pozo et al., 2006), in the settlements of the Irkutsk Region (280 – 2730 pg 28 PCBs/m^3) in June-August 2008 (Mamontova et al., 2009b). But the data obtained for Mongolia are comparable and higher than values found in air of China in September-November 2004 (21 – 336 pg 29 PCBs/ m^3) (Jaward et al., 2005), India in July-September 2006 (120 – 1080 pg 28 PCBs/m^3) (Zhang et al., 2008) and European countries (20 – 1700 pg 29 PCBs/ m^3) (Jaward et al., 2004).

The DDT and its metabolites levels in air of the area investigated are also higher than those found in the settlement of Khankh located on the northern coast of the Lake Hovsgol (18.4 pg/m^3) (Mamontova et al., 2009) and are in the range of values found in the Irkutsk Region (Russia) (65 – 1440 pg/m^3) (Mamontova et al., 2009b). They are comparable and lower than levels in air in India (16 – 2952 pg/m^3), where the pesticide is used at presence (Zhang et al., 2008). The DDE levels in air in Mongolia higher and DDT levels are comparable with the levels in air in European countries (< 0.4 – 25 and 0.6 – 190 pg/m^3) (Jaward et al., 2004).

The ratio of DDT/DDE in both air and soil samples from Mongolia are higher 1 (1.43 in Tarialan somon Hovsgol aimag, 1.15 – in Yargayt-bogino and 1.3 – in the zone of atmospheric influence of electrice power station in Ulaanbaatar) and indicate relatively recent entrance of DDT in the environment. In spite of the default of data of the DDT application in Mongolia, the recent entrance of the pesticide in the Mongolian environment is possible as a result of atmospheric transboundary transport from the territory of China or India where DDT is used at presence (Chen et al., 2005; Zhang et al., 2008). It is possible that the local entrance of DDT can result from an application of housekeeping.

α-HCH levels in air in Tarialan somon Hovsgol aimag and Yargayt-bogino are 2.2. and 1.5 times lower than in air in the settlement of Khankh in the northern coast of Lake Hovsgol (34 pg/m^3) (Mamontova et al., 2009a). α-HCH level in air in Ulaanbaatar is higher than one in Khankh. They are comparable or lower than levels obtained in GAPS-study (0.1 – 1700 pg/m^3) (Pozo et al., 2009), European countries (< 14 – 100 pg/m^3) (Jaward et al., 2004), the Irkutsk Region, Russia (19 – 365 pg/m^3) (Mamontova et al., 2009b), India (12 – 1691 pg/m^3) (Zhang et al., 2008).

HCB levels in air in Tarialan somon Hovsgol aimag is lower in 1.9 times, and those in Ulaanbaatar and its suburb are slightly higher than HCB air levels in the settlement of Khankh (14 pg/m^3) (Mamontova et al., 2009a). HCB air contents in Mongolia are comparable or lower than HCB levels in European countries (11 – 50 pg/m^3) (Jaward et al., 2004), China (10.4 – 461 pg/m^3), Singapore (9.5 – 24.5 pg/m^3), Japan (14 – 95 pg/m^3), South Korea (26 – 136 pg/m^3) (Jaward et al., 2005).

Human health risk assessment. Data obtained were used for the carcinogenic and no carcinogenic human risk assessment as a result of the exposure with complex of POPs investigated (PCBs, DDT and its metabolites, α- and γ-isomers HCH and HCB) under inhalation of atmospheric air and accidental ingestion of soil particles. The concentrations found in air and soil in the three site of sampling are typical of the areas investigated (the central areas of Hovsgol aimag, Ulaanbaatar and its suburb was assumed.

Hazard Indexes (HI), the indexes of noncarcinogenic risk, were calculated for the following target organs and systems: liver, the central nervous system, immune and endocrine systems, development. HI was 2 – 4 orders lower than 1 in all cases (Table 3). The exceeding of 1 indicates the possibility of an appearance of disturbances in target organs and systems (R 2.3.10.1920-04). Carcinogenic risk (CR) under human exposure with POPs from soil changes from $5.78*10^{-9}$ in rural area in the Hovsgol aimag to $2.32*10^{-8}$ in Ulaanbaatar. The CR values correspond to the first diapason that is taken by the population as negligible risk, not different from usual everyday risks. Such risks don't require additional measures for reducing risks and their levels are subject of periodical control (R 2.3.10.1920-04). PCBs bring the main contribution in total CR (89 – 95 percents). The PCB contribution was followed by the contribution of HCH (2.7 – 8.3 percents), DDT and its metabolites (1.8 – 2.2 percents) and HCB (0.33 – 0.92 percents). The mean CR value under POPs inhalation exposure comes to $1.25*10^{-6}$. The value corresponds to the second diapason – the maximum permissible risk or upper level of permissible risk (R 2.3.10.1920-04). The values can result from the overestimation of air levels (discussed above) and require an additional and seasonal investigation. The constant control is needed if the values will be confirmed.

Table 3. The hazard indexes (HI), carcinogenic risk (CR) and the contribution of individual groups of POPs in total CR under human exposure as a result of the inhalation of atmospheric air and accidental ingestion of soil by adult.

	HI	CR	the contribution in total CR, %			
			total PCBs	total DDTs	total HCHs	HCB
air, mean	0.029	$1.25*10^{-6}$	92.5	1.97	4.9	0.60
soil, mean	0.0003	$1.46*10^{-8}$	91.2	2.58	5.9	0.32

Conclusions

1. The concentrations of persistent organic pollutants in soil from Mongolia come to 3.97 – 13.2 ng 28 PCBs/g dry weight, including 1.43 – 5.94 ng indicator PCBs/g, 0.06 – 4.28 ng total DDT/g, 0.02 – 0.81 ng α+γ-HCHs/g, and 0.0045 – 0.031 ng HCB/g. The POPs distribution in the atmospheric air repeats the POPs distribution in soil on the area investigated.
2. The POPs levels in soil and air are lower than maximum permissible concentrations (MPC) and preliminary permissible concentrations (PPC) of PCBs and OCP accepted in Russia. POPs levels in Mongolian soil obtained in the investigation are comparable with those from background areas of the world. POPs levels in Mongolian air are in the frame of concentrations found in the world.
3. The PCB homological pattern in soil near electric power station in Ulaanbaatar corresponds to the homological pattern in technical mixture of PCB (Sovol or Arochlor 1254). The homological patterns in soil from other sites changed due to the redistribution of PCB congeners in the environment.
4. The ratio of α- and γ-isomers of HCH in soil from Mongolia indicate to predominant application of the technical mixture of HCH in the area of Ulaanbaatar and lindan in the Hivsgol aimag
5. The ratio DDT/DDE is higher in both soil and air from Mongolia that indicates fresh entrance of DDT in the environment of Mongolia due to the atmospheric transboundary transport from countries using DDT (China, India) or from local agricultural sources.
6. Hazard indexes as a result of the human exposure with POPs in soil and air are 2-4 orders lower than 1 that denotes the lack of disturbances in target organ and system. CR

under the exposure with POPs from soil particles correspond to the first diapason that is taken by the population as negligible risk, not different from usual everyday risks. Such risks don't require additional measures for reducing risks and their levels are a subject of periodical control. The CR values under inhalation exposure with POPs correspond to the second diapason – the maximum permissible risk or upper level of permissible risk (R 2.3.10.1920-04). The values can result from the overestimation of air levels (discussed above) and require additional and seasonal investigations. The constant control is needed if the values will be confirmed.The investigation was supported by RFFI № 10-05-93173-Mong_a and 10-05-00663-a

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