МОНИТОРИНГ ПРИРОДНЫХ ПРОЦЕССОВ

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Effects of Self-protection of Natural Ecosystems on the Example of Artificial-Natural Geochemical Barriers in Landfills of North-Western Russia

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Abstract. For Nature any landfill is foreign and aggressive object, it impairs and pollutes atmosphere, soils, grounds, surface waters and groundwater. Usually these environments are taken as bearing invariable natural characteristics and so under artificial impacts they need in artificial protection. The most expansive efforts in this usually are targeted to groundwater, mainly it is construction of waterproof screens around waste masses. But our numerous data from landfills in the North-Western Russia allow to conclude that the pollution of groundwater when compare to surface waters is always insignificant and does not depend nor on a size of the landfill, nor on a time of its operation, and even nor on ground conditions, because of the widespread natural development of geochemical waterproof barriers in grounds under the influence of leachate. This is the prominent example of the self-protection of the environment against an aggressive object according to Le Chatelier principle and ideas of biogeochemical energy of Vernadsky.

***Keywords**: geochemical barrier, gleying, groundwater, landfill, leachate, pollution, waste, surface water*

Эффекты самозащиты природных экосистем на примере искусственно-природных геохимических барьеров на полигонах ТБО Северо-Запада России

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Аннотация. Для природы любой полигон отходов является чужеродным и агрессивным объектом, она нарушает и загрязняет атмосферу, почвы, грунты, поверхностные и подземные воды. Обычно эти среды воспринимаются как обладающие неизменными природными характеристиками, поэтому при искусственном воздействии они нуждаются в искусственной защите. В этой защите от свалок наиболее масштабные усилия обычно направлены на грунтовые воды, в основном это строительство водонепроницаемых экранов вокруг массивов отходов. Но наши

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многочисленные данные по полигонам Северо-Запада России позволяют заключить, что загрязнение подземных вод по сравнению с поверхностными всегда незначительно и не зависит ни от размеров полигона, ни от времени его эксплуатации, ни даже от грунтовых условий, вследствие повсеместного естественного развития геохимических водонепроницаемых барьеров в грунтах под воздействием фильтрата. Это яркий пример самозащиты окружающей среды от агрессивного объекта в соответствии с принципом Ле Шателье и идеями биогеохимической энергии В.И. Вернадского.

Ключевые слова: *геохимический барьер, глейзинг, подземные воды, полигон ТБО, фильтрат, загрязнение, отходы, поверхностные воды*

Introduction. Setting and state of the problem

A waste from production and domestic activities is an undesirable but inevitable product of human civilization. By their physical state a waste may be or solid, or liquid, or gaseous, or wave one. It is clear that it is the solid phase of waste is most persistent in environment and it will produce most durable contaminations. But the vast majority of solid waste (average of 98.5 % by mass) is waste of low classes of hazard (IV-V classes by Russian scale).

By their sources solid waste may be divided to production waste (mostly industrial, ISW) and domestic (municipal, MSW) waste (Grinin, Novikov, 2002). Although ISW accounts about 90–98 % of total volume of waste in different countries, their composition vary, depending on respective industries, but rather predictable, while MSW tends to be geographically more constant in volume and composition, but more difficult than ISW in dealing with them.

In Russia the present calculation standard only for household solid waste in different regions is from 0,2 to 0.5 t/person·year (Denafas e.a., 2014). The actuality of the problem for Russia is confirmed in 2014 by the adoption of new essential (110 pages!) amendments to the “Federal Law on waste of production and consumption” (1998), which came into force from the beginning of 2016 (On amendments…, 2014).

There are variable methods of waste utilization, as compression in a sealed container for the burial, or gasification, or combustion, including combustion in boiling layer, or pyrolysis, or sorting and further processing to secondary raw materials, etc. — a total of at least 20 technologies (Flawed…, 2007), in the best variants reducing to the secondary usual of waste. All of them are still limited in Russia, because, typically, of their expensiveness. Really the main way for waste utilization is and will be their burial to landfills.

As practice shows, the most vulnerable environment from landfills is hydrosphere. In particular, 75 % from available 55.000 landfills in USA are contaminants of groundwater (Jones, Lee, 1981). In Russia, 12 % of groundwater pollution sites are associated with wastewater and solid waste. The largest number of sites of total (from any contributors) contamination of groundwater in Russia is located in the Volga (32 %), Siberian (24 %); Southern (15 %) and Central (13 %) federal districts (Galitskaya, 2005) (Fig. 1).

Fig. 1. Federal districts of Russia and location of North-Western Federal District (NWFD).

Рис. 1. Федеральные округа России и расположение Северо-Западного федерального округа (СЗФО).

In a humid climate, which is typical for the most part of Russia, the adverse impact to the environment from MSW increases by strongly contaminated *"leachate*" (Canziani, 1994). It is a dark murky liquid, enriched with pollutants arising under the passage of precipitation through the waste mass and under related microbiological processes (Noble, 1991). Getting into the groundwater, the leachate determines the spacing of pollutants. In different countries, from Italy to New Zealand, the numerous complicated theoretical chemical models are built for the processes of formation of leachate (Grungaletti e. a., 2016). They may be based on the equations of fluid motion in porous media, on a damped flow of contaminants, on some equilibrium reactions, on biogeochemical effects in soils, on kinetics of reactions, until the application of the principles of Ben Robinson and Gibbs and so on (Islam e.a., 2001; Noble, Arnold, 1991; Qiu, Sun, 2011; Shuzong e.a., 2011, etc.). The key model parameters are waste moisture and the rate of compression. However, the yield and the composition of leachate is subject to seasonal and weather changes (as rain and air humidity), they complicate numerical solutions and forecasting in the models (Cossu e.a., 1988; Sivakumar e.a., 2009).

The waterproofing from the leachate is accepted to be necessary. In accordance with the requirements of normative documents of Russia (Federal Law…, 1998; Construction Rules…, 2001) the focus and accordingly the main expenditure fall on protecting of the groundwater. The most expensive requirement (40…70 % of total cost of landfill) is the construction of a waterproof screen at its base to protect the ground against a leachate. There is the typical complex of works during the construction and operation of the landfill recommended in the normative documents (Goldberg, 1995; Manual…, 1996), but in practice this sequence of works is almost never enforced totally.

Two main kinds of waterproof screens are: 1) natural (usually clay) screen or 2) artificial (usually plastic) screens. The advantage of plastic screens is their almost complete preventing of ingress of leachate into the grounds. However, recent studies show that all the landfills allocate the leachate. Even the plastic screens with a thickness of 2.5...10 mm used in the United States, permit the leakage of liquids and gases through the pores and through mechanical damages. The older landfills, the cracks in screens wider. So, in 2000, according to the LLSI

Company, 82 % of landfills had holes in the screens (Comments…, 2006).

But the main deficiency of artificial screens is their high cost. So, in Russia the average cost of construction of 1 sq. m. of polymer screens is about $7, while the construction of a clay screen is about $0.9 (Belyi, 2003). That is, ceteris paribus, the polymer screen leads to the increase of investment in 142.4 % when compare to using of clay screen (Raznoschik, Abramov, 1983). Clay screens are cheap, so used widely in many countries and in Russia too (Weisman, Korotaev, 2001; Tagilov, 2002), but are not possible anywhere. They are not absolutely impenetrable, especially in arid climate.

For example, in conditions of humid climate of Southeast China the highly soluble and thus the most rapid ions (chloride, sodium, etc.) served as markers of the contamination. Maximum penetration reported for chloride (3 m in the case of simple diffusion) (Zhan e.a., 2014). For all interactions it revealed that the effectiveness of a clay screen grows mostly only to a thickness of 1 m (Cuevas e.a., 2012).

The properties for clay screens in Russia are standardized by (Manual…, 1996); according to which the clay-bonded layer in basis of the trench should have a filtration coefficient no more than 0.0086 m/day (about 10^{-9} m/s) and a thickness no less than 0.5 m.

The considerable recent attention has been focused recently for detailed studies on the work of clay screens. So, in Madrid region the state of 15 large landfills was investigated, and the largest landfill of Madrid which had been covered with a clay screen 20 years ago was taken as indicative (Regadio e.a., 2012). The monitoring of overlying soils and of grounds of the discharge zone of the landfill indicates that contamination worsens with age. However, despite details of the chemistry, all the authors accept "*the mechanical inpermeability to be the main desired property for clay screens"* — the screen should not be broken mechanically. At the same time the direct experiments in Spain (the model was created for a 20-year landfill – see: Cuevas, 2012) show that in due course the clay screen of 0.5 m thickness in deep undisturbed layer decreases many times its exchange capacity and permeability coefficient, many times from original 10^{-7} m/day, due to the reduction in mechanical diffusion.

In addition to the natural and newly mechanically generated (natural-anthropogenic) clay screens, the active research is being conducted, particularly in China (Zhou e.a., 2014 etc.) to invent as artificial ways for disposal of groundwater contaminated by landfill leachate, and appropriate long-term barriers. As a variant there were proposed filter columns for that, with various-grained passive iron, activated carbon and zeolite, whose efficiency has been proved for the removal of organic impurities (particularly aromatic hydrocarbons). The cheapness of these substances allows to make a continuous filter reliable at least for 10 years. The other options in this direction are sand-bentonite mixture with natural clay or mixture of bitumens, comparable by it's efficiency with plastic screens (Maslov, 1999; Bezzar, 2010).

Thus, the modern study of water pollution from landfills moves in two main directions. On the one hand, the methods for the study of contamination processes are evolved including different models of leachate evolution, which, however, rarely provide reliable predictions. On the other hand, a variety of technologies to shield artificially and to eliminate contaminants are provided.

As to existing landfills, major American experts on solid waste Lee and Jones-Lee (Jones-Lee, Lee, 2000, 2007) — subdivide all landfills depending on the selected leachate control strategy into two types: 1) dynamically stable ("controlled bioreactor") and 2) buried ("dry", formerly known as "dry grave"). Also it is proposed to compare well-designed and managed landfills of both types (referring to the USA landfills) and landfills in "developing" (referring to the underdeveloped) countries, later conventionally classified to the third type.

The first type ("bioreactor") is focused to achieve a stabilization within 30...50 years and is applicable to a low-hazard biodegradable waste. The decomposition is accelerated by recirculation of water and leachate through the landfill mass. Together with a number of benefits (as ability for control and planning, constant volumes of liquid and biogas, etc.), numerous requirements can be placed in such landfills on suitable composition, moisture and amounts of waste and have a number of issues - in particular, the leakage of the filtrate. Buried (dry) landfills, a concept which is known in Russia as "dry grave" are focused to maintain the waste in a relatively dry condition above a groundwater level for a long time with the prevention of biodegradation and formation of leachate and biogas as long as possible. This type of landfills was designed for hazard and low-hazard waste and is the most widespread type of landfill in the United States and France (National…, 2000). However, currently the criticism of the "dry grave" concept increases (Comments…, 2006; Flawed…, 2007). In essence, the "dry grave" is a temporary care which can only to delay the pollution of groundwater by leachate. The estimated life time of these "graves" is hundreds of years for so long that the noticeable volume of leachate and the risk of proliferation would arise, including because of probable damage of the screen. In addition, there are problems of uncertainty of the evolution of the landfill and of control for that for such a long time.

Landfills in “developing countries”, differentiated by Lee (Jones-Lee, Lee, 2007) in the third type, are characterized by a predominance of food and vegetable waste and by small amounts of toxic materials. In addition, requirements for screens there can be weaker there than in industrial countries, and really the requirements are very different in every country. The rigidity of the legislation also is typically lower than in Europe or in United States. Most of landfills of the third type can be attributed to the type of "open dump", elaborated designs with screens and barriers are rare, and in general the waste management in general is not a priority.

With regard to the above, Russia occupies an intermediate position in relation to the development of the waste management system. On the one hand, there are numerous and voluminous normative documents regulating the methods of disposal, including requirements for landfills. On the other hand, in practice, due to financial and organizational obstacles, these requirements are carried out rarely. The landfills of "dry grave" type practically not occur in Russia due to the predominance of shallow groundwater level. The existing landfills can be attributed rarely and conditionally to the category of "controlled bioreactors" (rather uncontrolled). The largest number of landfills, in spite of new strong legislation (On amendments, 2014) up to day are illegal dumps and primitive landfills.

Materials and Data

Features of natural conditions of the North-Western Federal district

The North-Western Federal District (NWFD) is one of industrially developed regions in Russia, so one of most stressful situations associated with solid waste is observed here. However, as we have seen above, the North-West is not among the regions with the highest groundwater contamination. We can assume this is due to the natural features of the district.

NWFD is a plain region, located mostly in a zone of taiga, with boreal, transitional to marine, climate, characterized by significant precipitation under low evaporation, and so with excessive moistening (Fig. 2). The annual rainfall varies between 500...700 mm, while evaporation from water surfaces and areas occupied by forest is about 350...550 mm, from grassland – 300...500 mm, from marshes – 300...400 mm. Stable snow period in different areas ranges from 140 to 200 days (the Nature…, 1983; the Nature…, 2007).

Fig. 2. The orography and main cities of NWFD. Lowlands (greenish shades) dominate on the territory. Deep-blue points are investigated sites.

Рис. 2. Орография и основные города СЗФО. На территории преобладают низменности (зеленоватые оттенки). Темно-синие точки — исследуемые участки.

As a result of excessive moistening, it is the North-West that has **the highest in Russia degree of waterlogged soils and accordingly the high groundwater level**, which is held to be a very negative factor for landfills.In general two features of natural conditions of the Northwest are important for displacement of landfills: 1) the water regime, which forms in conditions of slow outflow on the background of positive water balance; that contributes to the intensification of water migration of pollutants; 2) the development of morains and heavy structureless soils derived from moraines with low filtration properties and significant adsorption capacity; that can be a natural geochemical barrier to contaminants. If the first factor is purely negative, the latter is considered to be favourable (De Vrier, Bakker, 1998).

Grounds of the region are diverse, but the vast majority of them are associated with activity of young (upper Pleistocene) Quaternary glaciations. Their essential properties for landfills properties are presented in table 1.

The main geochemical feature of the landscapes of the North-West is low background quantities of most microelements (Makarov, 1969, Trufanov, 2000). In the humus horizons of the soils, the concentration of all heavy metals except cadmium sometimes does not reach clark amounts. So the low background of trace elements in combination with a humid climate and with the prevalence of leaching regime in soils forms the conditions reducing the technogenic loads on the landscape (Fortesque, 1985; Perelman, Kasimov, 1999).

Table 1

Some parameters of typical grounds of NWFD

Таблица 1

Некоторые параметры типичных грунтов NWFD

Genetic type of deposits	Dominated grounds	Dominated clay minerals	Dominated absorbed complex	Mean absorption capacity mg-Eq	Order of filtration coefficients m/day
Glacial	loams, rarely	illite	Ca, Mg	5...12	0.5 in sandy

(moraines)	clays and sandy loams			(rarely up to 20...25 in loams	loams to 0.001 in heavy loams
Lacustrine-glacial	essentially clays	illite with an admixture of montmorillonite and kaolinite	Ca, Mg	10...15	pronounced stratification from 0.02...0.2 to 0.001...0.01
Fluvio-glacial	sands, loams	Quarts, carbonates	Ca	1…8	1...10 in sandy loam about 0.5–1.5
Alluvial	sands sandy loams	Quarts	Ca	1…5	About 1 m/day even in loams
Cover loams	Silt	illite (60 % or more) with numerous labile components, kaolinite, carbonates	Ca, Fe	14...28	0.03...0.15

In the soil cover of the North-West the acidic podzolic forest soils are dominated. Among non zonal types the hydromorphic soils of boggy areas, often gley ones (see “Discussion”), may be listed. We emphasize that gleying generally is an effective process for many subtypes of soils of the region (Glazovskaya, 1997).

Objects and methodology

Obviously for the Saint-Petersburg agglomeration new solutions to the problems of MSW are most relevant for the Saint-Petersburg agglomeration (Bashkin, Gregor, 1999; Common…, 2000). But as a parallel to St.-Petersburg the situation with landfills for some another large cities, more typical for NWFD can be considered. For example we took Vologda and Cherepovets. These two cities are similar by population and climate, but Cherepovets differs by development of a heavy industry and so by large volumes of specific ISW respectively.

The environmental monitoring and the engineering-geological and engineering-ecological prospecting at numerous sites in regions of NWFD hve been spent for 30 years. To summarize the data of surveys and monitoring of numerous firms, mainly limited liability companies (LLC) “Lentisiz”, “Vologdatisiz”, "Lengidroproekt", "EPIR", Russian Geo-Ecological Centre (RGEC) "Urangeo", "Progress", "Energogazizyskaniya" closed joint stock company (CJSC) "Test Center VNIIG" etc. are assigned. The data of 380 sets of chemical analyses of the atmosphere air, waters and soils have been used, from 6 to 24 components in each test. These analyses in different years, at different sites were performed on different equipment, by different techniques and with different sets of the analyzed pollutants. Because that for comparability of those numerous prospecting and monitoring data the results of water investigations have been interpreted in "EPIR" LLC using so-called generalized index of hazard marked as ***“R”***:

$$R = \frac{1}{\Sigma MPC_m} \sum_{i=1}^{m} \frac{a_i}{\xi_i} \quad (1)$$

where ***m*** is the number of considered substances in one limiting value of the index of injuriousness, MPC is Maximal normative (or permeable) Pollution Concentration.

The values under the sign of the sum represent the relative concentrations of the *i* substance and its MPC, i.e.:

$$\left\{ \begin{array}{l} a_i = S_i / S \\ \zeta_i = MPC_i / \Sigma MPC_i \end{array} \right\} \quad (2)$$

where *S* is the total concentration in water of all substances considered by one value of “***R”***; ***Σ MPC*** is the total value of these substances.

The values of the index in almost undiluted leachate can reach 25 and rarely higher. Its background values in natural waters of the rivers of the Vologda region are 0.15...0.85.

As can be seen from (2), the feature of “***R***” is that it depends only on the chemical composition and does not depends on a degree of dilution. So “***R***” could been used for the generalization and correlation between objects. In addition, such Russian standard indexes, as Complex Index of Water Pollution (CIWP), Conventional Complex Index of Water Pollution (CCIWP) were used for the correlation.

Data from some landfills

Our detailed data are presented in (Belyi, Shmakin, 2017), here we offer only some examples from Vologda region and vicinities of St.-Petersburg.

Vologda region is the middle region by many indexes for all Russia, and therefore it often serves as an indicator or base for socio-economic researches. Many natural features are close to the average Russian values too (Nature…, 2007). The number of landfills meeting requirements is the quite typical for Russia (Belyi, 2003; Construction rules, 2001). According to results of the inventory, there was 399 landfills on the territory of the region, only 36 of which had been included in the regional inventory of waste, which is the state register of waste disposal facilities. As an objective necessity, 82 authorized unequipped and 281 unauthorized dumps "serve" too (Report…, 2021).

The center of the region is ***Vologda*** city, and it can be taken as the first example. Old Vologda landfill (now destroyed) was located on the lacustrine terrace. First water confining bed (3…4 m) is a grey glacial-lacustrine clay with a permeability coefficient about $9{,}3\cdot10^{-8}$m/s, that meets the regulatory requirements for clay screens. At 1993 the landfill was equipped by four observation wells – in the center, up- and downstream of underground flow. The monitoring observations of groundwater of the first aquifer (1.8-2.5 m) were conducted for 30 years and for 20 contaminants as: nitrogen group, heavy metals, biological and chemical oxygen demand (BOD, COD). The results have been converted to "***R***", some of them are presented in Table 2.

Table 2

Index of hazard (R) according to the monitoring of groundwater in wells of Vologda landfill

Таблица 2

Индекс опасности (R) по данным мониторинга подземных вод в скважинах Вологодского полигона ТКО

Well No.	Date of sampling						
	22.07.99	14.10.99	14.06.00	02.11.00	14.06.01	31.10.01	10.12.07
282 (center)	8.754	3.341	18.988	3.085	4.206	3.362	13.3
279 (above)	5.541	3.342	16.1	3.272	3.401	3.481	–
283 (above)	3.699	3.345	10.071	2.325	3.495	3.366	–
281a (below)	3.972	3.341	9.723	1.962	3.457	3.393	–

– no data

As can be seen from the table, the major variations of "***R***" are associated with the seasonal factors. The pollution of groundwater for summer time, without influence of flooding, is high in the centre and decreases to the periphery. But **any significant difference between wells "below" and "above" is not observed**. The same results were obtained on the ***Cherepovets*** landfill, although it is located on the lacustrine-alluvial terrace covered by **sands**, with filtration coefficients up·to $2\cdot10^{-5}$m/s (!) — unlike Vologda, a natural clay screen under the waste body is missing here. But average for 6 years values of "***R***", average for 6 years, above and below the landfill differ not significant: 1.72 and 1.91 respectively (Belyi, Shmakin, 2023). So, in spite of permeable grounds the contamination is insignificant even when compare with Vologda, any significant increase in contamination due to penetration of leachate into ground water at the site of the waste body is not visible. As at the Vologda landfill, obvious is the synchronicity of fluctuations in the values of "***R***" on both wells is obvious, that caused by fluctuations in rainfall and snowmelt runoff. In addition to cities, to evaluate the environmental impact of rural damps we undertook the same studies in 4 rural mainly unequipped dumps of the Vologda district: "***Maysky", "Kurkino", "Fetinino", "Nadeevo"***. These dumps are exploited for

enough long time of 20 years or more. All of them are located in similar socio-economical (near large villages with a population about a thousand) and climatic conditions, but differ by their ground conditions.

The underground waters on all 4 objects were tested at three points on every site: 1) directly under the body of the waste mass, 2) at the border and 3) at 25…40 m, from the depths of 0.2…2.0 m, depending on hydro-geological conditions. The results evaluated to the hazard index ***"R"*** are presented on the chart (Fig. 3).

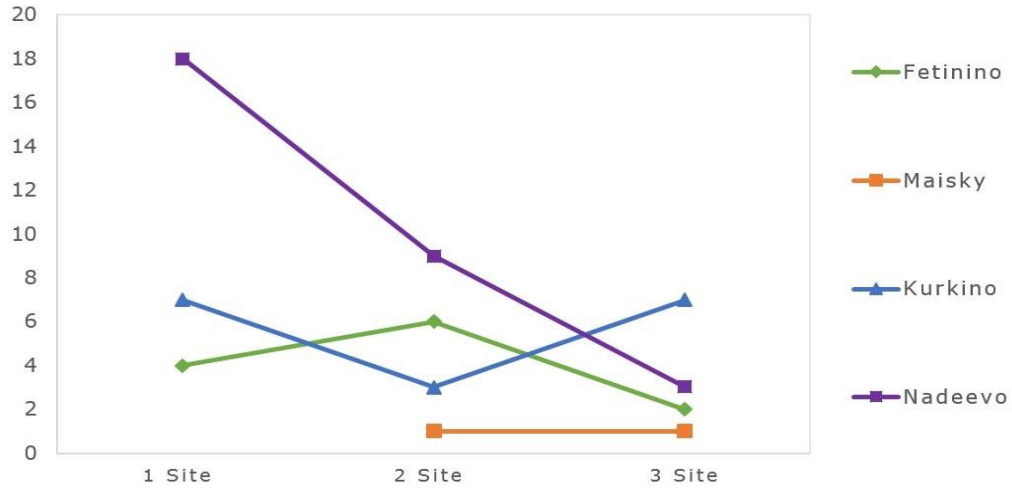

Fig. 3. The change in the index of hazard ***R*** at the sites of water sampling on rural dumps.

Рис. 3. Изменение индекса опасности R в местах отбора проб воды на сельских свалках.

It is clear from Fig. 1 that in most cases, contamination of groundwater from the center to the periphery decreases. But even throw 25…40 m (sites #3) the contamination is about background one. It is interesting that we can see the maximum levels of contamination as in the center, as on the bounder of the dump, in well-protected grounds of Nadeevo.

On 4 unauthorised and unequipped dumps inside Vologda city, and on 4 country dumps of Vologda district, and on 5 large fully equipped landfills in vicinities of Saint-Petersburg etc. the similar **low levels of pollution in groundwater** as below and above waste bodies were revealed. It doesn't depend nor on soil and ground conditions, not on screens and an arrangement.

In many cases at the base of the waste bodies **a bluish-grey colmated layer soaked by the leachate** of 0.2…0.8 m thickness was revealed, with the filtration coefficient on the order $1 \cdot 10^{-8} \ldots 1 \cdot 10^{-9}$ m/s; that is much less than normative one.The maximum value of the "***R***" index (186, that is much above than even in the leachate) is dedicated to the samples of bottom silt from so-named "Dead lake", formed by leachate, near ***Kungolovo*** landfill (40 km to the S from St.-Petersburg). We can assume that this is due to increased adsorption processes here and, ultimately, due to formation of a barrier, suppressing the farther emission of pollutants (Goldberg, 1995; Methodological…, 1987) – see below.

In the center of ***Kirovsk*** landfill the stratified sampling revealed that the contamination to be confined by the bluish-grey layer (outlined above) at the bottom of waste masses and is distributed no deeper the first 1m beneath, regardless of ground composition. Deeper, the contamination does not exceed the allowed level, as shown on Fig. 4.

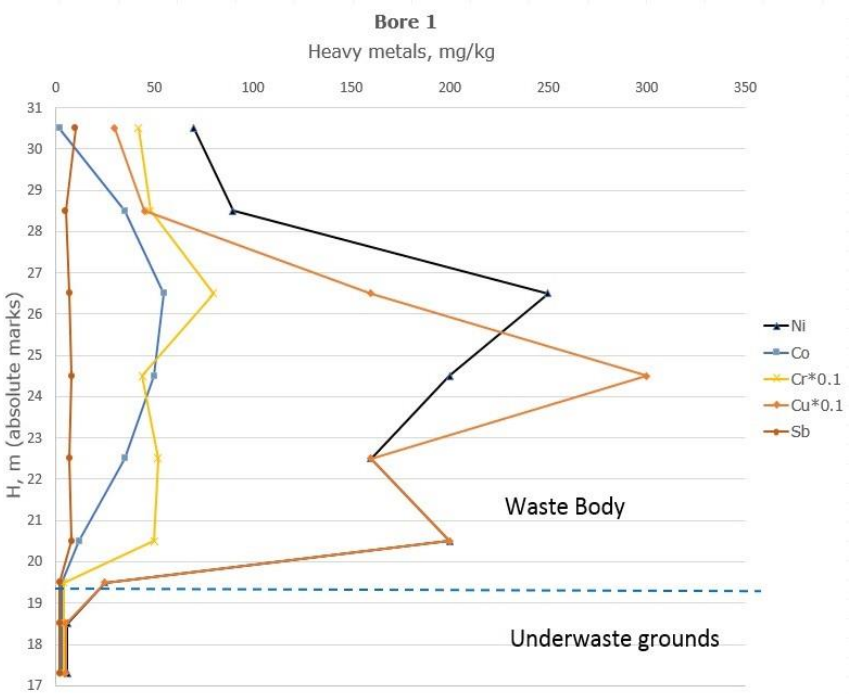

Fig. 4. An example of the distribution of heavy metals in soils in the central well of the Kirovsk dump (data of "Urangeo", plasma chromatography).

Рис. 4. Пример распределения тяжелых металлов в почвах центрального колодца Кировской свалки (данные "Урангео", плазменная хроматография).

In wells under the waste body mass it was discovered the **bluish-grey gleyed layer** with a thickness of 0.2–0.6 m.

For profiles of the wells downstream of groundwater flow (from North to South), a growth of groundwater contamination is not observed in the area close to the actual dump. The most contaminated water having CCIWP up to 350(!) was detected in the well # 55 at 300 m to the North of the dump, i.e. upstream of groundwater flow. This hurricane values of CCIWP outside the influence of the dump is mainly due to high content of iron and manganese from former peat quarries: 58 mg/l, which is practically ore concentration (Beus, 1972; De Vries e.a., 1998; Lietuvninkas, 2005) and due to huge COD, obviously at the expense of organic matter (up to 2200 mg O_2/dm^3).

In ***Saint-Petersburg*** city and suburbs the largest landfill is "Novosyolki" one, it have been closed at 2020. This large landfill inside the metropolis remained for decades as an object of detailed monitoring of all natural environments. According to these studies, the pollution of soil and air environments can't be traced by no one indicator more than 0.5 km.

The main unique natural feature of the landscape of this area, which was noticed in the very beginning of the landfill design, is the presence of a good natural waterproof horizon only at 2…4 m below the surface; it is layered clay packet (Riphean – Quaternary). The drilling throw waste masses has discovered **the colmated waterproof layer of up to 1 m by thickness** gained universally currency. So around and beneath of the landfill the groundwater (not counting perched groundwater on the same level with the body of the landfill) are well shielded and, as in other investigated landfills, remains virtually clean up to date.

In contrary, the surface waters flowing from the landfill are polluted very intensively. Formally they are "extremely dangerous" (CCIWP = 89(!) even 2 km downstream – table 3).

Table 3

Hazard Index (*R*) and Contamination index (CCIWP) for surface waters monitoring at the landfill "Novosyolki" before its close

Таблица 3

Индекс опасности (R) и индекс загрязнения (CCIWP) для мониторинга поверхностных вод на полигоне "Новосёлки" до его закрытия

indexes	point 1 (bypass ditch in the centre)	point 2 (a bypass ditch below)	point 3 (the "main ditch")	point 4 Chornaya riv. 2 km below
R	22.8	9.26	74.3	30.9
CCIWP	572.4	532.4	591.2	88.7

Thus, the most contaminated environment in the largest of St.-Petersburg municipal solid waste landfill is, once again, not groundwater, but surface waters, its contamination is higher by 2-3 orders of magnitude than the contamination levels in all other studied sites in the region. Note that extremely dirty river Chornaya ("Black" in Russian) in fact derives from the landfill, and not only flows completely through the territory of the largest metropolis of Russia, but also falls into the large lake of Lakhtinskiy Razliv on the shores of which the refuge territory is located (Yuntolovo reserve of ornithological directionality) and a new housing district is on construction.

Results

As can be seen from the foregoing, in all landfills of NWFD, different in size and natural (in particular geological) conditions, such elements of the environment as soil, groundwater and air are practically not contaminated already at the distances of few dozen meters from landfills and dumps. but the most vulnerable environment is surface water.

The paradox is that groundwater is significantly contaminated only at the best insulated Vologda landfill, but for a shallow depth and in small extent. The volume of the landfill, the presence or absence of the waterproof screen in the base of the waste body almost does not matter. So, even at landfills in Kirovsk and Kungolovo, placed on sandy soils, a substantial increase groundwater contamination have not been identified.

In general, our results can indicate the presence of **natural phenomena, suppressing the migration** of contaminants in groundwater. The only reason for that may be the formation of the **artificial-natural geochemical barrier** (Perelman, 1999). preventing further pollution in the underlying grounds under influence of leachate, even in sands. The time for formation of this barrier in a matter is not more than of 10 years.

Really such barrier as bluish colmated gleyed layer is visible in the bottom of every landfill. Its filtration coefficient ($\approx 10^{-4}$ m/day or $\approx 10^{-9}$ m/s) is far below the normative values for artificial screens. The bottom sediments, as shown in Kungolovo' "Dead lake", obviously are of a considerable buffer capacity, so one can concede an adsorption barrier to contaminants too.

Discussion. The role of geochemical barriers in natural protection of groundwater

As it is shown before (see "Introduction"), the natural properties of the environment around landfills usually **perceived as a given, or constant**, that can be changed only artificially. The natural-artificial processes of change in grounds and waters around landfills, in particular the processes in geochemical barriers, the mudding and especially gleying, attract the attention of researchers less likely. Also, in assessing of possible impact on the environment, which is performed at the design stage, the influence of complex of factors, which we call the natural shielding of areas, is practically ignored. The basis for determination of this shielding is the concept of the "*assimilative or environmental capacity"* (Bashkin, 2002). This concept is closely related to the primary function of landscapes stationed under aggressive impact as of artificial-natural systems and is determined largely by the presence of geochemical barriers, locking or suppressing the emission of pollutants into the environment. Essentially the complex natural geochemical barriers at the base of the landfill waste mass, which are dramatically alien and aggressive object for the existed ecosystem, are a

form of the **self-defense of dynamic natural ecosystems**.

The concept of dynamic interrelated systems with flowing equilibriums in upper geospheres generally has been elaborated already in the beginning of XXth century, especially in works of Russian academician V.I. Vernadsky and of his school of thought concerning the nature of the biogeochemical processes (Vernadsky, 1926, 1987). Essentially this concept bases on the principle of Le Chatelier-Braun, established primary by prof. H.L. Le Chatelier in 1884 for simple gas thermodynamic systems. In turn this principle was an advancement of ideas of J. H. van't Hoff, H.F.E. Lenz, J.W. Gibbs, K.F. Braun and other investigators in a field of thermodynamics by the end of XIXth century. Le Chatelier's principle or "*the principle of displaced equilibrium*" postulates that an external influence, disturbing the system from a thermodynamic equilibrium, causes some processes in the system tending to weak effects of such influence. In modern systemology it have been shown that Le Chatelier's principle is an example of wide equilibrium seeking principle, which implies that *"As systems are moved away from equilibrium, they will utilize all available avenues to counter the applied gradients..."* (fon Förster, 1960, p. 31).

In the case discussed here, the really new is the simultaneous use of only a few principles demonstrates such a high explanatory power in this context. It appears that even a very condensed version of the simple principles derived from ecosystem theory can simplify apparent problems to a degree where subtle speculations are no longer needed (Kay, 2000). Being applied to ecosystem theory (Műller, 2022) they are helpful in understanding of practical problems.

Remember that Le Chatelier was one of Vernadsky's main mentors, interlocutors and colleagues in Paris during 1889-1890. Later Vernadsky wrote: "*He was one of most remarkable person I met through my life. From him I had learned first time about works of American theorist and mathematician Gibbs… which became known in Europe some years later*" (Vernadsky, 1926, p. 86). The main idea of Vernadsky was that: "…the living matter… is geological force (biogeochemical energy) of outstanding value" (Vernadsky, 1987, p.43), "…geological Earth's envelope – the biosphere – is one … having a certain structure… named as an organization…" (ibid., p. 45). V.I. Vernadsky recognized 7 kinds of the matter in the biosphere. Soils, crust of weathering and so on belong in his concept belong to "biostagnant matter", created by organisms and non-organic processes simultaneously. "*These biostagnant organized masses are complex dynamically equilibrium ones, in which the geochemical energy of the living matter sharply manifests itself as biogeochemical energy*"(ibid., p. 52).

"*The living matter represents the most powerful geological force, increasing in the course of time. It… is sharply isolated… in the form of billions organisms… They presents autarkical centers for energetic and physico-chemical processes and are continuous connected with the environment by biogenic migration of atoms*…" (ibid., p. 119).

Later academician A.E. Fersman, as main follower of V.I. Vernadsky, has developed the concerning of biogeochemistry. Describing among other geochemical systems "*the system of living matter*", distinguished from different silicate's and other geochemical system, Fersman pointed that oxidation in this system usually initiates the high valences of cations, enhanced polarization and the occurrence of light and mobile anions. But "*the reducing medium, produced mainly by biogenic processes, otherwise tends to low valences, to lesser solubility and migrations of chemical complexes*." (Fersman, 1958, p. 221). Such is indeed the case around landfills.

This line of inquiry had been prolonged in works of A.I. Perelman, who was in its turn one of Fersman's follower. Perelman had developed Fersman's concept of geochemical systems, and had formulated the conception of "***geochemical barriers***". They are zones of Earth' crust characterized by accumulation of certain chemical elements and by weakening of migration flows. As to landfills the most important are sorption, acidic and reducing (gley) barriers, formed by soils as self-maintaining systems having certain assimilation capacity (Perelman, 1999; Alekseenko, 2003; Bashkin, 1999, 2002).

Some researches of landfills in this direction were conducted recently, but with no mention of the concept of "geochemical barrier". So,

Spanish researchers (Regadio et al., 2012) had revealed the absorbing and insulating layers. They proved that rains and smectite-illite-carbonate clays act as a "*reactor*" that reduce pollution from leachate.

In another article (Regadio et al., 2015) these authors examine more specifically the long-term effectiveness of natural barrier, developing on 12-year landfill, where the conductivity and content of soluble organic carbon, chloride, ammonium, sodium, used as markers for measurements of leachate pollution, have been measured in the groundwater. In the underlying soils the mineralogy, surface properties and cation exchange capacity have been studied; the associations with local geological conditions were identified too. Although the leachate was in contact with clays for a long time, the fronts of changes and electrical conductivity have been decreased in the clay layer at a distance of 0.2...1.5 m. Under such composition of a clay screen (at least 45 % illite-smectite component in layered silicates), it is considered to be sufficient to limit the pollution by leachate pursuant to the European legislation (Regadio et al., 2015).

Chinese investigators described the relevant differences between the composition of clays at 3 landfills, for which the index of "*Capacity reduction of liquid diffusion"* was derived (Shuzhong et al., 2011). The latter is the function of the degree of reduction parameters per 1 m of screen thickness, of age and area of the landfill, and of the amount and composition of waste. Obviously this term is very closed to the same of Bashkin's "*assimilation capacity*" (Bashkin, 2002).

The colmation (mudding) and gleyzation (gleying) deserves a special attention. The gleyzation have been studied mainly by Russian scientists and it was precisely on the North-West of Russia, where this phenomenon is most widely developed. The terms "*gley*" and "*gleeobrazovaniye*" (gleyzation) were introduced into the terminology of soil science by G.N. Vysotsky (1905) to denote a "*more or less dense rock, gray with a greenish tinge, formed in conditions of prolonged waterlogging*" (Cit.: Zaidelman, 1998). Vysotsky primary pointed to the biochemical nature of gleying, and established the role of transforming of iron from ferric oxide to ferrous oxide under the shortage of oxygen with the participation of anaerobic microfauna. In foreign (non-Russian) science exact terms "gleyzation" or "gleying" are rare; in soil science the process closest to it usually named "*redoximorphic*" (Fortesque, 1985; Visvanadham, 2007), but it is more wide term.

According to the generalization of the works devoted to the soil formation process in conditions of excessive moisture, prof. F.R. Zaidelman (1998) underlined that the gleyzation is one of the most global processes of soil formation. "*Three simple factors as waterlogging, as the presence of organic substances capable for fermentation, and the presence of heterotrophic microflora on acidic, leached and neutral grounds, free from sulfates, are the necessary and sufficient conditions for the emergence of the process of gleyzation*" (Zaidelman, 1998, p. 262). The ubiquity of these three major factors in base grounds of all investigated MSW landfills in conditions of excessive moisture testifies that the development of gley process is very likely there.

First, the storage of solid waste generates the steady positive landforms consisting of a fairly loose mass of waste quickly saturated by water precipitation, which is determined by the regime of excessive moistening.

Secondly, there are an anaerobic conditions at the base of the landfill in connection with high concentration of dissolved organics in leachate; BOD and COD exceed the MPC by several orders of magnitude (Grugnaletti et al., 2016).

Thirdly, a significant proportion of municipal solid waste is organic matter, primarily of biogenic origin, i.e. food waste, paper, wood to name but a few, rich in most diverse, especially in anaerobic microflora.

Thus, we see there all available three factors according to criteria of F.R. Zaidelman for the gleyzation. **It is gleying after mudding creates powerful artificially-natural geochemical barriers** at the base of all the studied dumps and landfills, regardless of the quality of soils and of quantity of waste.

A characteristic feature of gleyzation is the reduction of the oxide compounds of metals and unbalanced removal of iron (Alekseenko, 2003; Bashkin, 2002). Thus there is a number of crucial transformations of the mineral mass of the soil, which have a decisive influence on the ecological properties and processes subsequently

(Zaidelman, 1998). Under gleyzation the destruction of primary and secondary minerals is fobvious, the elements with variable valence (Fe, Mn, S, N) are subjects for significant changes. This process may be of enzymatic nature and occurs under the influence of metabolic products of anaerobic bacteria (N_2, H_2S, light organic acids, etc.).

It is important that the relative enrichment in silica and depletion in iron and to some extent in aluminum occurs when gleying, but manganese and sulfur reduce with the formation of movable compounds. In particular this is seen in the zone of influence of the landfill of Kirovsk.

The gleyzation is observed in the profile of the underlying grounds **at all the studied landfills**, regardless of the granulometric composition. Easily identifiable symptom of gleyzation is specific gleyed colouration from whitish-gray or gray-bluish in sandy loams, to gray-glaucous, glaucous-blue or greenish in loamy and clay soils, and to deep blue one in sands. The specific colour of these horizons is due to the loss of oxide pellicles of iron by primary and secondary minerals, which cover their own color, and appearance of new minerals under gleying. Under development of the gley process in the bottom of the landfill the permeability of grounds reduces. The increase of sorption capacity of soils under gleying is equally important.

As it was shown by B.V. Trushin in his thesis (1994), the intensity of barrier mechanism increases in sequence: Rock – Soil – Bottom silt. The levels of pollution accumulation in these environments increase linearly in time. The gained empirical dependences can be used for quantitative prediction. It is the time factor plays a special role. "*One of the most important characteristics determining the level of environmental pollution, is the lifetime of the landfill, and it gives you the ability to allocate the main stages of the impact of landfills on the environment. The landfill will have the greatest impact after 3-4 years of operation and in the first 15-20 years after closure.*" (Trushin, 1994, p. 20).

As it is shown by our research, the geochemical barriers mainly form for some first years (not more than 8) after the start of the landfill (the example of Kungolovo). During the initial phase, although the barriers are not fully formed, the leachate volume is small yet and the rate of pollution is small also. The uppermost estimate of the time of formation of the barrier is the actual data of Kungolovo landfill, where from 1984 to 2002 in sandy grounds already quite mature and reliable gley barrier have been formed (see "Data").

Among the studied objects the geochemical barriers are particularly well manifested at the city landfill of Vologda, Kirovsk and Kungolovo. It is reliably established here that the dispersion area of polluting substances in groundwater, which indicator is the concentration of heavy metals, is limited by the contours of the site.

The most powerful example of the barrier is bottom silt of the "Dead Lake" in Kungolovo, which absorbs huge amount of pollutants.

Conclusions

The results allow to suggest the importance of the assimilative capacity of the grounds under and near landfills. The effect of colmation and gleying processes proven under landfills in different ground conditions is equivalent to the artificial succeeding of the normative filtration coefficient for landfills.

The use of such natural processes, including reducing the toxicity of the leachate, as biodegradation, and chemical processes of decomposition and precipitation, etc. is of considerable importance (Cuevas e.a., 2012) with minimal risk and of high reliability. Also it is shown that many regulatory standards for old landfills are clearly excessive (Qiu, Sun, 2011).

During as researchs and making for a formation of artificial barriers the attention should be paid to forms of existing of chemical elements in the migration flow in waters, to their relative quantity and to characteristics of the migration environment, as they determine the process of deposition of elements (or their compounds) in different geochemical barriers.

For practical needs the use of artificial-natural geochemical barriers allows:

1) to achieve the desired rate of filtration coefficient for the underlying grounds within storage site of the landfill, reducing the cost of construction of the protective screen;

2) to expand the spatial capabilities for the choice of sites for the landfill.

The effect of colmation and gleying processes repeatedly proven under landfills in many different ground conditions leads to the decrease in conductivity and to the increase of sorption, which is equivalent to artificial achievement of the normative values of the filtration coefficient for the base of the landfill. **Under certain conditions it is possible totally to opt out the construction of waterproof screen.** This is important because a simple analysis of several dozen projects for the construction of landfills to the extent of even one region, shows that it is billions rubles of estimated cost. The neglect of the actual assimilative capacity of natural ecosystems leads to unjustified overestimate of costs for works of construction and installation.

From our studies it is clear, at least for waterlogged conditions, that the leading propagation way of the contaminants around landfills is not underground one, what today is the aim of normative documents and practice of engineering surveys. So in regulatory framework for the design and operation of landfills we can and we have to move the focus from soil and groundwater protection to surface waters. Under detailed investigations it is possible to work up the territorial building codes for the construction of landfills, close to local specific natural and socioeconomic conditions. As surface waters are extraordinary dirty, it should be targeted as by design work, and by engineering solutions for manual of landfills, the attention in normative documents must be directed to surface way of water pollution. But it is much easier for study and it is able to counteract its pollution effects.

Along with the undoubted economic benefit, the using of the properties of geochemical barriers broadens the choice of sites for landfills. The designer can be not attached to islets of waterproof beds, as was done by the design of "Novosyolki" landfill. This is especially true for large and largest cities with their sharp shortage of land resources.

At existing landfills it is necessary to carry out a radical reconstruction of drainage networks and everywhere, especially in excessively humid areas, to introduce a system of closed water (leachate) turnover, approaching the American concept of "bioreactor" (Jones, Lee, 1981).

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