https://doi.org/10.26565/2410-7360-2023-58-26
UDC 504:628.357.034.2-043.2](477.74-25)(045)

Received 3 November 2022
Accepted 15 March 2023

Biological wastewater treatment plants as sources of environmental pollution by persistent organic pollutants (on the example of Odesa industrial-and-urban agglomeration)

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ABSTRACT

Formulation of the problem. Effective mechanism of wastewater treatment (WT) are important components of reducing anthropogenic load on the environment. One of the most widespread mechanisms of urban wastewater treatment is the biological treatment on a Biological Wastewater Treatment Plant (BWTP). However, increasing the nomenclature of pollutants concentrated in urban wastewater seriously affects the effectiveness of WT on BWTPs, which are not intended for such a wide spectrum of specific pollutants such as, in particular, Persistent Organic Pollutants (POPs). The control of their intake into the environment must be regulated according to the Stockholm convention. The goal of the research is the evaluation of the intake of polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) into the environment from the wastewater of Odesa Industrial-and-Urban Agglomeration (IUA) and determining the volumes of their accumulation in the sea environment.

Purpose. The assessment of the amount of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/F) entering the environment with the wastewater from Odesa IUA and determining the amount of their accumulation in the sea environment.

Methods. In our research, all available official methods of calculation of the unintentional formation of POPs were reviewed, as a result – the main and most complete methods were selected: «UNEP (2013). Toolkit for Identification and Quantification of Releases of Dioxins, Furans and Other Unintentional POPs» and «EMEP/EEA air pollutant emission inventory guidebook. Technical guidance to prepare national emission inventories, 2019». For evaluation of PCDD/F accumulation in the sea environment an improved by authors methodology for calculation of PCDD/F accumulation with their cumulative effect and half life taken into account was used.

Results. The work provides justification for the necessity of use of calculation methodologies for determing the intake of PCDD/F to the sea environment adjacent to Odesa IUA; annual massed of PCDD/F entering the sea environment as part of treated, insufficiently treated and untreated wastewater are calculated; massed of PCDD/F that accumulate in waste activated sludge (WAS) on BWTP are calculated; volumes and specifics of PCDD/F accumulation in the sea basin, where the wastewater of Odesa IUA is being discharged to are determined using the improved methodology that enables taking into account the cumulative effect and half life period of these substances; mass and concentration of PCDD/F, immobilized in WAS, are determined, the excess level of concentration of PCDD/F in WAS compared to the maximum permissible concentration is determined. The total accumulation of PCDD/F in the sea environment over 2007-2017 period is determined.

Conclusions. It was established that the use of calculation methodologies for evaluating PCDD/F volumes in the water environment is the only and necessary condition for satisfying the requirements of the Stockholm convention due to the impossibility of performing a regular instrumental monitoring of PCDD/F intake into the water environment. The use of suggested by us improved methodology for calculation of PCDD/F accumulation with their cumulative effect and half-life period taken into account allows for calculation of PCDD/F masses that were formed throughout the year under consideration, taking into account the PCDD/F masses that were formed during previous years as well.

Keywords*: Persistent Organic Pollutants, polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans (PCDD/F), biological wastewater treatment, wastewater, waste activated sludge.*

In cites: Mykhailenko Vladyslav, Safranov Tamerlan, Adobovska Mariia (2023). Biological wastewater treatment plants as sources of environmental pollution by persistent organic pollutants (on the example of Odesa industrial-and-urban agglomeration). Visnyk of V. N. Karazin Kharkiv National University, series "Geology. Geography. Ecology", (58), 350-359. https://doi.org/10.26565/2410-7360-2023-58-26

Introduction. Wastewater discharge is an important component of antrophogenic impact on the environment of Odesa region of the North-Western part of Black Sea (NWBS), i.e. the water area from Small Adzhalyk Estuary on the North and up to Sukhyi Estuary on the South. This problem is particularly relevant for industrial-and-urban agglomerations in South-Western coastal waters of Black Sea due to large water consumption on a given multifunctional territory. One of the examples of such

impact is wastewater discharge by urban wastewater treatment stations of Odesa IUA to the adjacent waters of North Western Black Sea.

The evaluation of anthropogenic load on the Odesa region of North Western Black Sea is one of the key components of ecological safety enforcement in Odesa IUA. According to the requirements of the Stockholm convention [1], a separate vector of research on the condition of the sea environment must be the control of POPs content/concentration, which poses a set of properties, in particular immutability for decomposition, ability for bioaccumulation and toxicity in any concentration. In the work [2], we discovered the specifics of North Western Black Sea pollution by certain POPs based on expedite data from Ukrainian Scientific Center of Water Ecology. Though, the research data addressed a certain nomenclature of isomers of polychlorinated biphenyls, as well as hexachlorobenzene, which were accumulating in the sea environment throughout their continuous use in the former Soviet Union as part of POP-containing pesticides [3] and as a result of transformer and condenser liquids being released into the environment [4]. Although, in our other work [5] it was discovered that such substances as polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofuran (PCDD/F) enter the water environment as parts of the wastewater being discharged there, the instrumental opportunities of monitoring, however, are not allowing to receive information regarding the contents of these toxic compounds. Moreover, PCDD/F enter the sea environment along with the wastewater as a result of constant operation of the city sewage system and insufficiently effective technologies on BWTP. That's why using calculation methodologies is an important aspect of receiving this data. Since Odesa IUA plays a big role in pollution of the sea environment, and one of the key sources of POPs emission into the environment is the discharge of wastewater, we have considered the intake of these substances into the sea environment as a result of wastewater discharge only from Odesa IUA.

Literature review. The length of sewage network of Odesa IUA makes up to approx. 690 km. Sewage outlets of the buildings lead to pressureless collectors which are usually under the roadway of the streets. Down them the stocks proceed to Sewage Pump Stations (SPS). More than one hundred of pumps located on 26 SPS pump up to 465,000 м³ per day of wastewater to the operational BWTPs.

The treatment of wastewater on BWTP is being performed according to the non-traditional schemes (Fig.1).

Wastewater is supplied to the reception camera

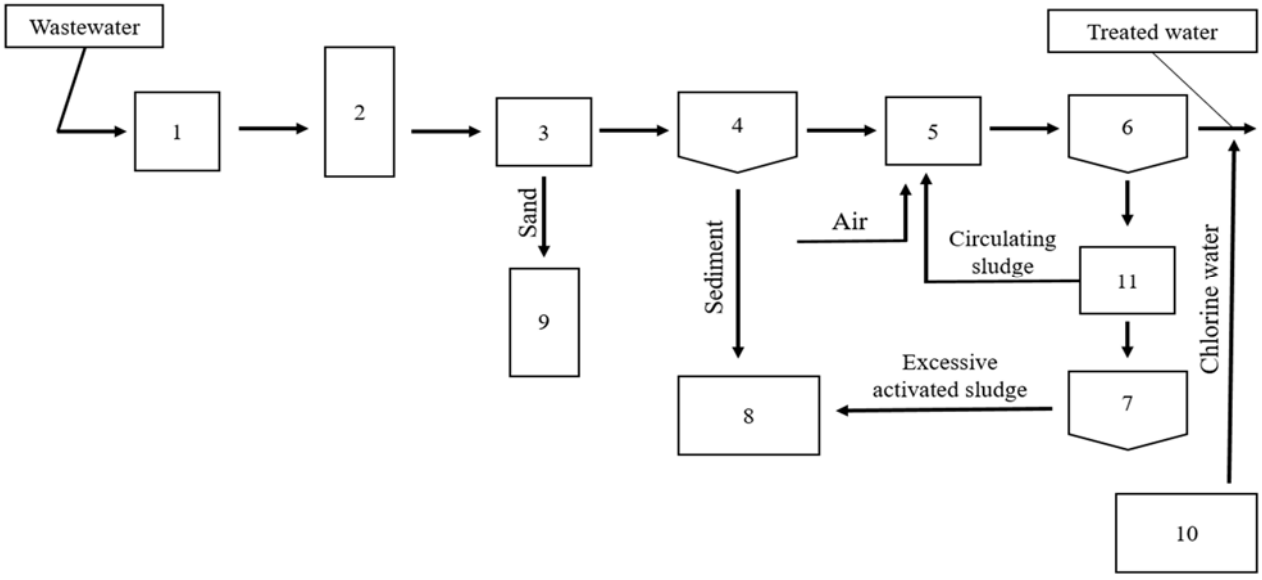

Fig.1. Principal technological scheme of wastewater treatment on urban wastewater treatment plants (Odesa) [6]

(1) where the high-pressure flow is being depressurized and then freely flows to the mechanical treatment facilities under gravity down special lots. Mechanical treatment facilities include cells (2), three horizontal sand traps (3) and six primary radial settling tanks (4). The sand captured in sand traps is deleted on sand platforms (9). The raw sediment separated in the primary settling tanks is pumped to the sludge platforms (8) by pumps located in the raw sediment pumping station. Full biological treatment of wastewater occurs in aerotanks by means of activated sludge microorganisms. Air supply occurs through tubular aerators from foamed polystyrene. After aerotanks the mix of wastewater proceeds to secondary radial settling tanks (6) where under gravity the separation of activated sludge from treated waste liquid occurs. The sediment of activated sludge from secondary settling tanks is collected and then

freely flows under gravity to the airlift camera (11) from where it gets back to regenerators of aerotanks. Excessive activated sludge is returned to sludge thickeners (7). Supernatant liquor from sludge thickeners flow under gravity to the receiving reservoir of the municipal and household wastewater pump station from where it goes to the receiving camera by means of water pumps (1) [6].

In general, wastewater doesn't have a high concentration of PCDD/F, however during the process of chlorination on BWTP (10) the concentrations of these substances drastically increase, in some cases in 50 times. As noted by L.A. Fedorov (1993), disinfection of drinking water with molecular chlorine or sodium chloride electrolysis products leads to the generation of environmentally dangerous concentrations of PCDD/F. They enter the sewer from the water supply network. Sources of PCDD/F may be organochlorine pesticides, phenols, chlorophenols and other chemical compounds in industrial effluents. Other factors can also cause these substances to enter the sewage system, such as washing clothes and textiles that have been treated with paints and biocides that contain PCDD/F. In addition, PCDD/F can become a part of wastewater as a result of intake of effluents containing PCDD/F from combustion sources or as part of untreated industrial wastewater.

Formed in the Odesa IUA wastewater can be discharged in both treated and untreated condition. However, both treated and untreated wastewater may be a source of constant unintentionally formed POPs emission into the environment. It is also worth mentioning that in the process of wastewater treatment at the BWTP, new POPs are not formed, but the existing POPs are redistributed between the wastewater itself that is being treated and waste activated sludge. Therefore, an important source of PCDD/F emission into the environment is also waste activated sludge, which does not enter the sea environment directly, but concentrate on sludge sites and accumulate significant concentrations of POPs, which is clearly demonstrated in the work [5].

However, waste activated sludge is often considered a high-quality fertilizer for use on agricultural land, which corresponds to the principles of circular economy [8, 9] and sustainable development [10]. Although a study of waste activated sludge use in various European countries showed that, despite the high concentration of biogenic elements in it, waste activated sludge can become a source of a wide range of pollutants entering the environment, in particular heavy metals, polychlorinated biphenyls and PCDD/F [11]. After being introduced into the soil, such toxic substances will be bioavailable for cultivated plants, provoking negative effects (for example, phytotoxicity) and being transferred along terrestrial food chains, becoming dangerous for people and the environment [12, 13]. This is why the proper WAS handling is important, since improper handling of them can lead to the uncontrolled spread of PCDD/F into the environment. On wastewater treatment stations of Odesa agglomeration WAS is placed on sludge platforms (see Fig.1), but in the case of their placement on irrigation fields, they can lead to an increase in the concentration of PCDD/F in the soil and plant layers with subsequent movement along the trophic chain. As for WAS, which was buried in burial grounds, it can contribute to the transition of PCDD/F into the leachate and further entering the groundwater. It should be noted that PCDD/F can also be formed during the thermal drying of excess activated sludge and sediments of wastewater.

Discharge of wastewater into open water bodies is a common practice of discharging insufficiently treated sewage directly into surface water bodies. The sources of PCDD/F in this case are the same as in sewage effluents. It should be noted that due to the lack of proper efficiency of sewage treatment, PCDD/F enter the given water body as part of the wastewater directly in full. The general scheme of biological wastewater treatment and, as a result, the entry of PCDD/F into the sea environment during the discharge of wastewater by BWTP of the Odesa IUA is presented in Figure 2.

The purpose of the article is the assessment of the amount of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/F) entering the environment with the wastewater from Odesa IUA and determining the amount of their accumulation in the sea environment.

Research methodology. In our research, all available official methods of calculation of the unintentional formation of POPs were reviewed, as a result - the main and most complete methods were selected: [7] and [14].

All official calculation methods are based on the concept of emission coefficient (factor), which is an experimentally determined numerical characteristic which is common for a technological source of unintentional POPs production. For the majority of the main technological processes, which are accompanied by the formation of POPs, listed in Annex C of the Stockholm Convention, the corresponding emission factors are established, which are given in the methodologies [14] and [7].

Unintentional formation of POPs by fuel combustion is calculated using these coefficients according to the formula:

$$E_{pollutant} = V_{WW} \cdot EF_{pollutant}, \text{ or } E_{pollutant} = M_{WAS} \cdot EF_{pollutant} \quad (1)$$

$E_{pollutant}$ – annual formation of pollutant;
$EF_{pollutant}$ – emission factor of pollutant
V_{WW}– volume of wastewater
M_{WAS} – mass of waste activated sludge

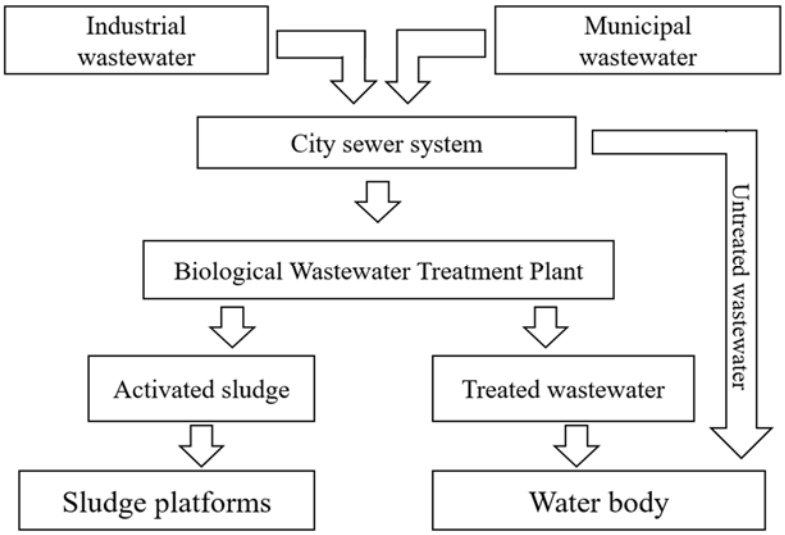

Fig. 2. Principal scheme of PCDD/F entry into marine environment during wastewater discharge by treatment plants of Odesa IUA

In our work [15] it is clearly demonstrated that the separate use of these methods does not make it possible to fully assess the unintentional formation of POPs. It is also worth mentioning that the data of the annual inventory will show a result that will reflect only the formation of these substances for the current year, but not the actual presence of POPs in the environment for the year under consideration. Therefore, we improved the existing methods taking into account the cumulative effect of POPs. It is a common knowledge that the half-life of any pollutant occurs according to an exponential law, taking into account its half-life. Based on this, the equation of POPs' half life will have the following form:

$$A_t = A \cdot e^{-t}_{\tau}, \quad (2)$$

A_t – the mass of POPs after half-life over a period of time t;

A – the initial mass of the POPs;

t – the time period under consideration;

τ – the time period during which the concentration will decrease by e times.

An important component of this formula is the value of the half-life of PCDD/F. Numerous toxic-kinetic studies show that, depending on the specifics of the natural environment, the half-life of 2,3,7,8-TCDD can range from 1 to 50 years [16-18]. In particular, under constant intense exposure to direct sunlight, the half-life can be 1-3 years, and in the deep layers of the soil cover, the half-life can reach 50 years [18]. Therefore, it is considered that on average the half-life of PCDD/F in terms of TE of TCDD (toxicological equivalent, which expresses the cumulative toxicity of complex mixtures of PCDD/F due to the toxicity of 2,3,7,8-TCDD) in the environment is 10 years. Therefore, in formula 2, τ (PCDD/F) is equal to 14.5 years.

Thus, suggested by us improved approach to the calculation of unintentional POPs formation taking into account the cumulative effect has the main advantages, namely: allows for taking into account all of the main sources of unintentional formation of POPs based on the generalized data of the main official methods, which are the basis for reporting on the implementation of the Stockholm Convention; takes into account the specifics of these substances and allows taking into account the effect of their accumulation, hence the obtained results reflect the real picture of presence of accumulated POPs in the environment; determining the accumulated masses of POPs will allow to create a high-quality monitoring system of POPs in Ukraine, because the discovered values show not only the annual masses of POPs, but their actual value for the year under consideration [19].

Presentation of the main research material. In order to assess the release of POPs into the environment during the discharge of the wastewater by the treatment facilities of Odesa IUA, it is necessary to take into account the specifics and volumes of treatment of all types of wastewater. According to [20], part of the wastewater in Odesa IUA undergoes the procedure of treatment by activated sludge, and part of the wastewater is discharged into the sea basin without treatment. Thus, when the wastewater is treated with activated sludge, part of the POPs is redistributed between the actual wastewater and the activated sludge itself, while when discharging untreated and insufficiently treated wastewater, all POPs enter the sea environment directly. As a result of the wastewater treatment by activated sludge, a significant amount of these pollutants is localized in the activated sludge itself and is concentrated on the sludge sites after its removal. That is why it is crucial to consider the release of POPs from given category

of sources separately for water and soil environments.

Using statistical data [20-27], we determined the release of POPs to the environment along with wastewater (both treated and untreated), taking into account their cumulative effect over 2007-2017. Obtained results are presented in the form of Figures 3 and 4.

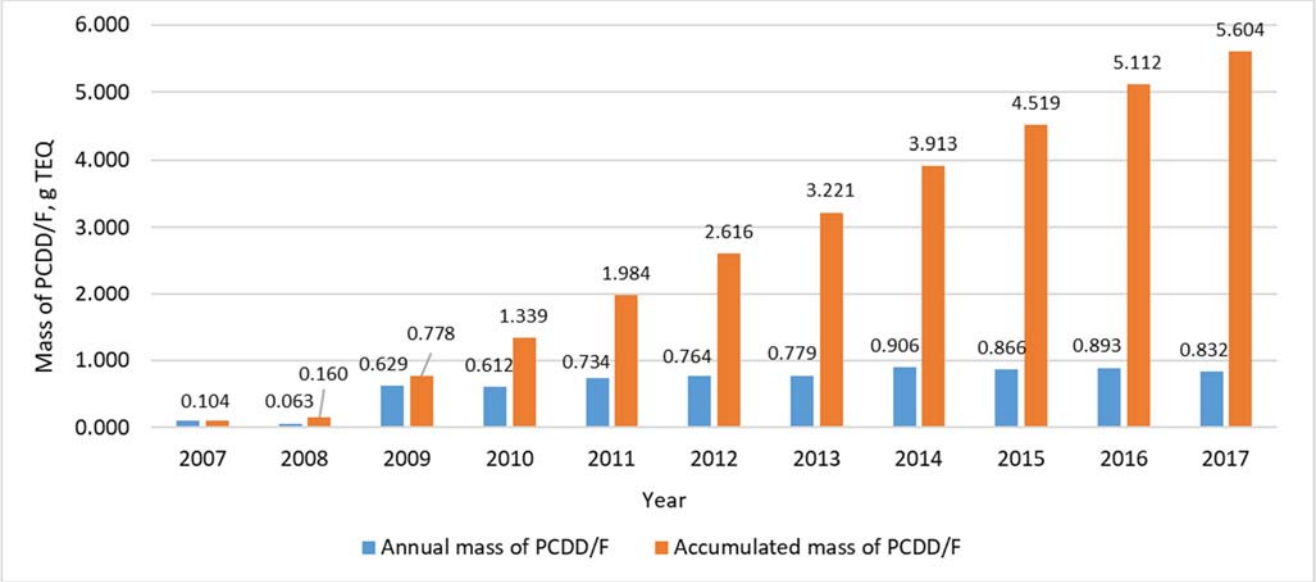

Fig. 3. Annual and accumulated masses of PCDD/F entering marine environment during treated wastewater discharge by treatment plants of the Odesa IUA

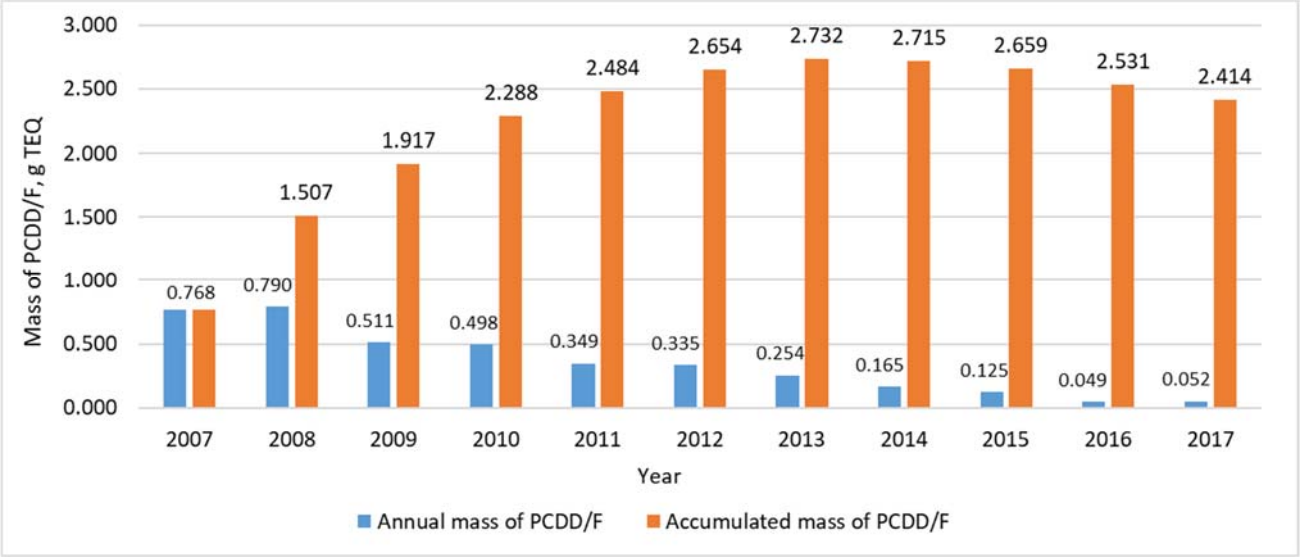

Fig.4. Annual and accumulated masses of PCDD/F entering marine environment during untreated wastewater discharge by treatment plants of Odesa IUA

Annual masses of PCDD/F, which enter the marine environment as part of untreated wastewater have a clear downward trend during the years 2008-2016, which is associated with a significant decrease in discharge volumes of the untreated wastewater. In turn, the amount of accumulated masses of PCDD/F increases until 2013, and from 2014 to 2017 a downward trend of POPs accumulation is observed. However, during the discharge of treated wastewater, there is an upward trend in the accumulation of PCDD/F masses in the marine environment, which is associated with a simultaneous sharp increase in the annual masses of PCDD/F until 2014 and approximately the same annual masses of these substances from 2015 to 2017. This trend is explained by the increase in the flow of wastewater that goes to the wastewater treatment plant and undergoes the activated sludge treatment procedure, in particular, due to the decrease in the flow of wastewater that was discharged without treatment. Along with this, the accumulated POPs masses for 2017 are significantly larger than the similar annual ones, and in the case of untreated wastewater, the difference is 46 times, and in the case of treated wastewater and activated sludge

– almost 7 times.

That is why it is important to determine the accumulated masses of PCDD/F in the activated sludge, which does not enter the sea basin, but accumulates on sludge sites and in the soil and plant environments. Based on the fact that the share of activated sludge is 1% of the volume of wastewater, and the humidity of activated sludge is 99.3% [28], we determined the volume of WAS formation, as well as the mass of PCDD/F accumulated in it, the calculation results are shown in table 1.

The obtained data show a continuous increase in the accumulated masses of PCDD/F in activated sludge. According to [28], the annual mass of POPs, which enter the soil environment at sludge sites exceeds the permissible by more than 600 times. However, taking into account the cumulative effect of these substances and their half-life period shows that the concentration of POPs over 2007-2017 exceeds the maximum limit value by at least 4261 times, which makes WAS a source of excessive environmental pollution, especially in the case of disposal outside the sludge sites. In particular, any use of WAS, especially as fertilizers, is unacceptable, and this type of waste requires the development of special methods of handling it.

The comparative histogram of the intake of POPs into the environment of Odesa IUA using the example of 2017 is shown in Fig. 5.

Table 1

Input of PCDD/F in the environment as part of activated sludge

Year	Mass of WAS, t dry residue	EF, mcg TEQ/t dry residue	Mass PCDD/F g TEQ	Accumulated mass of PCDD/F, g TEQ
2007	728.728	200	0.146	0.146
2008	441.441	200	0.088	0.224
2009	4407.403	200	0.881	1.091
2010	4288.284	200	0.858	1.876
2011	5143.138	200	1.029	2.780
2012	5353.348	200	1.071	3.666
2013	5458.453	200	1.092	4.514
2014	6348.342	200	1.270	5.484
2015	6068.062	200	1.214	6.333
2016	6257.251	200	1.251	7.164
2017	5829.824	200	1.166	7.854

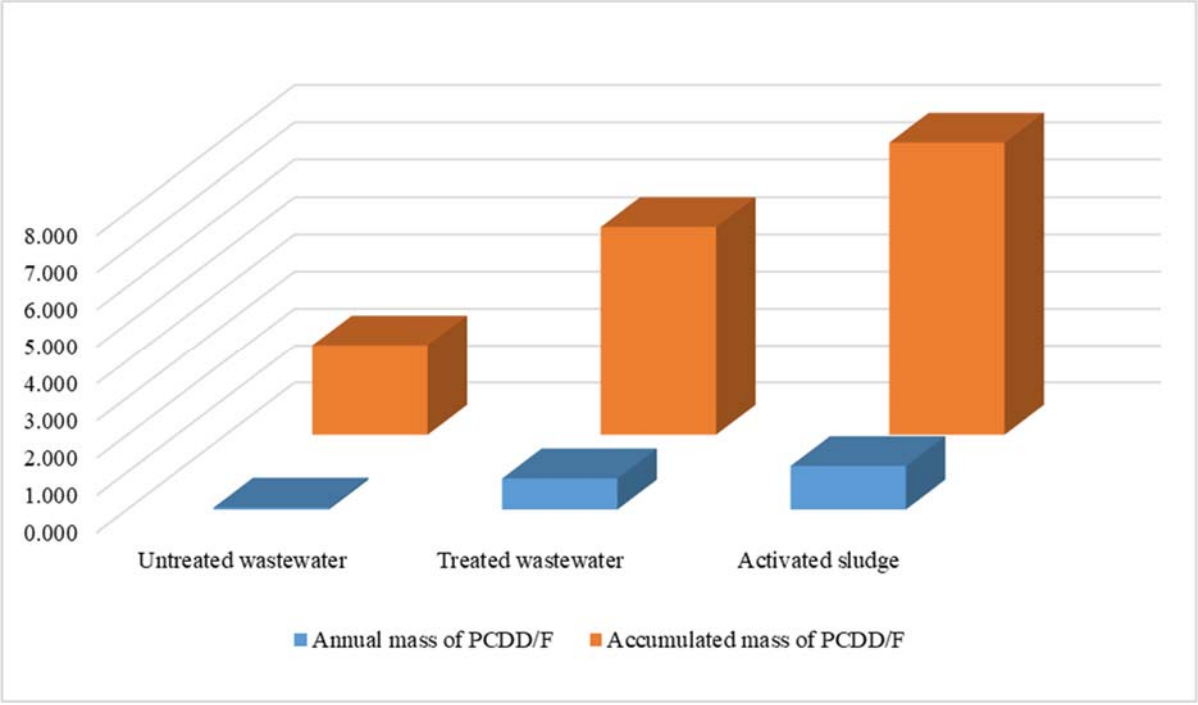

Fig. 5. Comparative histogram of the input of PCDD/F in the environment of Odesa IUA, 2017 (g TEQ)

Fig. 5 shows that the largest mass of PCDD/F is concentrated in WAS, and taking into account the cumulative effect of PCDD/F and their half-life period increases the results obtained by us by almost an order of magnitude, which indicates that the use of methodologies [7, 14] leaves significant amounts of

PCDD unestimated /F, which enter the marine environment and can accumulate in hydrobionts, as well as accumulate in bottom sediments, thereby remaining a source of possible secondary pollution of the water column under certain hydrological conditions.

Even though wastewater treatment by activated sludge is a recognized type of wastewater treatment from PCDD/F, it doesn't solve the problem of POPs pollution, because these substances just redistribute from the water environment to the soil one.

It is also worth mentioning that the mass of PCDD/F that enters water bodies as part of treated wastewater shows that biological treatment does not ensure full and complete treatment of wastewater from POPs, which requires the introduction of new, more effective methods of wastewater treatment.

Conclusions. As a result of the research, the following conclusions can be drawn:

– Due to the impossibility of regular instrumental monitoring of PCDD/F entry into the water environment, the use of calculation methodologies for determining the volumes of these substances to assess their intake into the water environment is a the only and necessary possible condition for satisfying the requirements of the Stockholm Convention;

– The use of existing European methodologies makes it possible to obtain data only for the current year, while the use of suggested by us method allows for obtaining accumulated concentrations for the current year, considering those concentrations that were formed over the previous years, taking into account the cumulative effect and half-life period of PCDD/F;

– The largest annual mass of PCDD/F that enters the aquatic environment comes as part of treated wastewater and amounts to 0.832 g TEQ (on the example of 2017), which is explained by the increase in the treated wastewater discharge due to a decrease of untreated wastewater discharge volumes into water bodies;

– The accumulated masses of PCDD/F entering the water environment of the Odesa IUA during the discharge of both treated and untreated wastewater is 8,018 g TEQ;

– The largest mass of PCDD/F is concentrated in WAS. The accumulated mass for the period 2007-2017 exceeds the maximum threshold by at least 4261 times, which makes WAS a source of excessive pollution of the environment, especially in the case of disposal outside the sludge sites and requires the development of new environmentally safe methods of neutralization and disposal of WAS;

– To confirm the obtained results, it would be expedient to implement instrumental measurements of PCDD/F concentrations in wastewater and WAS;

– Biological treatment does not provide full and complete wastewater treatment against POPs, it requires the introduction of new, more effective methods of wastewater treatment.

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Authors Contribution: All authors have contributed equally to this work

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Біологічні очисні спортування як джерела забруднення навколишнього середовища стійкими органічними забруднювачами (на прикладі Одеської промислово-міської агломерації)

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Ефективні механізми очищення стічних вод (СВ) є важливою складовою зменшення антропогенного навантаження на довкілля. Одним з найрозповсюдженіших механізмів очищення міських стоків є біологічна очистка, яка відбувається на станціях біологічної очистки (СБО). Однак збільшення номенклатури полютантів, які зосереджені у міських СВ, значно впливає на ефективність очищення СВ вод на станціях СБО, які не були призначені для такого широкого спектру специфічних полютантів, зокрема стійкими органічними полютантами (СОП), контроль надходження яких у довкілля необхідно регулювати згідно до вимог Стокгольмської конвенції. Метою дослідження є оцінка обсягу надходження поліхлорованих дибензо-п-діоксинів та дибензофуранів (ПХДД/Ф) у довкілля зі СВ Одеської промислово-міської агломерації (ПМА) та визначення обсягів їх накопичення у морському середовищі. Для розрахунку обсягів надходження ПХДД/Ф у довкілля нами було використано оновлені європейські методики по інвентаризації викидів забруднюючих речовин, для розрахунку накопичення – використано вдосконалену авторами методику розрахунку накопичення ПХДД/Ф з врахуванням їх кумулятивного ефекту та періоду напіврозпаду. В роботі визначено річні маси ПХДД/Ф, які надходять у морське середовище у складі очищених, недостатньо очищених та неочищених СВ; встановлено маси ПХДД/Ф, які накопичуються у відпрацьованих активних мулах (ВАМ) на СБО; визначено обсяги та особливості накопичення ПХДД/Ф у морському басейні, в який здійснюється скидання СВ Одеської ПМА; визначено масу та концентрацію ПХДД/Ф, іммобілізовану у ВАМ, ступінь перевищення концентрації ПХДД/Ф у ВАМ порівняно з ГДК і сумарне накопичення ПХДД/Ф у морському середовищі за період 2007-2017 рр. Встановлено, що використання розрахункових методик для визначення обсягів ПХДД/Ф у водному середовищі є необхідною та єдиною доступною умовою виконання вимог Стокгольмської конвенції через неможливість здійснення регулярного інструментального моніторингу надходження ПХДД/Ф у водне середовище. Використання запропонованої нами вдосконаленої методики розрахунку накопичення ПХДД/Ф з врахуванням їх кумулятивного ефекту та періоду напіврозпаду дозволяє враховувати ті маси ПХДД/Ф, які накопичилися на рік, що розглядається, приймаючи до уваги ті маси ПХДД/Ф, які утворилися у попередні роки.

Ключові слова*: стійкі органічні полютанти, поліхлоровані дибензо-п-діоксини та дибензофурани, біологічна очистка стічних вод, стічні води, відпрацьовані активні мули.*

Внесок авторів: всі автори зробили рівний внесок у цю роботу

Надійшла 3 листопада 2022 р.
Прийнята 15 березня 2023 р.