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THEORETICAL RESEARCH OF THE PROCESSES TO REDUCE THE CONCENTRATION OF CHLORINATED COMPOUNDS WITH NATURAL FIBERS

Ieva ANDRIULAITYTĖ*, Marina VALENTUKEVIČIENĖ, Aušra ZIGMONTIENĖ

Department of Environmental Protection and Water Engineering,
Vilnius Gediminas Technical University, Saulėtekio al. 11, Vilnius, Lithuania

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Abstract. Disinfectants based on chlorine are the most commonly applied biocides for public spaces disinfection. Various studies show the use of sodium hypochlorite went up several times during the pandemic. Chlorinated compounds after they enter stormwater react with organic, inorganic and anthropogenic pollutants there and form harmful disinfection by-products. The article aims to present theoretical research of the processes linked to reduce the chlorinated compounds concentrations in stormwater. Research theory is based on the scientific information and the experiments' results. The equations of mass transfers are used to describe the process. Analysis is focused on the case when chlorinated compounds concentrations in stormwater change depending on mass transfer from solution to filter fiber. Experiments present that after filtration with natural fibers the amount of disinfectants in stormwater decreased approximately 60 percent. The values of mass transfer reveal the efficiency of hemp and peat fibers to transport chlorinated compounds ($C_{22}H_{48}ClN$). Research found out the values of different filter materials are quite similar: 0.20 $D_{ef}\cdot 10^6$, cm^2s^{-1} (hemp) and 0.19 $D_{ef.}\cdot 10^6$, cm^2s^{-1} (peat).

Keywords: chlorine, disinfectants, stormwater, mass transfer.

Introduction

Theoretical research aims to justify adsorption by mass transfer processes of chlorinated compounds using natural fibers. Recent studies show adsorption and mass transfers are usually used to describe pollutants migration from luiquid medium (Georgin et al., 2021; Wen-Pei et al., 2022). Scientific information notes, that adsorption process is a simple, effective and less expensive way to remove organic compounds from environment, including water bodies (Gao et al., 2020; Mateos-Cardenas et al., 2021; Al-Hashimi et al., 2021; Kumar & Gupta, 2020; Veclani et al., 2020).

Public spaces and surfaces cleaning by chlorine based disinfectants is an effective measure to avoid viruses (Li et al., 2021). Chlorine compounds were detected in rivers (Valentukevičienė et al., 2018). Study present chlorine compounds in water have a regression relation by correlation with biochemical oxygen demand, dissolved oxygen concentration as well as dry residue, sulphite, phosphorus and chromium concentrations.

Latest research note the most widely applied biocide to kill microorganisms and viruses is sodium hypochlorite (Rutala & Weber, 2019). Research reports sodium hypochlorite (NaOCl) is assigned to chemical substances with longer residual effect (up to 10 min). This means, that after the solvent of the disinfectant evaporates, the active chlorine based substance remains on the surface and protect it (Chu et al., 2021). How long chlorine will stay in active form depends on the amount of sodium hypochlorite used for surfaces cleaning. Some studies present, for instance, during the twelve hours disinfection process with chlorine amount of 6700 mg/l was occurred the amount of active chlorine about 21.0–25.0 mg/l, which has caused the formation of various harmful compounds (Zhang et al., 2020). Considering the chemical properties NaOCl is described as a solution formed by chlorine reaction with a sodium hydroxide. When chlorine based disinfectants are flushed by water from surfaces, active chlorine reacts with water and form hypochlorous [HOCl] and hydrochloric acids [HCl]. HOCl further dissociates into hypochlorite (OCl–) and hydrogen (H+) ions depending on the solution's pH (Parveen et al., 2022; Paul et al., 2021). OCl ions decompose to a mixture of chloride and chlorate ions, where the chlorate ions are considered as toxic and the chloride is considered as nontoxic. OCl and HOCl are strong agents able to oxidize the sulfhydryl groups of enzymes

* Corresponding author. E-mail: *ieva.andriulaityte@vilniustech.lt*

causing disruption of DNA and protein synthesis (Estrela et al., 2002; Fukuzaki, 2006; Jones & Joshi, 2021).

Environment pollution by chlorinated compounds is a new research direction because of their negative impact on surface water bodies. Research found out, that disinfectants based on chlorine react with water‘s organic, inorganic and anthropogenic pollutants and form disinfection by-products such as haloacetic acids, trihalomethanes, dibromochloromethane, bromodichloromethane, tribromoethane, dichloroacetonitrile, chloramines and other organic compounds. These chemical substances are toxic, carcinogenic and mutagenic for aquatic flora and fauna and cause ecological imbalance (Chen et al., 2021; Ghafoor et al., 2021).

Theoretical research were carried out aiming to analyse the possibilities to reduce concentrations of chlorinated compounds in stormwater. Investigations are based on the scientific information and experiments' results (Valentukevičienė & Najafabadi, 2020). It is analysed the process to remove chlorinated compounds from stormwater by natural fibers (hemp, peat).

Studies report organic pollutants including chlorinated compounds are usually analysed by adsorption isotherm. Mostly adsorption models are described using Langmuir and Freundlich isotherms (Chen & Lo, 2022). Langmuir isotherm is commonly used to describe the homogeneous adsorption process and for modeling equilibrium data (Guo & Wang, 2019). It is mathematically acceptable, easy integrated and used for study heterogeneous adsorption in line with energy distribution functions and heterogeneity descriptions. However, research notes Langmuir isotherm does not accurately describe the adsorption process, especially in heterogeneous adsorption systems. Freundlich isotherm describes the adsorption process of heterogeneous substances (organic compounds) in heterogeneous adsorbents, when the adsorption capacity of homogeneous adsorbent is not used. Freundlich isotherm is widely used because its accuracy and is recommended to describe heterogeneous adsorption (Zhao et al., 2013). To compare with Langmuir isotherm, it provides much more accurate results in a wide range of heterogeneous adsorption systems. The weakness of Freundlich isotherm is its incompatibility with Henry's law and this not allow to describe the process. When $q \to 0$, the isotherm acquires thermodynamic incompatibility. In this research to anlayse chlorinated compounds migration in porous materials is used mass transfer equation and it is focused on the case when concentrations of chlorinated compounds change during the processes of mass exchange.

A multifunctional case is applied where concentrations of chlorinated compounds change depending on the variations in the amount of material absorbed by natural fiber under the limiting conditions. The multifunctional transfer is described as follows: first, as the external mass transfer of the solute molecules from the solution to inner fiber's particle. In this case, the process is fast and expands practically without any resistance, while the internal diffusion remains beyond the detection limit. Second, the diffusion occurred within fiber particle. In this case, the transfer of the molecular membrane takes a few minutes and internal diffusion can last up to several hours. Following scientific information a more porous fiber is characterized by the larger absorption surface area and by the more developed internal structure, forming a whole network of pores and channels, extended inside the particle (Gan & Gan, 2020). This explains the appearance of internal attractive forces and the dependence on internal diffusion in fiber's structure. The theory of chlorinated compounds solution transfer in homogeneous unsaturated natural fiber is based on the assumption that disinfectants dissolve and transport chlorine compounds to the depth of the fiber. The research is based on the sorption analysis by natural fibers and different concentrations of solutions. The one-dimensional flow is extended to the two-dimensional case, where the humidity of fiber surface and the boundary concentration of the removed material change. Theoretical research are defined according to these elements: mass transfer equation (to describe mass transfer and internal diffusion. It also includes mass transfer parameters, that determine the rate of sorption), equilibrium equations (to describe the material balance. The integrity of system is defined by continuity equations), substances removed by the system (natural fiber balance is presented as a functional dependence), initial and boundary conditions (to determine the relation of research elements).

All these effects can be identified qualitatively, but there is no ways to measure them quantitatively. Nevertheless, the results of multicomponent transfer can be theoretically predicted. One way to solve such problem is to apply simplifications in order to obtain formulas independent of the influence of the effects listed above. Equilibrium conditions in multicomponent systems might be described by derivative and correlation methods. Derived multi-component transfer process data is obtained from single-component process data (Zhang et al., 2022). Theoretical research reliability is assessed by the data of laboratory experiments focused on the analysis of stormwater samples contaminated by chlorine based disinfectants.

1. Materials and methods

Theoretical research are carried out using experimental data obtained in laboratory conditions. It is analysed how the passing flow impact fiber's humidity and how the boundary conditions of chlorinated compounds concentrations change as a result. The boundary conditions of transfer processes are determined by the following factors:

- contact time when equilibrium is established. It is determined by analysing transfer under laboratory conditions. A natural fiber is used to transport dide-

cyldimethylammonium chloride (C22H48ClN). The sorption time is taken no more than 1 hour in order to ensure the retention of the disinfectant within fiber.
- Natural fiber sorption capacity in relation to didecyldimethylammonium chloride and the required amount of natural fiber to reduce the concentration of chlorinated compounds to the limit according by national legislation. Laboratory experiments revealed the changes of chlorinated compounds concentrations impact on stormwater pollution indicators: pH, turbidity, color, conductivity and active chlorine concentrations. Research find out that filtration decrease chlorine concentration to value below 0.01 mg/l (detection limit) (Valentukeviciene et al., 2022).

Theoretical research equations are solved numerically or analytically. The initial data consists of the system variables and the final data are the kinetic data. It is verified how the theoretical research correspond to the experimental results. With the aim to describe the theoretical research equations, it is assumed that the transfer continues from the beginning of the process until the equilibrium between the concentration of chlorinated compounds in water and the concentration of the removed substance in fiber. The establishment of equilibrium represents the end of the static transfer process and reflects the ability of the fiber to absorb the available solute (organic chlorine). The transport equation is characterized by the values of their constants. The mass transfer equation looks like this:

$$N_i = k_i \cdot A \cdot \left(C_i - C_s\right), \tag{1}$$

where: A – constant, $C_i - C_s$ – equilibrium concentration, mg/l, k_i – mass transfer coefficient.

Theoretical research of chlorinated compounds removal from stormwater by natural fiber is an extension of the already known single-component transfer process. Noteworthy is not simple extension in present case, because of several forms of undesirable chlorinated compounds impurities in stormwater. It is important to evaluate following cases:

Reaction of removed substance with other substances characterized by different chemical and physical properties may take place in the solution (e.g. chlorinated compounds with organic substances) and continue on the fiber surface. This reduces the diffusivity of solution and impacts the resistance in the transmission system.

Variations of absorption capacity or homogeneity may occur within a fiber mass transfer space as regards to space increase. Such changes in the transmission system cause the new position of the transmission equilibrium point and impacts fiber integrity.

Irreversibility is the formation of permanent chemical bonds between the contact spaces and functional groups on fiber surface. Such connections disrupt leaching process and some fiber sides remain blocked. This causes an unpredictable influence on the transfer process becaus not all fiber particles are equally open. Irreversibility must be revealed by examining the transfer equilibrium during laboratory experiments. The transfer equilibrium curve usually not match and present the formation of irreversible bonds.

Molecular sieves effect occurs when diffusion in a natural fiber is blocked by relatively large molecules (e.g. chlorine and organic substances complexes). This excludes large molecules from the diffusion process within fiber micropores. This makes some areas of the fiber unsuitable for certain molecules, depending on their sizes.

This methods provide with the ideal transmission, i.e. the components are removed separately and independent of diffusion. The basics methods are:

1. The formulas are extended empirically: formulas for single-component functional dependencies are extended to predict multi-component transmission. Mathews' extension of Redlich Peterson's formula looks like this:

$$q_{si} = \frac{K_{Ji} C_{Si}}{1 + \sum_{j=1}^{n} b_{Jj} C^{\beta_{Jj}}{}_{Sj}}, \tag{2}$$

where K_J, b_J ir β are the one-component Redlich-Peterson constants.

Eq. (2) is based on theoretical classical formulas, but does not give very accurate results. It is proposed to carry out several laboratory experiments and to decide on their applicability to the absorption properties of natural fibers, removing chlorinated compounds from stormwater and making theoretical and practical functional dependencies.

2. "IAS" theory is a thermodynamic method independent of the type of isotherms used. It is the equivalent of Raoults law applied to solid-liquid systems. It states that several components are simultaneously removed from the aqueous solution, forming an ideal solution phase. The resulting mathematical expression is:

$$C_i = C_i^o Z_i \tag{3}$$

and

$$C_T = \sum_{i=1}^{n} C_i^o Z_i, \tag{4}$$

where: C_i^o – solute concentration in liquid phase, in the presence of a one component solution, mg/l; Z_i – molar concentration in the adsorbed phase, C_i – concentration of the liquid phase in the mixture, mg/l; C_T – total concentration of chlorine, mg/l.

Theoretical research can only be applied to systems linked to very dilute solutions and in the presence of very small fiber molecules, when their interrelationships are negligible. In transport systems with high concentrations of solutions and large molecules (molar mass ≥300 g/mol), such a theory cannot be applied, as inaccurate descriptions of the transport process are obtained for mathematical equations.

Correlation method is used to describe multicomponent transport processes. Often, the ideal transfer is represented with a slight mixing effect that does not correspond to the real transfer system (Graveleau et al., 2017). The lack of appropriate mathematical formulas does not allow to express mathematically the effects of the mentioned multicomponent solutions, which forces in many cases to use experimental data to evaluate them. This forms the basis for the correlational method. Data from single-component and multi-component transfer processes are used to construct interaction factors. Thus, is obtained the formula for the isotherm of the multicomponent transfer process. An improved extended isotherm with an interaction factor η that minimizes the differences between experimental and theoretical multicomponent transport data, i.e. when the following formula is extended (Girish, 2017).

Here is presented a transmission system of n number of components, the composition of which includes p experimental points, q_{ical} can be calculated from any well-known formula.

$$q_{ical} = \frac{K_{Li}(C_{si}/\eta_i)}{1+\sum_{j=1}^{n} a_{Lj}(c_{sj}/\eta_j)}, \tag{5}$$

where η_i is unique to each solution component i and is dependent on other fiber components. Such a method can also be applied using the Redlich-Peterson functional dependence:

$$q_i = \frac{K_{Ji}(C_{si}/\eta_i)}{1+\sum_{j=1}^{n} b_{Jj}(C_{sj}/\eta_j)^{\beta_j}}. \tag{6}$$

This formulation gives better results in transfer systems with finely dispersed organic molecules or weak solutions. This method allows to improve functional dependence, but cannot include all constants for different components in one formula. This may result in some inaccuracy.

2. Results and discussions

This section presents the research results based on chlorinated compounds transfer by natural fibers (hemp, peat). Sample tests collected from permanently disinfected areas. Transfer processes analysed at laboratory conditions using two different fibers: hemp and peat. Mass transfer values are determine. The mass transfer values defining fiber's efficiency to transport chlorine compounds see in Table 1.

Experimental research carried out at the laboratory using stormwater and chlorinated compound ($C_{22}H_{48}ClN$). Chlorinated compound concentrations varied from 5–25 ml/l. These concentrations were used aiming to analyse low quantities relation to surface disinfections as well following World Health organization recommendations (WHO, 2020). With the aim to find out and define which material could be used to transfer chlorine compounds have been tested two fibers: hemp and peat. Both materials selected according their physical parameters and economic indicators (recycled, cheap material with high sorption capacities).

Table 1. Mass transfer values of different fibers

Chlorineated compound	Fiber	Concentration, ml/l	$D_{ef.}\cdot 10^6$, cm^2s^{-1}
$C_{22}H_{48}ClN$	Hemp	5–25	0.20
$C_{22}H_{48}ClN$	Peat	5–25	0.19

Results showed, that chlorinated compounds' transfer depends on fiber structure, particle size, pore dimensions, pore volume, specific surface area of the fiber and type on transferred material (C22H48ClN). The efficiency of natural sorbents for stormwater treatment analysed in research Andriulaitytė and Valentukevičienė (2022) and Fridrick and Valentukevičienė (2021). It was found out that, for example, hemp has high efficiency to remove pollutants from stormwater and reduce the following indicators: conductivity, pH, color, turbidity as well. Table 1 present the transmission constants obtained during testing hemp and peat under laboratory conditions with C22H48ClN. To be noted these both materials have good transport properties and usually are applied to determine mass transfer values (Gumnitsky et al., 2020; Liu & Beckerman, 2022). Mass transfer values are similar: hemp transfer value is slightly higher when peat value. Research present both materials could be used to remove chlorinated compounds from stormwater.

Based on theoretical research and the results of laboratory experiments is prepared the characteristic of disinfectant amount transfer in solution by the residual COD (mgO_2/l) (Figure 1).

Figure 1 reveals all three isotherms could be used to describe chlorinated compounds transfer within fiber (hemp, peat). In order to obtaine more accurate data is recommended to verify theoretical and experimental results using other materials (i.e. wood chips, activated carbon and etc.). This would allow to determine transfer values and would provide with information which fiber transfers chlorinated compounds more efficient and is recommended to use for pollutants removal. Research reveal the probability of enzymatic hydrolysis of investigated chlorinated compounds lead to intermediate organic monomers and can be identified such as chloramines etc.

The first reults of chlorinated compounds transfer modelling have shown the relatively normal level of correlation R-squared is 0.715 (Table 2). It was used sixteen test samples for analysis. Regression value determine the strength of the relationship between one dependent variable and a series of other variables.

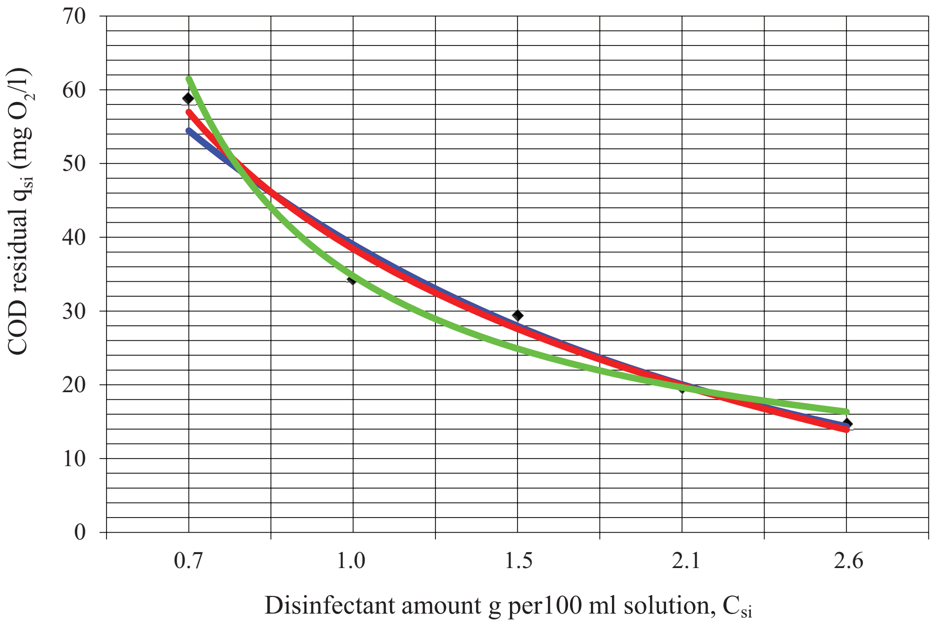

Figure 1. Chlorinated compounds adsorbtion isotherm
(theoretical results: — Langmuir, — Freundlich, — Redlich-Peterson; • experimental results)

Table 2. Chlorinated compounds transfer modeling

Dep. Variable	Cl	R-squared	0.715
Model:	OLS	Adj. R-squared	0.671
Method	Least squares	F-statistic	16.33
No. Observations	16	AIC	–119.6
Df Residuals	13	BIC	–117.2
Omnibus	1.150	Durbin-Watson	1.247
Pro(Omnibus	0.563	Jarque-Bera (JB)	0.506
Skew	0.435	Prob (JB)	0.776
Kurtosis	2.937	Cond. No	1.04+03
coef.	0.0436	std. err.	0.004
t	12.185	P>|t|	0.000

The Durbin Watson test is used to check autocorrelation in the residuals of a statistical regression analysis. (Draper & Smith, 2014). Table 2 indicates value 1.247 by the Durbin – Watson statistic. According scientific sources values from 0 to 2 point present positive autocorrelation. Modelling results show autocorrelation is relatively normal in this case. Scientific sources recommend to use the Jarque – Bera test for data analysis (Aslam et al., 2021). Jarque-Bera test define if data matches a normal distribution. It is evaluated by the skewness and kurtosis of data. The formula for the Jarque-Bera test statistics is:

$$JB = n[(\sqrt{b_1})^2 / 6 + (b_2 - 3)^2 / 24], \tag{7}$$

where: n – sample size, $\sqrt{b_1}$ – skewness coefficient, b_2 – kurtosis coefficient.

According the scientific sources the normal distrubution has kurtosis three and skeweness is zero. To obtaine more accurate data and to verify how regression and correlations are related to changes of possible variables is recommended to compare theoretical and experimental results by mathematic modeling.

Conclusions

1. Adsorption is an effective and less expensive way to remove chlorine compounds from stormwater. Experiments revealed that after test samples filtration with natural fibers (hemp, peat) disinfectants amount in stormwater decreased around 60 percent.
2. Theoretical research resulted the efficiency of chlorine based disinfectants ($C_{22}H_{48}ClN$) transfer from solution to filter depend on fiber material. The mass transfer values showed hemp and peat can be used to remove chlorine compounds from stormwater. Research present chlorine compounds ($C_{22}H_{48}ClN$) mass transfer values are similar: 0.20 $D_{ef.}\cdot10^6$, cm^2s^{-1} (hemp) and 0.19 $D_{ef.}\cdot10^6$, cm^2s^{-1} (peat).
3. In order to find out the influence of filter materials on the decrease of stormwater pollution further experiments should be carried out using other fibers such as sawdust and etc. This would allow to select of the most effective filter material for chlorinated compounds removal from stormwater.
4. Enzymatic hydrolysis of chlorinated compounds to intermediate organic monomers can be indentified such as chloramines etc.

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Contribution

Conceptualization, I.A. and M.V.; methodology, A.Z.; validation, I.A., M.V. and A.Z.; formal analysis, A.Z.; investigation, I.A.; resources, M.V.; writing–original draft preparation, I.A.; writing–review and editing, M.V., A.Z. and I.A.; visualization, I.A., A.Z; supervision, M.V. All authors have read and agreed to the published version of the manuscript.

Disclosure statement

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