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Treatment of drug residues (emerging contaminants) in hospital effluent by the combination of biological and physiochemical treatment process: a review

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Nadeem Ahmad
Civil Engineering, Jamia Millia Islamia Central University, New Delhi, India
Sirajuddin Ahmed
Jamia Millia Islamia Central University, New Delhi, India
Viola Vambol
Department of Applied Ecology and Environmental Sciences, Yuri Kondratyuk Poltava Polytechnic, National University, Poltava, Ukraine, and
Sergij Vambol
Life Safety and Law Department, Kharkiv Petro Vasylenko National Technical University of Agriculture, Kharkiv, Ukraine

Abstract

Purpose – All those effluent streams having compromised characteristics pose negative effects on the environment either directly or indirectly. Health care facilities and hospitals also generate a large amount of effluent like other industries containing harmful and toxic pharmaceutical residual compounds due to uncontrolled use of drugs, besides others. The occurrence of antibiotic in the environment is of utmost concern due to development of resistant genes. These get mixed up with ground and surface water due to lack of proper treatment of hospital wastewater. The effect of pharmaceutical compounds on human society and ecosystem as a whole is quite obvious. There are no strict laws regarding discharge of hospital effluent in many countries. Contrary to this, the authors do not have appropriate treatment facilities and solution to solve day by day increasing complexity of this problem. Moreover, water discharged from different health facilities having variable concentration often gets mixed with municipal sewage, thus remains partially untreated even after passing from conventional treatment plants. The purpose of this paper is to highlight the occurrences and fate of such harmful compounds, need of proper effluent management system as well as conventionally adopted treatment technologies nowadays all around the globe. This mini-review would introduce the subject, the need of the study, the motivation for the study, aim, objectives of the research and methodology to be adopted for such a study.

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Design/methodology/approach – Hospital effluents consisting of pathogens, fecal coliforms, *Escherichia coli*, etc, including phenols, detergents, toxic elements like cyanide and heavy metals such as copper (Cu), iron (Fe), gadolinium (Gd), nickel (Ni), platinum (Pt), among others are commonly detected nowadays. These unwanted compounds along with emerging pollutants are generally not being regulated before getting discharged caused and spread of diseases. Various chemical and biological characteristics of hospital effluents are assessed keeping in the view the threat posed to ecosystem. Several research studies have been done and few are ongoing to explore the different characteristics and compositions of these effluent streams in comparison so as to suggest the suitable conventional treatment techniques and ways to manage the problem. Several antibiotic groups such as ciprofloxacin, ofloxacin, sulfa pyridine, trimethoprim, metronidazole and their metabolites are reported in higher concentration in hospital effluent. The aquatic system also receives a high concentration of pharmaceutical residues more than 14,000 μg/L from treatment plants also and other surface water or even drinking water in Indian cities. Many rivers in southern parts of India receives treated water have detected high concentration drugs and its metabolites. As far as global constraints that need to be discussed, there are only selected pharmaceuticals compounds generally analyzed, issue regarding management and detection based on method of sampling, frequency of analysis and observation, spatial as well as temporal concentration of these concerned micropollutants, accuracy in detecting these compounds, reliability of results and predictions, prioritization and the method of treatment in use for such type of wastewater stream. The complexity of management and treatment as well need to be addressed with following issues at priority: composition and characterization of effluent, compatible and efficient treatment technology that needs to be adopted and the environment risk posed by them. The problem of drugs and its residues was not seen to be reported in latter part of 20th century, but it might be reported locally in some part of globe. This paper covers some aspect about the disposal and regulatory standard around the world toward hospital effluent discharge, its managements and treatment technologies that are adopted and best suitable nowadays various industries and monitoring the efficiencies of existing treatment systems. This mini-review would introduce the subject, the need, the motivation and objectives of the study and methodology can be adopted for such a study.

Findings – The compiled review gives a complete view about the types of antibiotics used in different health care facilities, their residue formation, occurrences in different ecosystems, types of regulations or laws available in different counties related to disposal, different type of treatment technologies, innovative combined treatment schemes and future action needed to tackle such type of effluent after its generation. The thesis also highlights the use of certain innovative materials use for the treatment like nanoparticles. It also discusses about the residues impact on the human health as well as their bioaccumulative nature. If the authors relate the past to the current scenario of pharmaceutical compounds (PhACs) in the environment, the authors will certainly notice that many diseases are nowadays not curable by simple previously prescribed Ab. Many research projects have been done in European countries that have shown the risk of such residues like Pills, Sibell, Poseidon, No pills, Neptune, Knappe, Endetech, etc. In the previous section, it was mentioned that there are no stringent laws for hospital wastewater and in many countries, they are mixed with domestic wastewater. Many difficulties are there with this research due to complex analysis, detection of targeted Ab, affecting waterbodies rate of flow, nature of treatment varies with season to season. The way nature is being degraded and harmful effect are being imposed, it is important to take immediate and decisive steps in this area. Wastewater treatment plants (WWTPs) serves as a nursery for antibiotic-resistant systems, hence monitoring with great attention is also needed. Many trials with different treatment process, in combination, were considered. Many countries are paying great attention to this topic by considering the severity of the risk involved in it.

Research limitations/implications – Previous studies by several scientists show that the pharmaceutical residues in the discharged effluent displayed direct toxic effects, and sometimes, detrimental effects in the mixture were also observed. The discharge of untreated effluent from hospitals and pharmaceuticals and personal care products in the natural ecosystem poses a significant threat to human beings. The pharmaceuticals, like antibiotics, in the aquatic environment, accelerate the development of the antibiotic-resistant genes in bacteria, which causes fatal health risks to animals and human beings. Others, like analgesics, are known to affect development in fishes. They also degrade the water quality and may lead to DNA damage, toxicity in lower organisms like daphnia and have the potential to bioaccumulate. A few commonly used nanoadsorbents for water and wastewater treatment along with their specific properties can also be used. The main advantages of them are high adsorption capacity and superior efficiency, their high reusability, synthesis at room temperatures, super magnetism, quantum confinement effect as well as eco-toxicity. This review will focus on the applicability of different nanoscale materials and their uses in treating wastewater polluted by organic and inorganic compounds, heavy metals, bacteria and viruses. Moreover, the use of various nanoadsorbents and nano-based filtration membranes is also examined.

Practical implications – A number of different pharmaceutical residues derived from various activities like production facilities, domestic use and hospitals have been reported earlier to be present in groundwater, effluents and rivers, they include antibiotics, psycho-actives, analgesics, illicit drugs, antihistamine, etc. In past few years environmental scientists are more concerned toward the effluents generated from medical care facilities, community health centers and hospitals. Various chemical and biological characteristics of hospital effluents have been assessed keeping in the view the common threats pose by them to the entire ecosystem. In this study, seven multispecialty hospitals with nonidentical pretreatment were selected for three aspects i.e. conventional wastewater characteristics, high priority pharmaceuticals and microbial analyses. The present work is to evaluate efficacy of advanced wastewater treatment methods with regard to removal of these three aspects from hospital effluents before discharge into a sewage treatment plant (STP). Based on test results, two out of seven treatment technologies, i.e. MBR and CW effectively reducing conventional parameters and pharmaceuticals from secondary and tertiary treatments except regeneration of microbes were observed in tertiary level by these two treatments.

Social implications – This review has aimed to identify the emerging contaminants, including pharmaceutical residues, highly consumed chemicals that are present in the hospital effluent, along with their physicochemical and biological characteristics. In this, the main objective was to review the occurrences and fate of common drugs and antibiotics present in effluents from hospital wastewaters. As far as global constraints that need to be discussed, there are only selected pharmaceuticals compounds generally analyzed, issue regarding management and detection based on method of sampling, frequency of analysis and observation, spatial as well as temporal concentration of these concerned micropollutants, accuracy in detecting these compounds, reliability of results and predictions, prioritization and the method of treatment in use for such type of wastewater stream are among the major issues (Akter *et al.*, 2012; Ashfaq *et al.*, 2016; García-Mateos *et al.*, 2015; Liu *et al.*, 2014; Mubedi *et al.*, 2013; Prabhasankar *et al.*, 2016; Sun *et al.*, 2016; Suriyanon *et al.*, 2015; Wang *et al.*, 2016; Wen *et al.*, 2004). This paper covers some aspect about the disposal and regulatory standard around the world toward hospital effluent discharge, its managements and treatment technologies that are adopted and best suitable nowadays.

Originality/value – This study many multispecialty hospitals with nonidentical pretreatment were selected for three aspects i.e. conventional wastewater characteristics high priority pharmaceuticals and microbial analyses. The present work is to evaluate efficacy of advanced wastewater treatment methods with regard to removal of these three aspects from hospital effluents before discharge into an STP. Based on test results, two out of different treatment effectively reducing conventional parameters and pharmaceuticals from secondary and tertiary treatments except regeneration of microbes were observed in the tertiary level by these two treatments were studies followed by ozonation and ultraviolet-ray treatment.

Keywords Characteristics, Drugs, Residues, Emerging, Contaminants, Hospital effluent, Treatment process

Paper type General review

1. Introduction

Fecal coliforms, *Escherichia coli*, pathogens, etc. as well as copper (Cu) and cyanide, nickel (Ni), iron (Fe), phenols, detergents and other toxic elements are founded in current hospital wastewater (HWW) (Cosgrove *et al.*, 2018; Kalhor *et al.*, 2019; Panwar and Ahmed, 2018; Tomenko *et al.*, 2007). These undesired compounds, along with emerging pollutants, usually are not extracted before discarding (Babbar *et al.*, 2017; Huang, 2010; Nigam and Srivastava, 2020; Papa *et al.*, 2015; Roy *et al.*, 2020). Many biological and chemical indicators of hospital sewage are taken into account to view the threat posed to the natural components (Diwakar *et al.*, 2015; Eum *et al.*, 2019; Mahanta *et al.*, 2015; Shekhar *et al.*, 2015).

Various studies' types have been carried out, and only a few continuing to investigate the most important characteristics and proportions of these sewages in comparison, in order to develop suitable conventional treatment methods and solving the problems' ways. Several antibiotic groups such as ciprofloxacin, ofloxacin, sulfapyridine, trimethoprim, metronidazole and their metabolites are reported higher in sewage from health facilities (Machiwal and Jha, 2015; Shekhar *et al.*, 2015; Watto and Mugera, 2015). More than 14,000 μg/l of pharmaceutical residues fell into water bodies from sewage treatment plants (STPs) and other polluted surface waters (Chen *et al.*, 2014; Kulkarni *et al.*, 2015; Shahul Hameed *et al.*, 2015). It should be emphasized that many rivers in southern India that receive processed water contain high concentration medical supplies and their metabolites (García-Ávila *et al.*, 2020; Guilherme and Rodriguez, 2015; Li *et al.*, 2017; Saeed *et al.*, 2012).

With regard to global constraints requiring discussion, only selected pharmaceuticals were the focus of attention. With regard to the management and detection of pharmaceuticals, sampling has been carried out in accordance with the required observation frequency; the spatial and temporal concentration of micropollutants were taken into account; the detection accuracy, the results' reliability and the forecast presented were analyzed. In addition, the priorities and the applied method of wastewater treatment of a certain type are taken into account. (Bond *et al.*, 2010; Leavey-roback *et al.*, 2016; Legay *et al.*, 2015; Serrano *et al.*, 2015; Wang *et al.*, 2015). Determination of wastewater composition and characterization, selection of treatment technologies that are compatible and effective for implementation, and the environmental risk associated with the use of these technologies are priorities that need to be quickly addressed by effective management methods (Bull *et al.*, 2011; Chen and Westerhoff, 2010; Hanigan *et al.*, 2016; Richardson and Postigo, 2016; Shah and Mitch, 2012; Wei *et al.*, 2010; Xie *et al.*, 2016). The problem of pharmaceuticals in the environment and their residues was not urgent in the second half of the 20th century, however, this problem can be highlighted in some parts of the world in a local way (Chhetri *et al.*, 2016; Dongmei *et al.*, 2015; Feretti *et al.*, 2008; Igbinosa *et al.*, 2013; Xiao *et al.*, 2016). This paper covers various aspects of the regulatory and disposal standard in different countries of the world regarding the discharge of medical facilities' wastewater, these streams' management and way for their treatment, which are currently accepted and best suited. The following works (Abu-shanab, 2013; Garcia-villanova *et al.*, 2010; Yang *et al.*, 2019; Zhai *et al.*, 2014; Zhang *et al.*, 2016) present the results of monitoring existing treatment systems for various industries. This mini-review would introduce the subject, the study's need, the motivation for the task, aim, objectives of the research and methodology that can be adopted for such a study (Awual *et al.*, 2011; Benanou *et al.*, 2010; Dad *et al.*, 2018; Gopal *et al.*, 2007; Kogevinas *et al.*, 2016; Mazhar *et al.*, 2020; Yang *et al.*, 2019).

2. Guidelines for hospital wastewater for its managements

The members of the European Union have adopted their own laws, assessment criteria and disposal ways of health care facilities' wastewater to effectively their manage. This is due to the fact that there are currently no specific guidelines in this area of activity. For example, Germany has established that the medical institutions' wastewater is domestic. In this regard, there is no special permitting procedure for the discharge of this wastewater into the sewer system (Cen *et al.*, 2020; Huang *et al.*, 2020; Lin *et al.*, 2020; Madureira *et al.*, 2020; Manigrasso *et al.*, 2020; Pompilio and Di Bonaventura, 2020). If HWW meets specific characteristics concerning sewage, no further consideration is required regarding the discharge of these streams to wastewater treatment plants. If an Italian medical institution has 50 beds or less, then the wastewater of this institution is discharged into the sewer without additional analysis.

In society's health and care process, the hospital plays a vital role. In the course of health care procedures, various types of unwanted and harmful products are generated. Due to the environmental hazard of biomedical wastes, their management is the most pressing issue in maintaining public health and a favorable environment. In 1998, India promulgated its original regulations on biomedical waste. At the same time, a deadline for their validity was set in December 2000, which was subsequently extended until December 2002. However, the on-ground situation remains far from satisfactory. According to these rules, medical institutions are obliged to sort, ensure effective disinfection and disposal of waste in an environmentally friendly manner. However, few hospitals are sincerely complying with such stipulations (Grasso *et al.*, 2020; Lee *et al.*, 2020; De Matteis *et al.*, 2018; Mubarakali *et al.*, 2012; Pantidos and Horsfall, 2014; Philip, 2009; Prasad *et al.*, 2007; Vigneshwaran *et al.*, 2006). The World Health Organization guidelines are very understandable and clear and in document form for medical wastewater as "Safe Management of Wastes from Health-care Activities." It documents collection, treatment and separates medical wastewater into three streams:

(1) A blackwater has a high content of waste from toilets or urinals (i.e. faeces);

(2) A greywater is water with other types of contamination (no faecal contamination), which is formed after showers, laundries, film cleaning, laboratories, X-rays, etc. and

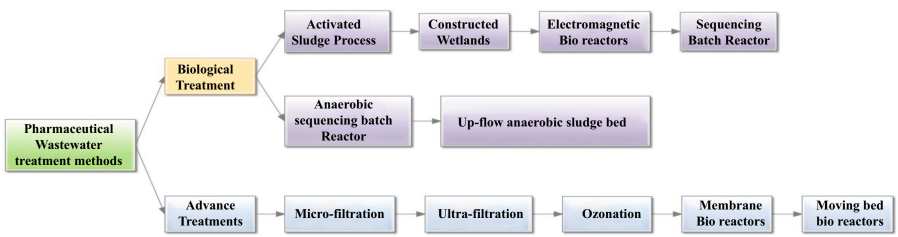
(3) Storm water, which is not classified as wastewater, and is collected from sediments on the Earth surface.

HWWs can be used with benefits for various purposes; however, they usually have different degrees of pollution, which depend on the maintenance of the wastewater discharge and accumulation systems and the tasks that the particular medical institution performs. It is in developing countries that there is the highest degree of environmental risk associated with the release of HWW into the environment. This is due to the fact that uncontrolled discharges of such wastewater occur in practice, which contributes to the penetration of hazardous pollutants into ground and underground aquifers. With that said, it should be summarized that the best way to manage HWWs is to ensure their local sorting into primary, secondary and tertiary (Khan *et al.*, 2019a, b, c, d, e, f, g; Khana *et al.*, 2019). Detailed information on silt disposal and possible water reuse, including the introduction of new and innovative techniques the HWW treatment, is also provided in this document. It also directs to the minimum necessary actions regarding the HWW management, in view of the fact that suitable sanitation facilities are missing in developing countries.

3. Treatment techniques available for PhACs

The effective removal of medical preparations, remnants, cleansers and sanitizers from the HWW needs special attention due to the inhibitory effects during the biological treatment. A membrane bioreactor (MBR) is used in many countries for secondary treatment. However, a sudden increase in formic acid has been noticed due to the presence of certain contaminants in the wastewater from medical facilities. This situation is capable of providing pH shock in the reactor and a decrease in the efficiency of its operation, since the sludge is destroyed. Among the various technologies known today for one of the three treatment steps, the most common are firstly conventional activated sludge (CAS), and secondly, an MBR. Several factors affect the efficiency of removing new pollutants from wastewater, and at the same time, any treatment method has undeniable advantages and disadvantages as shown in Figure 1. However, it is not possible to claim that any of the known technologies are absolutely appropriate for HWW purification as shown in Table 1.

Previous studies by several scientists show that the pharmaceutical residues in the discharged effluent definitely render toxic effects, and in some instances, cumulative harmful effects were also discovered. The discharge of untreated HWW, and pharmaceuticals and personal care products to the natural ecosystem poses a significant threat for human beings. Others, like analgesics, are known to affect growth in fishes. They also degrade the water



Source(s): Changani *et al.*, 2020

Figure 1. Different methods adopted for pharmaceutical effluent treatment

quality and may lead to Deoxyribonucleic acid (DNA) damage, toxicity in lower organisms like daphnia and have the potential to bioaccumulate.

The well-known nanoscale adsorbents can also be used to remove contaminants from wastewater with their special properties. This is promising, since among their many advantages, the main ones are high efficiency due to excellent adsorption capacity, reusability, environmental safety, synthesis at room temperature, supermagnetism and quantum confinement effect (Andersen *et al.*, 2020; Hai *et al.*, 2014; Hard, 2000; Heberer, 2002; Khan *et al.*, 2019a, d; Lotfi *et al.*, 2020; Lu *et al.*, 2020; Thompson *et al.*, 2001). Therefore, this review is focused on the possibility of using some nanoscale materials for the removal of heavy metals, organic and inorganic compounds, and disinfection of viruses and bacteria. In addition to this, the same nanoadsorbents' use and nano-based filtration membranes is being studied.

Several different pharmaceutical residues derived from various activities like production facilities, domestic use and hospitals have been reported earlier to be present in groundwater, effluents and rivers; they include antibiotics, psychoactives, analgesics, illicit drugs, antihistamine, etc.

So the details of some nanoadsorbents given in Table 2 indicate the expediency of their use for the purification of pharmaceutical wastewater.

In the past few years, to see the common threats to the entire ecosystem posed by wastewater generated in health care facilities, public health centers and hospitals, environmental scientists assessed the various chemical and biological characteristics of this wastewater. In current investigation, seven multidisciplinary hospitals, which use the different pretreatment, were selected in accordance with three aspects, namely, conventional wastewater characteristics, microbiological testing and high priority pharmaceuticals. These three aspects are important in order to have a Wastewater Discharge Permit (STP). According to the test results, an effective improvement in conventional parameters and quantitative reduction of pharmaceuticals after secondary or tertiary treatment was observed using two methods, namely, MBR and constructed wetlands (CW), from seven different processing technologies, but with the exception of microbial regeneration.

Table 1.
Removal efficiency of pharmaceuticals from medical institutions' wastewater

Treatment schemes	Effectiveness	Reference
MBR	22%	Kovalova *et al.* (2012), Khan *et al.* (2019g)
	39–60%	Göbel *et al.* (2007)
Three-stage moving bed biofilm reactor (MBBR)	More than 20%	Casas *et al.* (2015)
Sequential aerobic + anoxic/anaerobic treatment	75–100%	Wiest *et al.* (2018)
Filtration + CAS	59–76%	Lien *et al.* (2016)

Table 2.
Removal efficiency of some heavy metals by nanoadsorbents from medical institutions' wastewater

Contaminant	Nanoadsorbent	Effectiveness	Reference
Cr^{6+}	Carbon nanotube supported ceria nanoparticles	30.20 mg/g	Di *et al.* (2006)
	Akaganeite nanocrystals	80 mg/g	Lazaridis *et al.* (2005)
Cd^{2+}	Ascorbic acid-stabilized zero	79.58%	Savasari *et al.* (2015)
As^{5+}	Hematite	$2{,}899 \pm 71.09\ \mu g/g$	Dickson *et al.* (2017)
Cu^{2+}	Chitoson-bound Fe_3O_4 nanoparticles	21.5 mg/g	Khan *et al.* (2019a, b, c, d, e, f, g), Khana *et al.* (2019)

Source(s): Khan *et al.* (2020)

This review aims to identify the emerging contaminants, including pharmaceutical residues, highly consumed chemicals present in the hospital effluent, and their physicochemical and biological characteristics. At the same time, the primary direction of the study was to analyze the factors influencing the emergence and distribution of medicines and antibiotics present in the wastewater of medical institutions.

4. Future scope of research

Application of nanotechnology-based membrane filters will further gain acceptance in future, due to their high efficiency in removing inorganic, organic and biological impurities, with metal selectivity as well as, being durable, low cost and resistant to fouling. Research studies can have future prospect involving below said direction for the attainment of better management of hospital effluent. Research studies can have future prospect involving below said direction for the attainment of better efficiency regarding HWW:

(1) The effect of physiochemical parameters on PhACs concentrations. Aerobic, anaerobic and facultative ponds can have future in the removal of Ab with fewer energy requirements.

(2) Proper studies can be performed about the intermediate compounds that are formed during various treatment processes. Their chemical identity can be established with full-scale investigations.

(3) Sludge matrix on PhACs can also be studied keeping in mind the removal efficiency of the different treatment process.

(4) Degradation of PhACs and Ab in natural environments can be further studied considering the nature and types of microbiological activities taking place.

(5) During degradation process, formic acid and acetic acid formation were reported in many studies which can be further investigated and correlation can be developed with a model.

5. Conclusion and recommendations

The compiled review gives a complete view about the types of antibiotics used in different health care facilities, their residue formation, occurrences in diverse ecosystems, types of regulations or laws available in various counties related to disposal, other type of treatment technologies, innovative combined treatment schemes and future action needed to tackle such kind of effluent after its generation. The thesis also highlights the use of certain innovative materials used for the treatment, like nanoparticles. It also discusses the residues impact on the human health as well as their bioaccumulative nature.

A comparative analysis of early and current PhACs scenarios in the environment shows that at present it is extremely difficult, and sometimes even impossible, to overcome some diseases by simple previously prescribed Ab. Many investigation projects that have been carried out in European countries demonstrate the existence of an environmental risk associated with residues such as Sibell, No pills, Pills, Endetech, Neptune, Poseidon, Knappe, etc. Research also shows that many countries do not have specific legislation regarding pharmaceutical preparations in wastewater, and therefore it is allowed to mix such wastewater with household. Significant difficulties accompany such complex analysis studies due to the fact that it is difficult to detect of targeted Ab that affect the flow rate in water bodies and the treatment type varies depending on the season. Undoubtedly, due to such a situation, the degradation of nature occurs, as a result of which it negatively affects humans, as well as the animal and plant world, which forces us to immediately take decisive action. The overwhelming majority of modern wastewater treatment plants not only fail to solve this important environmental problem but also contribute to the development of antibiotic resistant systems. This issue needs serious and careful monitoring. Countries that care about the health of the population and about future generations take into account the risks associated with the problem of pharmaceuticals in the environment and strive to solve this problem most effectively; therefore, many trials have been investigated with a combination of different treatment processes.

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Corresponding author
Nadeem Ahmad can be contacted at: er.nadimcivil@gmail.com