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Occurrence and Removal of Persistent Organic Pollutants (POPs)

Siyabonga Aubrey Mhlongo, Linda Lunga Sibali, Kholofelo Clifford Malematja and Peter P. Ndibewu

Abstract

Since the revelation in the detection of the persistent organic pollutants (POPs) in industrial wastewater in the early 1990s, a notable progress has been achieved on the research and different removal applications or methods of this challenge at hand. This book chapter entails a decent understanding on the occurrence, effects, and amputation of POPs in the water sector in advancement of municipal performances of treating industrial wastewaters and environment at large. This current chapter also presents an overview of research associated to the amputation of persistent organic pollutants (POPs) from various water bodies, i.e., river sediments, sewage plants, industrial sludges, and wastewater. Also, discussing the relationships with actual pre-treatment and removal rates. Vital characteristics such as the wastewater matrix, location, sources of POPs, materials and modules, operational parameters and problems are presented with a clear focus on removal of these organic pollutant's different sources (like, textile wastewater). The particular methods to the removal of POPs can be associated with the application of ultrafiltration, nanofiltration and reverse osmosis as advanced treatment stages are considered in correlation with the textile wastewater characteristics and removal efficiencies requirements. This gives significance to the amalgamation of physico-chemical and biological treatment with membrane processes which is likely to represent an efficient solution for the removal of POPs from textile wastewater. However, since membrane fouling and hydrophilicity are apparent in the execution of this process, this chapter also covers the effective strategies like fabrication of membrane with a suitable additive to counterattack these challenges, which are often used in membrane technological research. This chapter also proposes an updated understanding of fouling and improvement of membrane properties.

Keywords: persistent organic pollutants (POPs), ultra-filtration (UF) membranes, blending, fouling, hydrophilicity

1. Introduction

There has been an advanced progress regarding the persistent organic pollutants (POPs) - i.e., elongated-lived, lethal organic composites such as PCBs, PAHs, OCPs and dioxins which have predominantly pursued their way into the environmental sector - constitute the theme of a research programme launched in the early 1990s

by the Swedish Environmental Protection Agency (SEPA). This environmental programme has raised funds estimated to SEK 50 million for the research facilities in Sweden to bring focus only on persistent pollutants [1]. Equally concerning, the data obtained by World Health Organization (WHO) in late 2016, an estimated that 1.2 billion public does not have access to clean water [2, 3]. The occurrence of persistent organic pollutants (POPs) in river water and water treatment plants has raise serious concerns, especially due to the high costs and energy consumption that comes with mitigation of these challenges – because it involves variety of steps, and over thirty processes have been primarily used [4, 5]. The apparency or the occurrence of POPs in industrial wastewaters and textile industries have led to more of ecological negative effects, these includes, i.e., good taste and odor issues of the downstream water supplies, and further forming foam. This results in inhibition of the natural self-purification processes, and worse case - negative effects on the marine life and living organism in the society [6].

Persistent organic pollutants (POPs) remains nothing else but a bunch of different chemical compounds that constitutes of different pedigrees but have common traits, viz., semi-volatility, hydrophobicity, bio-accumulative, high toxicity, and alarming persistency in the environment, and they can also drift into food chains [7]. Research have indicated major contributors of POPs in the environment, these are typically chemical industry, textile industry [8], pulp and paper industry [9], and treatment of landfill leachate [10, 11].

1.1 Textile industries contribution to POPs in waste streams

During the early months of 1990, several studies reported on textile industrial sector being the major contributor of POPs, and worse, discharging very high absorptions of different hazardous POPs, i.e., polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) [12–14]. A bigger portion of polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans from textile industry are sourced all through - washing into sewage sludge, which is often used as an agricultural fertilizer, and are also the source of dioxins in the food chain [15]. A Few of these POPs like polychlorinated biphenyl, phenols, benzenes and dichloro-diphenyl-trichloroethane (DDT) are purposely formed in different of commercial applications for their significant nature or properties they have intermediates or pesticides **Table 1**.

Persistent organic pollutants are toxic chemicals which belong to the families of chemicals such as aliphatic and polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), organochlorines (OCPs), and organophosphorus pesticides (OPs) [17]. They tend to accumulate in the environment and have shown to resist photolytic, chemical, and biological degradation [18]. Persistent organic pollutants have been described by Stockholm convention as a wide range of chemicals which poses a greater risk to human life and biota due to their toxic, persistent and bio-accumulative nature [19]. Their exposure may lead to birth defects, dysfunctional immunity, change on the reproductive and/or nervous systems [20]. Therefore, their continuous detection in the environment even in low concentrations has been a genuine concern for many years. This chapter aims at giving a deep understanding on the occurrence of POPs, their nature and detection methods.

1.2 Occurrence of persistent organic pollutants (POPs)

The industrial application of POPs can be traced back during the early 1900s when these chemicals were commercialized, and used for pest and diseases control [7]. For a number of years, researchers have focused their attention on studying the

POPs	Effects	Processes or methods
Aromatic amines	Cancer-causing effect	Textile industry, dyeing
Dioxazine	Hazardous effect	Textile industry, dyeing
Antraquinone	Oncogenic effect	Textile industry, dyeing
Pentachlorophenol	Carcinogenic effect	Processing of cotton
Chloranil	Carcinogenic effect	Textile industry, dyeing
Phthalocyanine	Oncogenic effect	Textile industry, dyeing
Phenolic compounds	Harmful effects on neuraxins, liver and kidney, cancer-causing effect	Textile industry, dyeing

Table 1. Processes and effects of some persistent organic pollutants from textile industry assembled by Mustereț and Teodosiu [16].

persistence factor, bioaccumulation, and toxicity of the common POPs such as, viz., PCBs (polychlorinated biphenyls, PAHs (polycyclic aromatic hydrocarbons) and OCPs (organochlorine pesticides) [21]. Although many POPs have been prohibited due their adverse effects however, they are still detected in considerable levels in the environment around the globe [22]. Several studies around the research space have reported significantly elevated levels of POPs in various matrices, including biota, sediment, soil, surface water, and drinking water [23–25]. It is no doubt that the rapid increase in human population, urbanization and industrialization have had a great impact in the rapid increase of the POPs in the environment [26]. Moreover, farming practices such as discharge of pesticides and fertilizers into the environment also lead to significant increase of POPs in the environment [27].

1.2.1 Compositional patterns and properties of different POPs

i. Sources of PCBs, their toxicity and nature in the environment

Polychlorinated biphenyls (PCBs) have been identified as a group of chlorinated organic pollutants consisting of 209 isomers and congeners that resulted from the variation in number and position of the chlorine atoms connected to the biphenyl rings [28, 29]. Most of these chemicals which are synthetic, have been used as coolants and lubricants mainly in electrical equipment such as electrical capacitors, generators and transformers owing to their insulating properties [30, 31].

They are characterized as persistent pollutants due to their low water solubility, high fat solubility, resistance to degradation and bioaccumulation in the environment [32]. The major concern associated with PCBs is their high level of toxicity even in extreme low concentrations. Despite their prohibition and also classified as one of the “dirty dozen” in the grouping of POPs, they are still detected in the different environment matrix [33]. Research conducted on monitoring of PCBs in the environment show that sediments are the major sources of PCBs [26]. This is because POPs such as PCBs have high organic carbon partition coefficients (K_{oc}), making them to easily adsorb to sediments. Polycyclic biphenyls are often discharged into the environment from industrial discharge, storage leaks, volatilization, urban discharge [34].

ii. Sources of PAHs, their toxicity and nature in the environment

Polycyclic aromatic hydrocarbons are a group of lipophilic chemicals which exist in the environment in different forms (colorless, white, or yellow solids). These

chemicals exist as a mixture containing two or more benzene rings fused together in linear, cluster, and angular arrangements as shown below in **Figure 1** [35]. They have been listed under Stockholm convention as POPs due to their bio-accumulative and toxic nature in the environment, while they also have been found to exhibit toxic properties such as; carcinogenic, mutagenic and teratogenic making them harmful to human health and aquatic life [36]. Naturally, PAHs can be produced from incomplete combustion of renewable materials (e.g. wild fires) [37], and volcanic eruptions [38]. However, literature shows that anthropogenic activities such as garbage burning, coal combustion, exhaust from motor vehicles, etc. dominate the sources of PAHs in the environment [39]. The persistency of PAHs tends to increase with increasing molecular weight **Figure 2** [40].

iii. Sources of OCPs, their toxicity and nature in the environment

Organochlorine pesticides are chemicals often used in agricultural activities mainly for pest control purposes [41]. They have been listed as POPs owing to their toxic, bioaccumulation and non-biodegradability nature [42]. Improper disposal from domestic use such as indoor residual spraying of pesticides plays a significant role in the increased levels of OCPs in the environment [43]. The increasing demand of agricultural practices and the persistent fight against pests mean more pesticides residues produced **Figure 3**.

According to a study by Jayaraj, Megha [44], only 0.3% of the pesticides used on crops interact with the target pest while the rest becomes excess. Therefore, these chemicals end up in different environment matrix including soil, sediments, and air. Of all the environmental matrix contamination, sediment contamination has reported to have detrimental effect on the source of food chain [45]. Furthermore, literature shows that considerable levels of OCPs have been detected in various honey samples [46–48], which is a proof of the impact that OCPs have on the food chain.

iv. Removal of persistent organic pollutants (POPs) in wastewater

Due to the continuous released of POPs into the environment, this has prompted researchers across the globe to find solutions for treating POPs. Physico-chemical methods such as coagulation, ion exchange, oxidation and adsorption have over many years been applied for removal of wide variety of POPs in the environment [18, 49]. However, many of these methods have been associated with several setbacks such as high cost. Equally important, POPs have been reported to be resistant to physico-chemical methods such as flocculation, coagulation, filtration, and oxidation process [50]. More so, bioremediation has proved to have more advantages over some physico-chemical methods due to its cost effectiveness, wide variety of the microorganisms or bio-sorbents and non-destruction of the material site [51–54]. The *in situ* bioremediation process which involves carrying out treatment

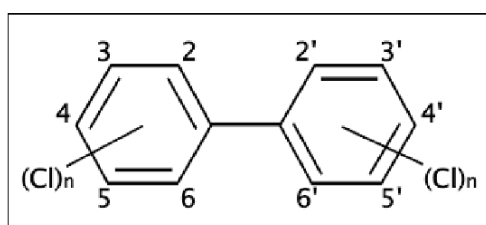


Figure 1.
Structure of PCBs.

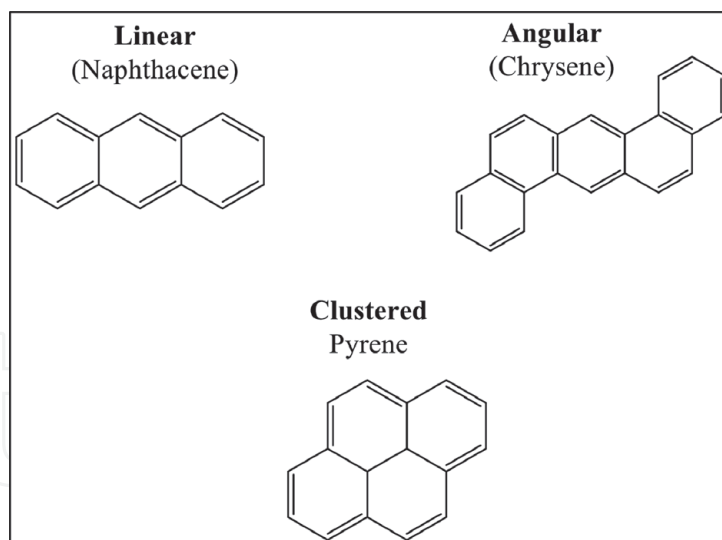


Figure 2.
Different arrangements of PAHs [35].

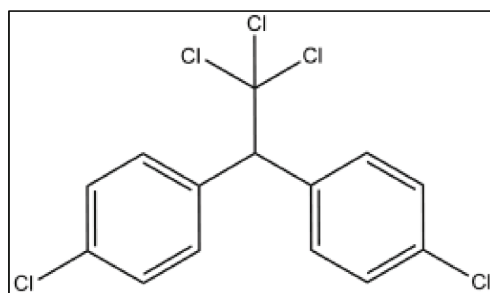


Figure 3.
Chemical structure of OCPs.

process at the contaminated site, has been regarded as a cheap, non-destructive and reliable method for degrading POPs in polluted sites [55].

Advanced oxidation process, defined as oxidation process in which hydroxyl radicals acts as oxidants, has drawn considerable recognition as a potential method for treating POPs in the environment [56]. An advanced oxidation process such as heterogeneous photocatalysis, has been widely used in degradation of POPs in the environment due to its cost effectiveness, wide availability and non-toxic properties [57]. In this heterogeneous photocatalysis, the decomposition and mineralization of contaminants using TiO_2 as photocatalyst is based on the principle of the separation of light-induced electrons/holes (e^-/h^+) pairs [58].

1.3 Removal of persistent organic pollutants (POPs) in wastewater membrane method

Membrane technology have caught so much attention in the research sector due to the drastic growth over a short space of time. This is due to its approach with advantages of using reasonable energy, less chemical matrix, good film forming ability, flexibility, robustness, separation properties, and recently, they can easily integrate with a number of methods [59, 60]. Ultrafiltration (UF) membranes are likely to be the approach having replaced macromolecular separation technique such as proteins – and apart from being the newest approach, UF membranes have some very good attributes like, low energy consumption, mild operating conditions, no phase change and they are environmentally friendly [61]. There are many polymeric materials that have been used before in the membrane processes,

however, poly(ether)sulfone (PES) is mostly preferred material in (UF) membranes because of excellent properties (mechanical, thermal, and chemical stability). Some of the famously highlighted challenge about PES is the factor of hydrophobicity. This shortcoming have led to the announcements of membrane fouling from previously reported studies [62, 63]. The other polymer material previously investigated are cellulose [64], poly(vinylidene fluoride (PVDF) [65, 66], polyetherimide [67, 68], polysulfone (PS) [69] and polyethersulfone (PES) [70]. Nevertheless, PES remains the preferable membrane materials in the synthesis of UF membranes, for decades because of its convenient features.

Now, surface modification of polymeric membranes can be physical, chemical, or said to be bulky modified (i.e., polymer blends) [71]. Any type of membrane modification, be physical or chemical method - after the membrane is formed, it creates a more hydrophilic surface. These vast modification techniques can be classified into three processes, (i) graft polymerization, this is when smaller particles with hydrophilic nature are smoothly distributed or chemically infused onto the membrane scaffold; (ii) physical pre-adsorption of hydrophilic components to the membrane surface plasma treatment, this is slightly different because, there is rather a selected or a change of a functional group to the membrane surface [i.e., sulphonation, carboxylation, etc]; and (iii) Former studies confirms different kinds of modification procedures for the modification of PES membranes, namely, physical methods like blending and surface-coating methods [72, 73], and chemical methods including photo-induced grafting [74], and plasma treatment and plasma-induced grafting [75, 76].

1.3.1 Membrane blending method as an effective technique for the removal of POPs

1.3.1.1 Blending method

Usually, for an improved PES polymer property, blending method should be taken into considerations because of its simplicity and efficiency it has shown over the years. In order to observe a noticeable change in the performance of the membrane, blending method should be a necessity – this is when both PES polymer is mixed together with poly vinyl pyrrolidone (PVP), and thawed in *N*-methyl-2-pyrrolidone (NMP). The resultant polymer resin formed from the mixture should be left to be stable until handled further as normal casting technique [77]. Nearly, the idea of blending is to consortium or improve a material in a hydrophobic nature into a good mechanically hydrophilic material. This is achieved by directly blending a hydrophilic polymer like as PVP [78, 79] and poly (ethylene) glycol (PEG) [80], in that way, PES membranes are easily modified.

In this case, PVP is considered for the formation of micropores, in that way, the hydrophilicity and the antifouling properties of the membrane are increased [80]. Therefore, polymer blending technique gives rise to polymeric membrane with much improved performances and improved properties in reference to the pristine or bare PES membrane. Some researchers have encountered significant shortcomings, based on miscibility of the polymer [81]. In one way or other, there are going to be unexpected challenges with the miscibility which is limited to a narrow concentration range of vinyl pyrrolidone. These challenges are eventually resolved by blending sulfonated PES with the original PES, this is what has been done before [82, 83]. This positively outcomes the higher water permeability, and high rejections in the synthesized membrane – hence the confirmation by the sudden appearance of smaller pore sizes [84, 85]. Hence a clear indication that hydrophilicity can be wide-ranging by changes in the composition ratios of blending.

1.3.2 Considerations affecting the removal of POPs by NF/UF PES membranes

1.3.2.1 The membrane characteristics

Throughout the process of eliminating POPs from the source of waterbodies (or wastewater samples), PES material membranes become a vital factor if you consider the nature of the apparent POPs. This accurate selection of a PES membrane largely plays a role in the removal mechanism since the process is strongly related to the type and functional groups in the membrane chosen. Subsequently, there is also a significant aspect to contemplate in a suitable membrane selection, and that is - the molecular weight cut-off (MWCO), normally articulated in Dalton. This indicates the molecular weight of a hypothetical non-charged solute lying between 85 and 90% rejection, the porosity of a membrane, the surface charge, and the membrane material (polymer composition) as well as the degree of ionic species rejection [86]. In conclusion, the effect of each constraint on the removal of POPs is specifically related to the actual solute properties (molecular weight, molecular size, acid dissociation constant-pKa, and hydrophobicity/hydrophilicity — $\log K_{ow}$), with which this governs the strength of the POPs-membranes physical and chemical interactions.

1.3.2.2 Membrane charge

Usually referred to as zeta potential, membrane surface charge is another vital factor to primarily study in membrane properties. The fundamental principle of the above factor lies in the fabrication of the membrane where you have to consider if the membrane has either a negatively or a positively charged surface [87]. Sometimes a membrane is pre-known to reject negatively charged pollutants (in this case, anions), such as nitrates, sulphates, and sulphites, henceforth, these fictional membranes should be negatively charged for them in order to be effectively repel the pollutants. This phenomenon therefore results into a reduced membrane fouling [88, 89]. This genius analysis of a membrane charge was discovered PVP micro particles were dispersed onto PES membrane for the membrane to give rise into an increased water permeability [90]. Thus, the zeta potential could result to many functional groups, such as, O=S=O, that comes with PES, and O=C-N of PVP that was dispersed across the scaffold of the membrane. The practical functional groups become the primary source of a negatively charged membrane [91, 92]. Consequently, this boldly confirms that an increase in the PVP particles likely to increase the hydrophilicity of the synthesized membrane – which by default leads to high permeability. Hence, the membrane charge increases as the PES and PVP dosages are varied.

1.3.2.3 Persistent organic pollutants (POPs) hydrophobicity or hydrophilicity

Hydrophilicity and hydrophobicity extremely defines the adsorption on the rejection of POPs during membrane applications process [93, 94]. Studies clearly shows that the interface between the non-polar hydrocarbon segments of POPs and the used membrane is primarily the cause of hydrophobic bonding - this has advanced the membrane progress on the extensive adsorption of POPs and of other organic pollutants onto the membrane technology [94–96]. A book published in the early 2000s vividly show that beyond hydrophobic interactions, adsorption could possibly occur over hydrogen bonding between the organic molecules and the hydrophilic groups of the membrane material [97]. Henceforth, hydrogen bonding and hydrophobic interactions may both occur independently

or concurrently. Therefore, according to the studied POPs in this book, the literature approves that the hydrophobic interactions is the driving force for the organic pollutants adsorption on the membrane surfaces - this constitutes the primary step of the rejection mechanism as Nghiem and Schäfer [97] have indicated in his study. In both ways, these observations have implicitly concluded that the rejection of hydrophobic compounds should (by experimentation) be examined after the used membrane is saturated with the target compounds, otherwise, the rejection could be incorrectly mistook for adsorptions are misread [98].

1.3.3 Alternative method - Surface modification

Several studies have been done on surface modification of membranes, and it has shown decent suitability for PES material for the amputation of POPs in wastewater. This includes, self-assembling nanoparticles [99] and/or nanotubes in PES membranes. Nonetheless, this book chapter solely focuses on PES polymer as an adsorbent blended for the improvement of the membrane properties. Another method still to advance in the membrane technology is - surfactant modification. However, a little progress has been observed in literature and still requires more work to be reported on using PES membranes. In the late 2000s, Boussu, Van Baelen [100] showed an increased in the flux for the nanofiltration membrane of waterbodies comprising of surfactants. Lastly, a brief study confirms that hydrogen fluoride could be considered to advance the membrane performances. Fourteen [14] days of immersion in a hydrogen fluoride solution, an increased permeability was obtained without any loss of rejection capacities [101–103].

1.3.4 Factors to consider during the removal of POPs

1.3.4.1 Effect of the feed water composition

A modified membrane performance with real water normally consists of (i) solutions containing salts, (ii) other organic matters, (iii) pesticides, hence, POPs rejection value is likely to vary significantly depending on the feed water composition. Importantly so, pH of the water becomes a prominent influent in the POPs rejection. Below is a brief discussion of how pH is an important parameter as the driving force in rejection values.

Influence of water pH: pH in the rejection role is vitally imperative in these experiments – as it directly involved in the membrane surface and membrane charge because of the dissociation phenomenon of functional groups throughout the adsorption of POPs. Different researchers have found membrane charge (zeta potential) suddenly leaning more to negative charge whilst the pH of the water body is increased, thus, resulting in functional group deprotonation [104–106]. Moreover, another prominent researcher, Freger, Arnot [107] verified about the varying of the pore sizes likely to take place reliant on the electrostatic interactions amongst the dissociated functional groups within the membrane material. Pang, Gao [108], also showed a study where high pH ranges seemed to cause reduced rejection rates, with permeate flux also going up. And this this was ascribed by the increased number of pore sizes at high pH values. These tests were conducted during the removal of one of the top four POPs (PCBs, OCPs and DDT, etc), and expectedly, the outcome exhibited that membrane rejection achieved the highest value at pH 7, and repeatedly gave lower rejection values at pH 2.5 [109]. This clearly indicates, the ion adsorption on the membrane scaffold, and predominantly at higher pH, OH⁻ ion adsorption is increased - which automatically leads to an increase in the zeta potential of the membrane.

1.3.4.2 Effect of membrane fouling

Fouling of a membrane is apparent and continue to be a challenge within the membrane technology scope and industrial applications – this includes the wastewater treatment processes [86, 110]. This takes place as the undesirable particles accumulate to cause clogs in the water flow across the membrane. This results in the shortening of membrane life. Membranes looks at advancing the progress by creating membranes with better or improved properties – this means fabrication or modification to create low fouling propensity. The achievement relies on transformation of hydrophobic polymers into hydrophilic nature [111, 112]. However, it is of emergency that cost-efficiency efforts be applied in order to mitigate membrane fouling as much as possible. For this to be counter-attacked effectively, mechanisms of membrane fouling should be studied expansively, hence, to develop dynamic anti-fouling methodologies.

2. Conclusions

The contamination regulator of persistent organic pollutants (POPs) due to industrial and textile discharged effluents has become more severe and, clearly demands for interventions of more efficient wastewater advanced treatment. This leads to a combination of physico-chemical and biological treatment using membrane methods – which in-fact, embodies an efficient solution for the removal of POPs from these industrial and textile wastewaters.

In conclusion, application of membrane methods could successfully rely on several factors for its optimum use, i.e., material composition membrane selection, type of modules, wastewater characteristics and the interactions between contaminants (POPs) and the synthesized membrane. Membrane procedures potential use, for the removal of significance organic pollutants in industrial water bodies and from textile effluents are ultrafiltration (UF), reverse osmosis (RO) and nanofiltration (NF). These employed methods are cost-effective and easier to carry out. However, for fiscal and monetary reasons, these applications remain a disadvantage in the case where the effluents or wastewaters can be recuperated for re-use.

Application of PES polymeric membranes for these procedures or the removal of POPs contains increased removal rates, and the choice of a membrane material becomes paramount important considering properties of PES like, permeability, selectivity, chemical and mechanical resistance. But PES also have some integral operational challenges, such as: fouling and concentration/polarization phenomena. This further leads to an unexpected decrease of the permeate flux, and the vital aspect in the operational procedure of PES membranes and the performance of the membrane inevitably decrease. However, washing of membrane by the use of physical and chemical procedures could discreetly recover the permeate flux between the membrane processes cycles, yet fundamentally irreversible fouling could possibly emerge.

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Conflict of interest

The authors declare no conflict of interest.

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