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Emerging organic contaminants in drinking water sources: A review of types and sources, occurrence, health risks, and remediation

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ABSTRACT
A number of chemical contaminants are classified as emerging organic contaminants (EOCs) because they are either new or recently detected. They collectively encompass micropollutants such as per- and poly-fluoroalkyl substances (PFAS), plasticizers, solvents, pharmaceutically active compounds, endocrine-disrupting compounds, gasoline additives, personal care products, food colourants, artificial sweeteners, musks, and fragrances. Multiple pathways has investigated through which EOCs can invade the drinking water sources. Global occurrence of EOCs is discussed in the drinking water sources. Countries like China and USA carried out maximum studies to detect the presence of a wide range of EOCs. Due to their potential risks to public health, EOCs have garnered significant global attention and research efforts in both developed and developing nations. These EOCs, along with other organics, lead to the formation of carcinogenic disinfection byproducts (DBPs) in community drinking water treatment plants. Researchers have explored various remedial measures ranging from in-situ treatments to ex-situ treatments. Despite the recent progress, it still need for proper field-scale experiments before being successfully used in community treatment plants to provide clean water to the masses to achieve sustainable development goal (SDG).

Keywords: Emerging organic contaminants, Drinking water, Health risk, Remediation, Sustainable development goal.

Introduction

The population on earth is ever-growing, and the majority of the earth's usable freshwater is extensively used to provide for the same in agricultural, industrial, and domestic sectors [1]. However, human activities like the overuse of pesticides and fertilizers for irrigation, improper disposal of solid wastes, leakage of septic tanks, or dumping of industrial effluents into the water bodies have led to immense damage to our surface and groundwater resources [2,3]. These activities lead to the introduction of emerging organic compounds (EOCs) in the aquatic environment. EOCs are organic pollutants that are newly detected or discovered in the environment due to recent analytical developments.

They include a divergent group of chemicals, their metabolites, and their transformation products, which set foot, disseminate and persist in the environment [4,5]. These organic contaminants bioaccumulate in the aquatic environment and exert toxic effects on aquatic animals, which can easily get transferred to humans via the food chain [6]. The surface water is well known to be susceptible to contamination and is backed by various research works. The occurrence and concentration of

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EOCs in the surface water are much more studied and reported than groundwater because of the complexity of the subsurface [7,8]. It has been reported worldwide that the conventional wastewater treatment plants, are not capable of complete degradation of certain EOCs [9]. Hence, these compounds pollute the downstream surface water. Surface water can also get easily polluted by agricultural runoffs and animal farms.

On the other hand, it is often assumed that the groundwater is devoid of anthropogenic pollution. Most of our liquid freshwater is groundwater, and around 2.5 billion population depend on it for basic consumption needs without any hesitation. Groundwater contamination should be of a more significant concern than surface water because they are the only perennial water source [10]. The occurrence and concentration of contaminants in the subsurface depend on various aspects like the aquifer material, the organic carbon content of the soil, category of impurity present, molecular weight of the contaminant, and the depth at which it is measured, which adds to the complexity [11]. Moreover, the groundwater and the surface water interact with each other, so contamination of one will affect the other [12]. Many regions face scarcity and contamination of the groundwater, but the number of research work is comparatively less [13]. However, our present concern about EOCs is not only related to the ecosystem but also regarding the degradation of the drinking water sources, which can have detrimental effects on our health in the long run.

The United Nations identifies access to safe and clean water as critical for improving the health, well-being, and socioeconomic situation in developing countries. The human population is dependent on surface and groundwater for their daily activities. Water from various surface and groundwater sources reaches the community drinking water plant. They undergo various treatments to ensure clean water distribution to the consumers to achieve SDG 6 [14].

This review work integrates present knowledge on the EOCs in community drinking water sources ranging from its origin, occurrences, eco-toxicity, health impacts, and remedial measures on a global level.

Sources of EOCs in surface water and groundwater

It is the prime concern to ascertain the predominant sources that contribute to groundwater contamination to various degrees so that proper steps can be initiated to reduce further contamination. Fig. 1 indicates multiple pathways through which EOCs can invade the drinking water sources [15].

Point source

In the case of point sources, pollutants are generated from distinct sources whose intrusion and transference into the environment is in a spatially constricted pattern and can be mathematically modeled. Effluents from industries and municipal treatment plants, dumping grounds, and leakage from septic tanks are some of the potential point sources of subsurface and surface water contamination. If the septic tanks are considered for individual households, they may become point sources for the contaminant leakage, however, an urban sprawl with multiple leakages may be considered as a non-point source.

Non-point sources

The origin of the non-point or diffuse pollution sources is primarily unknown and hard to detect. Examples of non-point sources can be agricultural or stormwater runoffs, which usually carry harmful EOCs from the land to water bodies gradually to groundwater.

Occurrence of EOCs in surface and groundwater

EOCs pave their way into the environment through various sources. Researchers have carried out several studies or experiments to demonstrate the existence of EOCs in drinking water sources. Countries like China and USA carried out maximum studies to detect the presence of a wide range of EOCs. Fig. 2 shows the global occurrence of EOCs in the drinking water sources [15]. Tables 1 and 2 summarize the global occurrence of EOCs in the groundwater and surface water, respectively.

Analysis of global data on pharmaceutical pollution in rivers showed that the cumulative concentrations ($\mu g\ L^{-1}$) of pharmaceuticals (mean ± standard error of the mean) were statistically more than two times significantly higher ($p = 0.011$) in South America (11.68 ± 5.29), Asia (9.97 ± 3.33), Africa (9.48 ± 2.81) and East Europe (8.09 ± 4.35) than in high-income regions (2.58 ± 0.48). Maximum cumulative concentrations of pharmaceuticals (μg L-1) decreased in the order; Asia (70.7) had the highest value followed by South America (68.8), Africa (51.3), East Europe (32.0) and high-income regions (17.1) had the least concentration. The corresponding human intake via ingestion of untreated river water was also significantly higher in low-and middle-income regions than in their high-income counterparts. For each region, the daily intake of pharmaceuticals was highest in infants, followed by children and then adults [16].

Eco-toxicological assessment and impacts of EOCs in the aquatic environment

There has been an increase in EOCs in the aquatic environment over the past few decades. Hence, eco-toxicological assessment is required to control and manage the drinking water resources properly. Eco-toxicological assessment involves laboratory analyses of living organisms where they are exposed to a certain contaminant at various concentrations for an established period. The use of biomarkers to indicate exposure to various contaminants provided reliable results.

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It can be effectively used as a warning signal for proper monitoring and eco-toxicological risk assessment of contaminants. Most EOCs pose acute and chronic toxic effects of EOCs on model animals and humans (Table 1). Here, an overview of the human health risks of various EOCs is presented (Table 1).

Although EOCs are generally present in low concentrations, their interaction with other compounds in the aquatic environment leads to synergistic mixtures. The synergistic combinations can be more toxic than the parent compound causing detrimental effects on human health and the aquatic than the parent compound causing damaging effects on human health and the aquatic biota. The photochemical transformations of the water contaminants can also result in toxic byproducts, which add further complexity to the process of risk assessment. EOCs in a water source can also enter the water treatment plants and generate new EOCs (disinfection byproducts, DBPs) with higher toxicity than the original compound. For example, the DBPs of gemfibrozil, such as chlorogemfibrozil and bromogemfibrozil, induced higher toxicity on male Japanese medaka than its parent compound (gemfibrozil). Various health impacts associated with EOCs on humans were depicted in Fig. 3. EOCs remain biologically active in the environment for a long time, affecting non-target living organisms. Asian vultures are on the verge of extension due to diclofenac exposure is an example where EOCs like pharmaceuticals have resulted in a population-level impact on non-target living organisms. The use of pesticides is widespread in developed as well as developing countries. In North-Western Europe,Japan, and the United States, pesticide consumption is three-fourths of total world pesticide usage. Pesticides are usually non-reactive; they either get dissolved or remain suspended in water and persist in nature for years.

Remediation of EOCs from drinking water sources

As discussed earlier, EOCs in water can cause immense damage to the overall health of the people consuming it. It also affects the aquatic flora and fauna, disrupting the environmental balance. Therefore, it is necessary to remediate the pollutants by different physical, chemical, or biological means. Several remedial techniques are available for water treatment, as listed in Table 4. Still, the main aim is to find a method that would be technically and economically feasible for a particular circumstance and region. Presently, researchers are investigating new remedial techniques by integrating various successful strategies for the economical and effective removal of emerging contaminants in natural waters.

Remediation of groundwater

The remedial technologies used for remediation of groundwater work on the principles of extraction/removal (pump and treat, air stripping) or transformation/degradation (in situ chemical oxidation, biodegradation), as represented in Fig. 4 [17]. Remediation of EOCs can be achieved through ex-situ and in situ techniques. Factors that affect which type of remediation technique will work depend on hydrogeological conditions, the aquifer's geochemical conditions, and the contaminant type. For example, karst aquifers consist of a particular kind of hydrogeological condition due to the presence of a highly permeable conduit, so it becomes highly challenging to remediate it [15].

In-situ chemical oxidation (ISCO)

In-situ chemical oxidation is the most frequently used system for the degradation of EOCs in groundwater to date. In-situ chemical oxidation (ISCO) is a very effective method of remediating groundwater polluted by stubborn Non-aqueous phase liquid (NAPL). It involves injecting suitable chemical oxidants in the subsurface, which can degrade a wide range of stubborn EOCs [18]. Fig. 5 represents the schematic diagram of the ISCO system. Oxidants usually employed in ISCO are persulfate, catalyzed hydrogen peroxide, and permanganate. Activating agents like heat, surfactants, alkali, metals, and glucose have successfully activated the oxidant [19]. ISCO can be very effective in remediating groundwater contaminated by phenolic compounds and halogenated organics. Another study by Dou et al. [20] examined the in-situ remediation method on four types of β-lactam antibiotics. The synergistic effect of persulfate and g-C3N4 (graphitic carbon nitride) resulted in excellent degradation of the pollutants. ISCO involving ferrous ion-activated decomposition of persulfate was used to remediate groundwater contaminated by ciprofloxacin and sulfamethoxazole. Better degradation of ciprofloxacin was observed under the same treatment condition. Hence, different contaminants respond differently to identical hydrogeological parameters and remediation techniques. Hybrid processes involving ISCO followed by in-situ bioremediation have also gained popularity due to their promising results. Although the oxidants initially affect the microbial population by changing the pH and redox conditions. Eventually, it increases the bioavailability and biodegradability of the contaminants, which supports the biodegradation process.

The oxidants can be directed to oxidize fewer biodegradable parts of the contaminant, which can aid in the production of more bioavailable byproducts. The ISCO involves the formation of various reactive intermediates in the subsurface that can be difficult to control and predict which can be disadvantageous when applied in the field, which emphasizes the requirement of more field-based experiments for proper insight [15,18-20].

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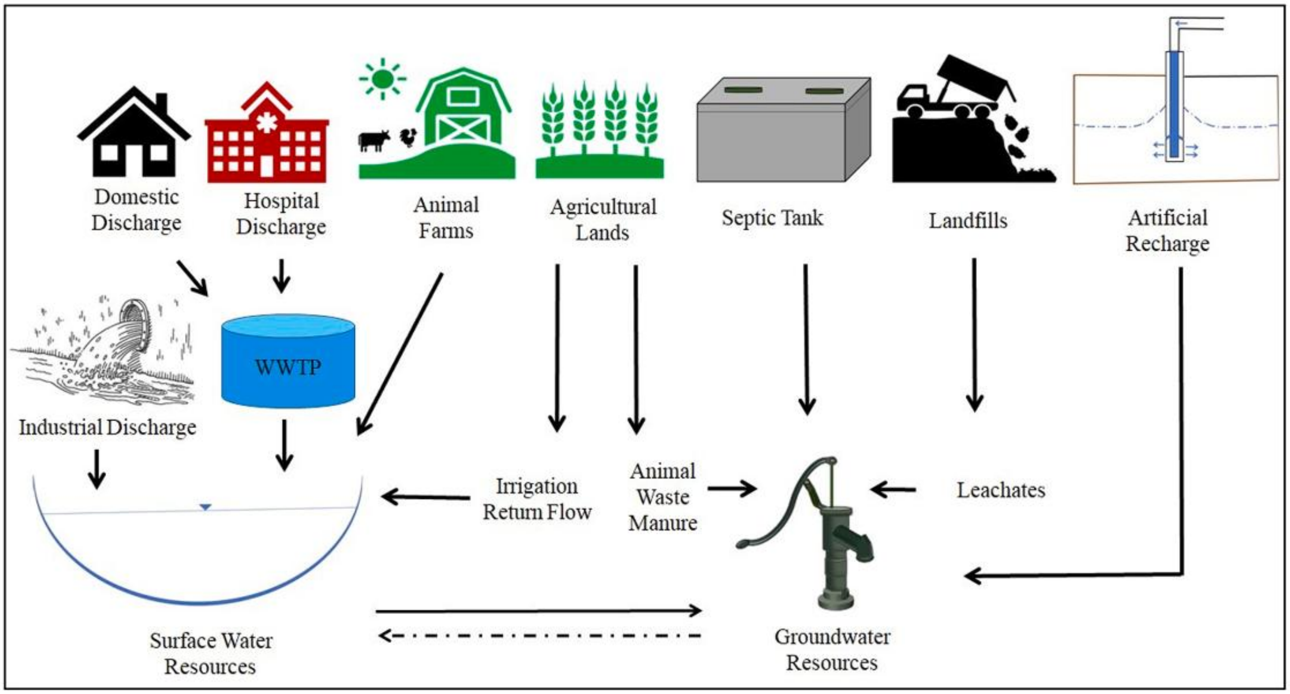

Fig. 1. Schematic diagram stating potential sources and pathways for pollution of drinking water sources by EOCs [15].

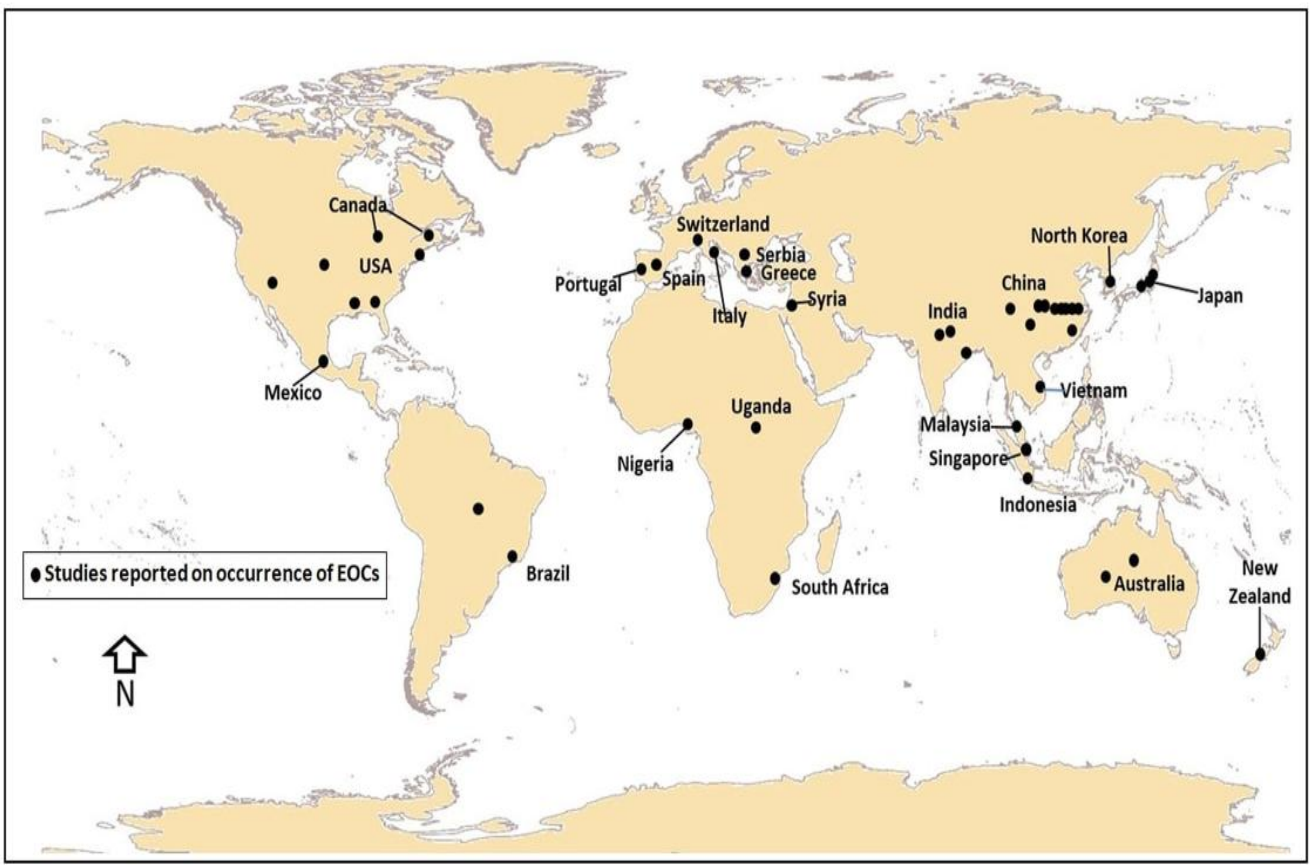

Fig. 2. The global occurrences of EOCs in drinking water sources [15].

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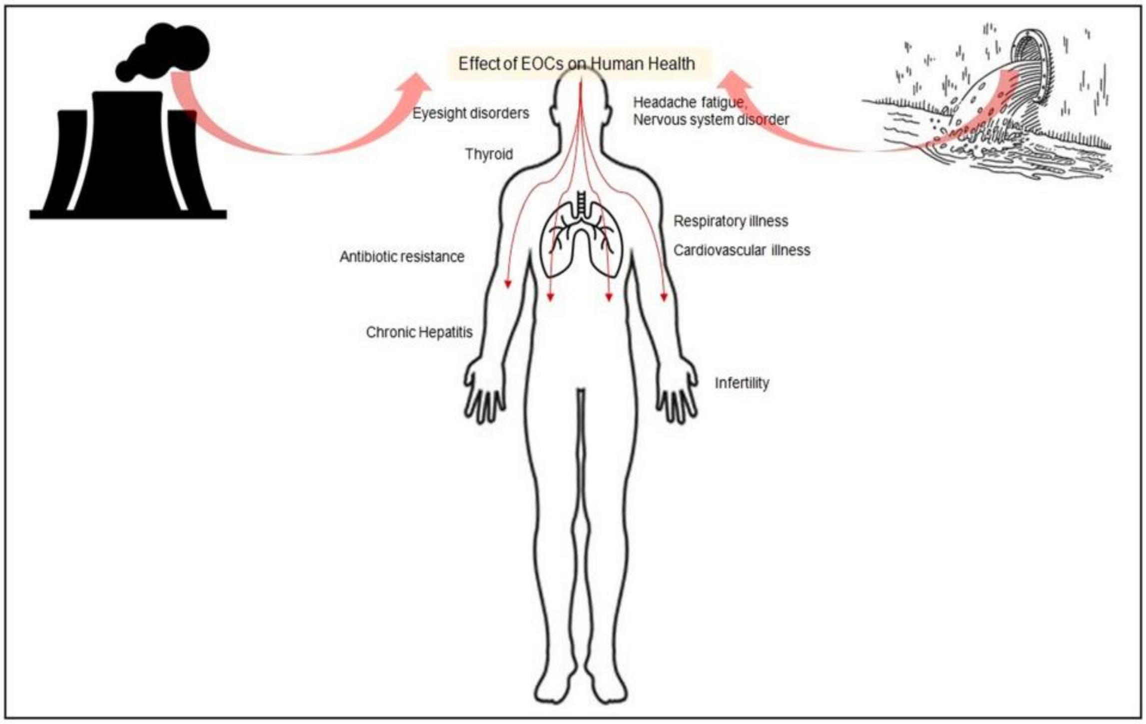

Fig. 3. *Effects of EOCs on human health [15]*

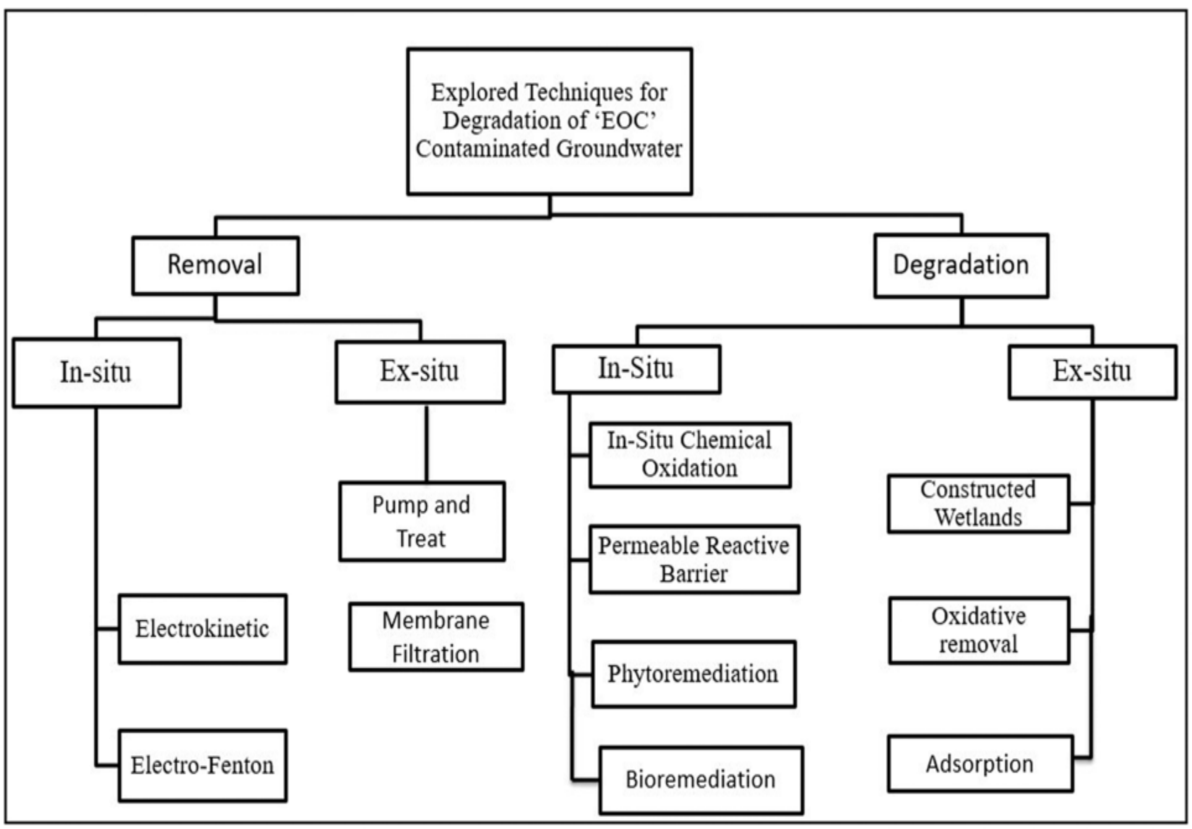

Fig. 4. Various techniques for treatment of 'EOC' contaminated drinking water sources [17].

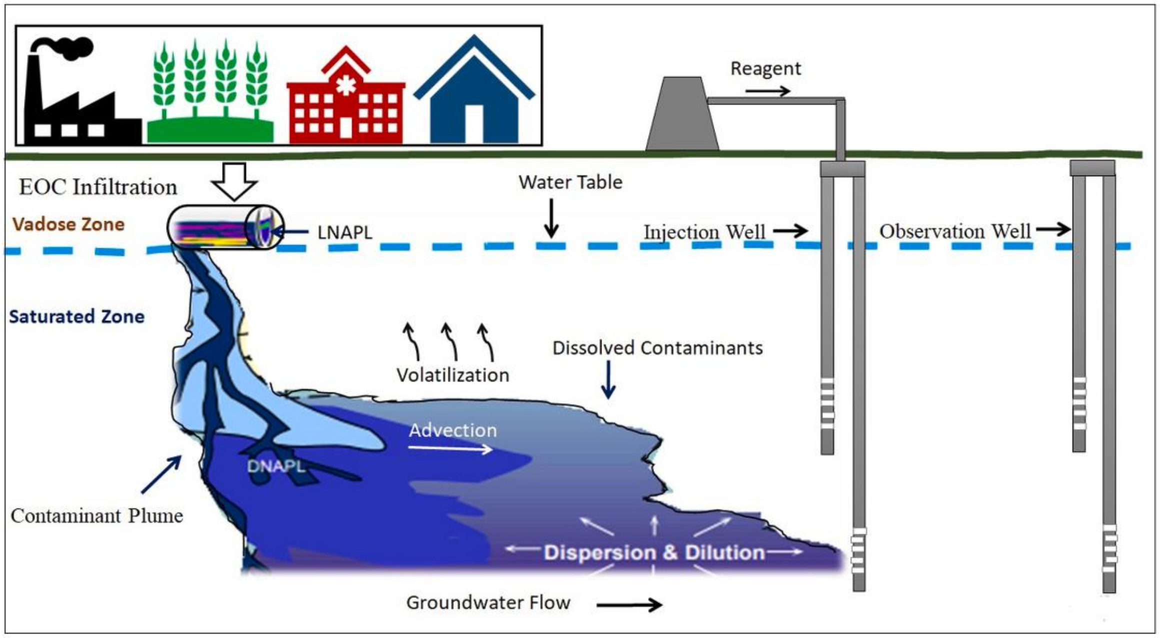

Fig. 5. Schematic diagram of the ISCO system [15].

Table 1- Types of emerging organic compounds, concentration values found in drinking water sources and the effects of each of them on community health [16].

Type of EOC	Concentrations	Major Findings and
1. Pharmaceuticals		
Sulfonamides	20.06 μg L^{-1}- 1281.50 μg L^{-1}	Human health risk for age groups less than 12 months was reported.
Chloramphenicol, ciprofloxacin, sulfapyridine,	0.5–21.4 ng L^{-1}	Resistance election.
Doxycycline, enrofloxacin, florfenicol, levofloxacin,		
metronidazole, norfloxacin,		
Estrogen and sulfapyridine	0.09–304 ng L^{-1}	Resistance selection.
Ciprofloxacin		Ciprofloxacin resistance.
Bromate, perchlorate and chlorate		A potential cancer risk.
fluoroquinolones, β-lactams, macrolides, phenicols, sulfonamides, tetracyclines	0.0064 ng mL^{-1} to 0.0089 ng mL^{-1}	Accumulations in the body and detected in the urine.
30 common antibiotics	226.8–498.1 ng L^{-1}	The non-carcinogenic risk of different groups of people was reported.
2. Personal care products		
Cyclic siloxanes D4(octamethylcyclotetrasiloxane) and D5 (decamethylcyclopentasiloxane) (personal care products)		Affects the liver and also induces liver cell enzymes. The primary target organ for D5 exposure by inhalation is the lung

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Nitro Musk (personal care products)	5–10 mmol L^{-1} in surrounding media	Competitive binding capability to estrogen receptors.
Fragrances (personal care products)		Facial dermatitis.
Glycol ether (personal care		Motile sperm count and infertility.
Glycol ether metabolites		Negative impact on the neurodevelopment of
Insect repellents (personal care products)		Cause vomiting.
Butylparaben & propylparaben (Personal care products) Parabens		Adverse birth outcomes decreased gestational age birth weight & body length of neonates.
Methylparaben & propylparaben (personal care products) Parabens		Increases gestational age.
3. Plasticizers	13.42–265.48 ng L^{-1}	Potential cancer risk.
Polyethylene terephthalate bottles	0.046 and 0.71 μg L^{-1}	Carcinogenic risk of 2.8 × 10^{-7}.
Plasticizers		Bio accumulation in the blood plasma and urine
4. Surfactants		
Linear alkylbenzene sulfonates		Acute contact dermatitis.
5. Flame retardants		
OPFRs	85.1 ng L^{-1}-325 ng L^{-1}	OPFRs in drinking water did not pose a major health concern for adults and children
Polybrominated diphenyl ethers (PBDEs)		Rapidly bioaccumulating doubling every two to five years.
Organophosphate flame retardants (OPFRs)	median: 48.7 ng L^{-1}	The estimated daily OPFRs intake was less than the oral reference dose values.
Triphenyl phosphate, tris(1,3-dichloropropyl) phosphate	1.1 ng mL^{-1} and 1.2 ng mL^{-1}	Endocrine-disrupting, reproductive toxins, carcinogens, neurotoxins Flame retardants were detected in all urine samples, suggesting bioaccumulation bio-accumulation.
Organophosphate esters (OPEs)	13.42–265.48 ng L^{-1}	Potential cancer risk (>1.00E-6), no non-carcinogenic effects (<1). Total OPEs in advanced DWTPs were much lower than those in conventional DWTPs. Relative to advanced DWTPs removed approx. 65.6% and 36.5% of the carcinogenic risk and non-carcinogenic risk of OPEs, respectively.
Flame retardants (FRs)	Up to 71.05 ng L^{-1}	The estimated daily intake (EDI) for all FRs was higher in children than adults. Both children and adults were at low risk (HQ < 1)
Organophosphate flame retardants (OPFRs)		EDI through drinking water was ≤9.65 ng kg^{-1} body weight day^{-1}.

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6 Endocrine-disrupting chemicals		
BPA	<LOD to 65.3 µg L^{-1}	Unexplained infertility was present in 29% of the subjects decrease in peak oestradiol
7. Gasoline additives		
MTBE	8 µM MTBE	Insulin formed a molten globule-like structure. This shows MTBE promotes protein oxidation and the production of reactive oxygen species
8. Per- and Polyfluoroalkyl Substances (PFAS)		
Perfluorooctanoate (PFOA) and perfluorooctane sulfonate (PFOS)	concentrations of 40–50 ng L^{-1}	No significant alterations in its activity were observed.
9. Artificial food sweeteners Artificial sweeteners	0.5–25 ng L^{-1}	ASWs in groundwater did not appear to pose human a health risk.
Artificial sweeteners	Up to 21 µg L^{-1}	Bio-accumulation detected in human waste.
10. Ultraviolet sun screening agents or UV blockers/filters		
Benzophenone-3 (BP-3)		The decline in male birth weight and decreased gestational age in humans. A decrease in epididymal sperm density and a prolonged oestrous cycle for females were observed in rats. DNA damage in epithelial cells.
Benzophenone-3	1 µM–5µM	
UV-B filter octylmethoxycinnamate (OMC)	BP-3 1, 10, and 50 µmol L^{-1}	A decrease in vasorelaxation of human umbilical arteries was revealed. Possibility of OMC acting as an inductor of pregnancy hypertensive disorders. inductor of pregnancy hypertensive disorders.
2-hydroxy-4-methoxybenzophenone; 4-		mRNA expression of cytokines up-regulated. UV
dene camphor; 2-ethylhexyl 4-methoxycinnamate; and butyl-methoxydibenzoylmethane		the immune system functions of humans and may lead to the development of asthma or allergic diseases.

Conclusion

number of chemical contaminants are classified as emerging organic contaminants (EOCs) because they are either new or recently detected. Due to their potential risks to public health, EOCs have garnered significant global attention and research efforts in both developed and developing nations. An analysis of global data on pharmaceutical pollution in rivers showed that South America, Asia, Africa, and East Europe had more than two times higher concentrations of cumulative Pharmaceuticals than high-income regions. Groundwater are typically major sources of the community drinking water plants. EOCs can get introduced in the community drinking water sources via numerous point and non-point sources.

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However, conventional drinking water treatment plants fail to remove EOCs from the sources. These EOCs, along with other organics, lead to the formation of carcinogenic disinfection byproducts (DBPs) in community drinking water treatment plants. Researchers have explored various remedial measures ranging from in-situ treatments to ex-situ treatments. Despite the recent progress, it still need for proper field-scale experiments before being successfully used in community treatment plants to provide clean water to the masses to achieve sustainable development goal (SDG).

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