The Potential Influence of Forests and Climate Change on the Environmental Fates of Organic Compounds in Tropical Watersheds

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Authors’ contributions

This work was carried out in collaboration between both authors. Author KSCB reviewed and synthesized existing literature and wrote the first draft of the manuscript. Author FMJ organized and edited the manuscript. Both authors read and approved the final manuscript.

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ABSTRACT

This exploratory review synthesizes current knowledge on the use, exposures and environmental fates of PBDEs, dioxins and dioxin-like chemicals, and current-use pesticides, as well as purports the potential for forests and climate change to affect their fates at the catchment scale in tropical regions. Organic compounds such as these are under global scrutiny because of their widespread distribution and potential for adverse health impacts. PBDEs and current-use pesticides are produced for their beneficial services as flame retardants and pest controls, respectively, whereas dioxins and furans are the by-products of combustion events involving many organic compounds. However, these chemicals distribute to various environmental media and are associated with adverse health effects, including neurotoxicity and carcinogenicity. Previous studies have shown that temperate and boreal forests influence the environmental fates of some organic chemicals by transferring them from the atmosphere to the soils, therefore potentially reducing atmospheric exposure. Changing climate variables, such as temperature change, are also expected to be important in the environmental distribution of organic contaminants. However, the effect of these factors, forests and climate change, on the environmental fates of organic pollutants in tropical

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watersheds have not been adequately examined. Knowledge of the fates and impacts of organic pollutants in tropical regions is critical for environmental management and policy development therein, and this review therefore explores the potential influences of forests and climate change on contaminant fates in tropical regions. Finally, the areas where more research is needed to assist in these endeavors are elucidated.

**Keywords:** Climate change; environmental fates; forests; organic pollutants; tropical watersheds.

1. **INTRODUCTION**

Many organic compounds used have caused tremendous concern, due to their global distribution, as well as potential for adverse environmental and health effects [1] and require monitoring and management. Among these are polybrominated diphenyl ethers (PBDEs), dioxins and dioxin-like compounds, such as furans, as well as pesticides. They are among the ubiquitous environmental pollutants [2].

The pollutants differ by their sources and release media [3] and display varying environmental distribution behaviors, patterns and fates. The differences are controlled by the intrinsic physicochemical properties of the chemical [4]. Such properties include partition coefficients, media-specific half-lives, aqueous solubility and vapor pressure [5]. These intrinsic properties dictate the persistence, long range transport potential, bioaccumulation and environmental state, among others. Contaminants that display unfavorable attributes – persistence, bioaccumulation potential, long range transport potential and toxicity – are recommended for regulation, management or bans [6]. In fact, the possession of these four attributes is a criterion applied by the United Nations Environment Programme for the regulation of persistent organic pollutants (POPs) [7]. For example, PBDEs, dioxins and furans display high octanol-air partition coefficients and long environmental half-lives and, consequently, they bioaccumulate, bio-magnify, and persist in the environment. They are also toxic. Hence, they are classified as POPs [5].

Properties relating to the environment or landscape govern the environmental fates and behavior of organic compounds. Precipitation provides a key mechanism via wet deposition for the transfer of chemicals from the atmosphere to terrestrial and aquatic compartments [8,9]. Also, processes such as volatilization and evaporation, which increase atmospheric concentrations, are often positively influenced by ambient temperatures. Vegetative covers, such as forests, have been shown to influence the environmental distribution of some organic compounds, whether by enhanced uptake from the atmosphere and subsequent transfer to soils or via re-volatilization from leaf surfaces [10,11]. Therefore, climatic conditions and vegetation influence the fates of organics [12].

Once in the environment, these organic compounds may cause adverse health effects ranging from skin disorders to neurological dysfunction, endocrine disruption and immuno-toxicity [2,13-16]. Therefore, understanding all the factors and mechanisms governing the environmental fates of these organics, as well as their impacts is critical for environmental management and the development of appropriate policies. Fig. 1 provides an illustration of the information necessary for solutions to reduce the negative impacts of organic pollutants.

In this exploratory review, one of the main objectives is to synthesize current information on the uses, exposures, health effects and environmental fates of select organic contaminants (PBDEs, dioxins, furans and current-use pesticides), and these are therefore first provided. Subsequently, information is given on that which is known of the influence of select environmental factors, such as climate change, as well as temperate and boreal forests, on the distribution of the pollutants. However, for tropical countries, there are limited studies evaluating the effects of climate change and forests on organic pollutant fates. Therefore, to meet another objective of this review, we next explore the potential for tropical forests and climate change to affect the environmental fates of the chemicals in tropical watersheds. Finally, we highlight the research gaps, which, once addressed, will provide the necessary guide towards the environmental and public health management of these pollutants, to mitigate their impacts, especially in tropical regions.
2. USES, EXPOSURES AND HEALTH EFFECTS of PBDEs, PCDDs and PCDFs

Polybrominated diphenyl ethers (PBDEs) are semi-volatile organic compounds and are among the cheapest flame retardants, whose purpose in items or materials is to interfere with combustion [17,18]. There are 209 congeners of PBDEs, with 1 to 10 bromine atoms possibly attached to the diphenyl ether molecule. Commercial mixtures of the PBDEs include pentabrominated BDE (pentaBDE), octabrominated BDE (octaBDE) and decabrominated BDE (decaBDE) [19]. PentaBDE and octaBDE have been banned in select states in the US and in Europe [19]. The main constituents of the pentaBDE formulation are PBDE-47, 99, 100, 153 and 154, whereas PBDE-153, 154, 183, 196, 197, 203, 207 and 208 are the components of the octaBDE mixture [20]. Commercial decaBDE is the most globally used and its main constituent congener is PBDE-209 (also called decaBDE). The extensive use of PBDEs in recent years has led to global distribution and, as such, they are now ubiquitous [21,22]. These organic compounds are not only persistent, but they also bio-accumulate, bio-magnify and cause adverse human and ecosystem health. A number of manufacturers in the US had committed to gradually stop the use and production of decaBDE [23].

OctaBDE and decaBDE are mainly used in electronic housings, whereas foams and textile materials typically contain pentaBDE [22]. As a result, PBDEs are found in furniture, clothing, our homes and other indoor environments, vehicles and electronic devices. PBDEs are dissolved in the polymers of the material, and the lack of chemical bonding means that PBDEs are constantly being emitted, from the material to which they have been added, during use and disposal. Point sources of PBDEs include manufacturing, recycling and waste disposal facilities. Other sources include back-yard burning [22].

Polychlorinated dibenzo-p-dioxins (PCDDs) and dibenzo-furans (PCDFs) are semi-volatile organic compounds, that are the unintentional
thyroid hormone disruption and developmental impairments [2,24]. They are also impurities in chlorinated products/materials. Natural events, such as volcanic eruptions and forest burning, are among the point sources of PCDDs and PCDFs. There are 75 PCDDs and 135 PCDFs, of which only seven (7) PCDDs and ten (10) PCDFs are of interest, because they have been found to be toxic [25,26]. These 17 dioxins and furans have chlorine atoms on the second, third, seventh and/or eighth positions on the dibenzo-p-dioxin or dibenzofuran parent molecule, respectively. PCDDs and PCDFs adsorb onto particulate matter and are soluble in octanol and lipids, with their solubility positively correlated with their chlorine-atom content [27]. PCDDs and PCDFs are subject to long range atmospheric transport and this, in combination with them being by-products of common events, has resulted in global distribution and ubiquitousness [28].

Atmospheric and aqueous concentrations appear to be high for the pentaBDE, whereas higher concentrations of decaBDE are often found in soils and sediments. PBDEs in the atmosphere and sediment have been extensively assessed, but less is known about concentrations in the soil and aquatic media, although soil appears to be a major sink [22]. In the environment, PBDEs degrade to form lower congeners. Humans are not only exposed to PBDEs via the abiotic environment, but also through diet [29], with fish being the major dietary contributor [30] – this is because PBDEs are lipophilic. Although decaBDE is not well absorbed and is quickly eliminated from animals [31], the lower PBDE congeners bioaccumulate in lipids and bioconcentrate, such that decaBDE has teratogenic potential [16,20].

PCDDs and PCDFs are among the most toxic organic chemicals, achieving status amongst the United Nations Environment Programme’s (UNEP’s) ‘Dirty Dozen’, which are the twelve most toxic POPs initially identified by the UNEP [24]. The most toxic is 2,3,7,8-tetrachlorodibenzofuran (TCDD). Specifically, the toxicities of 17 PCDDs and PCDFs are compared to that of TCDD, via the World Health Organization’s (WHO’s) 2005 toxicity equivalence factors (TEFs), using half-order increments on a logarithmic scale (0.03, 0.1, 0.3, etc.) [32]. The TEFs are used to calculate toxicity equivalence quotients (TEQs), which are compared to a tolerable daily intake of 1-4 pg TEQ/kg-bw. PCDDs and PCDFs have been associated with a variety of effects on humans and animals, not limited to skin lesions, disrupted liver function, disrupted metabolism, behavioral and developmental disorders, immunotoxicity and neurotoxicity. TCDD has also been found to be teratogenic and carcinogenic [2,13].

3. USES, EXPOSURES AND HEALTH EFFECTS OF CURRENT-USE PESTICIDES

The negative health and environmental effects associated with many currently used organic pesticides have placed pesticides under much scrutiny. Major classes include organochlorine insecticides, organophosphate insecticides, pyrethroids, fungicides, rodenticides, carbamates and herbicides [33]. Organochlorine (halogenated) insecticides are considered to be POPs [34], and many such pesticides are banned from use in a number of developed countries. There is instead a global shift towards non-halogenated pesticides, including organophosphate insecticides and herbicides [35]. These increasingly popular non-halogenated pesticides will herein be referred to as current-use pesticides (CUPs). In this review, the focus will be on select categories of current-use pesticides: pyrethroids; carbamates; organophosphate insecticides and herbicides.

Pyrethroids are neuro-toxic insecticides, affecting the sodium and chloride channels of the nerves, and are also associated with respiratory, dermal and gastrointestinal disorders [36]. Organophosphate pesticides are also neuro-toxic insecticides. However, they inhibit the cholinesterase enzyme, resulting in elevated neurotoxicity, and decaBDE has carcinogenic potential [16,20].
levels of acetylcholine at the nerve-muscle junctions, causing muscular spasms [37,38]. Carbamate insecticides operate in a similar fashion to organophosphate insecticides by inhibiting acetylcholinesterase, and are therefore also neurotoxic [39]. They have caused endocrine disruption, muscular disorders, respiratory distress, and some are potentially carcinogenic [40]. Herbicides control weeds primarily through the inhibition of photosystem I, photosystem II, acetyl-CoA carboxylase enzyme, EPSP synthase or the acetolactate synthase enzymes [41]. Glyphosate-based herbicides are among the first used herbicides, and are still globally used [42], despite concerns being raised about probable carcinogenicity [43]. These herbicides have been found to cause adverse liver and kidney effects in animal studies [44].

Human exposures to the pesticides are primarily occupational, with some environmental exposure. Whereas acute exposures to pyrethroids, organophosphate and carbamate insecticides cause the typical symptoms of pesticide exposure, such as muscle weakness, sweating and gastrointestinal upset, chronic exposures have been associated with nausea, vomiting, blurred vision, among other symptoms [45]. Although, there is limited data on the health effects of exposure to herbicides, exposures to some herbicides, such as phenoxy herbicides, have been associated with chloracne [14], whereas acute ingestion of glyphosate herbicides may cause gastrointestinal erosion, hemorrhaging, as well as sore throats [46].

4. THE EFFECTS OF FORESTS ON ENVIRONMENTAL FATES

Environmental fates and distribution studies about organic pollutants give information on the short- and long-term environmental concentrations, exposure media, time period during which the hazard potential is greatest, as well as the factors affecting their respective fates. Further understanding of pollutants and their behavior in the environment comes from physiochemical studies of the organic compounds, field measurements of environmental concentrations/distribution and from transport models.

It is well known that climate affects land cover. However, it is being recognized that land cover also influences climatic conditions. Increasing the vegetative density has been shown to increase albedo and total latent heat flux, while simultaneously decreasing sensible heat flux, thereby resulting in cooler and wetter climatic conditions [47]. The vegetative cover and soil distribution in ecosystems affect climate through changes in the balance of water, momentum and energy, and grasslands and pastures display lowered humidity, lowered precipitation and higher surface temperatures [48]. Tropical deforestation has been associated with climatic changes, such as increased temperatures and albedo, as well as with decreased evaporation, cooling and precipitation rates [49,50]. There is clearly a feedback system between climate and land cover. Since climate affects the fates of organic compounds, it was reasonably hypothesized that land cover also influences the fates of these chemicals.

Land use/cover influences the fates of organics in the environment because the retention capacity of the surface for the chemicals is determined by conditions such as vegetative cover [51]. There are studies that correlate land use/cover with either pollutant fates or some influential factors of their fates. For instance, the contamination of surface waters depends on land use [52]. The uptake by vegetation is more important in reducing atmospheric concentrations than uptake by soil for organics with log \( K_{OA} > 6 \) [53]. Also, land use/cover influences the volatilization fluxes of POPs to the atmosphere [54] whereas plant cover controls run-off [55]. Hence, contaminant fates depend on land use/cover.

There are a few studies assessing the influence of specific land covers, such as forests, on the fates of organic contaminants. Temperate forests are thought to reduce the atmospheric concentrations of a subset of organics, specifically semi-volatile organic compounds with \( 7 < \log K_{OA} < 11 \) and \( \log K_{AW} > -6 \), by transferring these to forests soils [56]. With reduced atmospheric concentrations of these semi-volatile organic compounds, there is the resultant reduction in atmospheric deposition to surface waters. However, there is increased delivery to the surface waters via forest soil run-off, although to a lesser degree [11]. In examining the role of forests in a Canadian watershed, the vegetative cover was alternated between forested and urban coverage, and it was found that forests soils may be reservoirs for semi-volatile organic compounds [57]. Therefore, these forests are filters of many organic contaminants and their soils, the reservoirs.

First, the forest canopy uptakes the chemicals from the atmosphere via gaseous diffusion, wet
The filter effect for organic contaminants, especially the more polar current-use pesticides. It is yet unexplored whether the combined high temperatures and precipitation rates of some tropical regions allow for organic contaminants to be filtered from the atmosphere. A recent study demonstrated that tropical forests exhibit a filter effect for PBDEs [61]. However, it is not known how the probable filtering effect of tropical forests may vary with forest structure and coverage, as well as climatic conditions.

Although vegetative compartments affect the atmospheric concentrations of organic contaminants, by uptake from the atmosphere [53], re-emissions to the atmosphere are expected under select conditions. Organics with higher $K_{oa}$ values ($8 \leq \log K_{oa} \leq 10.7$) can be re-emitted during seasons with higher temperatures [10]. Therefore, forests may act as secondary sources of organic pollutants at the local or regional scale. The role of tropical forests in acting as secondary sources of organic pollutants has not been adequately explored [12]. To add to considerations of forests’ influences on environmental fates, changes to agrochemical use, due to changes in land use, such as shifts from arable to forested land, were found to influence environmental distribution of pesticides with reductions in atmospheric concentrations associated with increased forested acreage [62]. Therefore, despite a potentially smaller filter effect, tropical forests may use other processes to control the fates of organic contaminants, including those that are less persistent.

Many current-use pesticides, such as organophosphates and carbamates are more polar (with low air-water and octanol-water partition coefficients) and less persistent [63]. Some possess partition coefficients outside of the forest ‘filter window’. While as yet unknown, it is possible that the filter effect extends to these products, especially since the forest filter effect is expected to vary widely because the filter factor is dependent on a number of parameters including deposition velocities, temperature, the canopy storage capacity for lipophilic organic contaminants, as well as particle-air partitioning [56]. Whereas it is internationally accepted that tropical forests provide critical environmental and economic services, such as habitat, biodiversity conservation, timber production and carbon sequestration [64,65], these biomes may also provide the additional benefits of human and ecosystem health preservation as they regulate organic contaminants.
Hazard assessment end-points, such as overall persistence and long-range transport potential, as well as steady state concentrations, are often used for the purpose of evaluating or predicting environmental impact [11,66-71]. The long-range transport potential (LRTP) of organic contaminants speaks to the ability of the locally emitted contaminants to be transported over large distances [72] via air (LRTP\textsubscript{a}) or water (LRTP\textsubscript{w}), and is therefore indicative of the capacity to cause adverse effects on larger scales – regional, continental or global [7,73-75]. Quantified metrics that allow for the evaluation of long-range transport potentials include those that are transport based, such as the characteristic travel distance, as well as those that are target oriented, such as the arctic contamination potential [75]. The characteristic travel distance is defined as the distance over which the contaminant is transported such that its concentration is 1/e (37%) of its original [74]. For an organic contaminant evaluated using the characteristic travel distance (CTD) and travelling at a height of 1000 m in the atmosphere at 25 °C, employed classifications for the contaminant are as follows: Class 1 – atmospheric CTD of greater than 2000 km; Class 2 – atmospheric CTD of 700 to 2000 km; and Class 3 – atmospheric CTD of less than 700 km [7].

The overall persistence at its simplest is a measure of the tendency or ability to maintain presence in the given environmental system [76]. Whereas single media persistence considers contaminant presence in an individual medium (air, water, among others), overall persistence assumes total presence, given a set of linked environmental media. Overall persistence is measured using half-lives [76] or steady-state residence times [75]. The residence time is the time taken for the contaminant to be degraded to 1/e of its original concentration. Therefore, overall persistence depends primarily on degradation losses from the entire environment [76,77].

It is recommended that hazard assessments focusing on hazard end-points, such as overall persistence and long-range transport potential, be conducted prior to toxicological assessments [77]. The overall persistence and long-range transport potential of a few persistent pollutants have been assessed [7,70]. Also, some current-use pesticides, which tend to be more polar than their halogenated counterparts, have been found to display regional transport potential [78]. There is little information about the way forests affect the above hazard assessment end-points and any associated health effects of the organic pollutants. In a study on the contribution of forests to the fates of select organic compounds, the observed reduced atmospheric concentrations led the authors to suggest that forests may be important in reducing the long range transport potential of many organic contaminants [11]. In a global study, forests reduced atmospheric and aquatic (oceanic and freshwater) concentrations of the studied organic compounds [59]. Therefore, in reducing media-specific concentrations, forests alleviated media-specific health risks. However, the overall global persistence/residence times due to delivery to forest soils was increased [59], with potentially enhanced risk via this medium.

Therefore, with the potential decreases to long range transport potential and persistence in the atmosphere in temperate regions and globally, in addition to observed global increase in overall persistence, it is possible that tropical forests may exhibit similar effects on these variables for organic pollutants – persistent and less persistent. One important question that comes to mind is: are tropical forests among those that can influence the fates of a chosen subset of organics? This area of research has been little explored. Also, studies assessing the fates and health effects of POPs and current-use pesticides in tropical regions, given the presence of forests at a site with other co-existing land uses, such as agriculture or urban centers, have not been identified.

To summarize, the impact of tropical forests on the environmental fates of some organic pollutants, such as PBDEs, dioxins, furans and current-use pesticides, especially at the watershed scale with co-existing land uses, is yet unknown. Information as these would aid in the natural resources management of tropical watersheds.

5. CLIMATE CHANGE EFFECTS

The major determinants of climate are precipitation and atmospheric temperature [79]. These variables influence parameters responsible for the environmental distribution of organic pollutants. For example, partition and transport coefficients are functions of temperature and/or precipitation rate. The mobility of persistent organic pollutants is dependent on climatic conditions, such as temperature, wind and precipitation distribution
Increasing temperatures result in increases in the primary volatilization of select organic contaminants [80-83]. For each 1°C increase in temperature, the half-life of pesticides in soils is expected to decrease by as much as 60% [84]. Increased precipitation is associated with increased wet deposition and delivery to terrestrial and aquatic surfaces [8]. Therefore, changes to the climate variables, temperature and precipitation rate, are expected to alter the environmental fates of organic chemicals. These are the direct impacts of climate change on the fates of organic contaminants.

The indirect impacts of climate change on the fates of organic contaminants are difficult to quantify and assess [83]. Agrochemicals will herein be used for elucidation. Regional changes in climate are expected to result in subsequent shifts in the types and size of pest populations. For instance, insect proliferation is expected with rising temperatures [84]. Also, as the climate changes, geographical shifts in the types and quantity of crops produced are expected [51]. To add to this, pesticide losses may occur from processes such as volatilization, degradation, erosion or run-off with consequential increased applications to compensate for the losses [8].

Since pesticide use influences the fates of these chemicals, climate change may indirectly affect their fates, by governing their use patterns.

All these studies [62,71,80,82-84] demonstrate that it may be difficult to predict the exact impact of climate change on environmental fates, because of the complex nature of climate processes. Nevertheless, efforts to quantify the potential impacts of climate change on the fates of organic pollutants, under a limited number of climate change scenarios, are on the rise. It has also been shown that pesticide leaching was dependent on the specific climatic conditions [85]. For example, they found that leaching was generally higher when pesticides were applied in the autumn than in the spring. In another study, the atmospheric concentration records, since the 1990s, of select organic chemicals, were compared with arctic variables, such as surface air temperature and sea-ice extent, and it was found that the increasing atmospheric concentrations of these compounds as the arctic warms were due primarily to revolatilization [81].

In yet another study using the multimedia model EVn-BETR, climate change scenarios included changes to land cover, precipitation and temperature, in the time periods 1991-2020, 2021-2050, 2051-2080 and 2071-2100, to assess the resultant impacts on the fates of polychlorinated biphenyls (PCBs) and PBDEs [82]. It was found that temperature was the major determinant for atmospheric fates, with reduced concentrations due to increased degradation and volatilization rates when the temperatures increased. To show that temperature most strongly affected the global atmospheric fates of some organic contaminants, BETR Global was used to create two climate change scenarios (for the periods 1981-2000 and 2080-2099) by varying select climate variables, such as temperature fields, wind fields, ocean current fields and precipitation rates [80]. Using BETR Research, a global-scaled seven compartment (upper air, lower air, sediment, soil, fresh water, ocean water and vegetation) multimedia model, to compare emissions with climate change scenarios, it was observed that increased temperature was the chief determinant of increases in arctic ocean and atmospheric concentrations of α-hexachlorocyclohexane (α-HCH), due to atmospheric transport to the arctic [62].

Increasing numbers of studies are comparing climate and land use change effects on the fates of organic compounds. Shifts from arable land to forests reduced volatilization fluxes from soils, whereas increases in temperature caused increased volatilization fluxes from soils [54]. This suggests that land use/cover may counteract or temper some direct effects of climate change.

The assessments described above were primarily conducted for the arctic region, or otherwise at a global scale. Watershed scaled assessments of climate change effects on the environmental fates of organic contaminants, in tropical regions, have not been identified. The studies involving climate change impacts are predictive and, as such, require the use of multimedia models. Some of these models, such as G-CiEMS [86], BasinBox [87], and SESAME [88], are site-specific and not applicable in other places. In other models, such as GIM3 [89] and MUM [67], only one vegetative cover is incorporated, with the application of weighted parameters, limiting their application in multi-use tropical watersheds. Finally, there are watershed models, like LOIS [90] or even GREAT-ER [91]; however, these are specifically for evaluating water quality and not environmental distribution. Consequently, the influence of any vegetative cover, especially tropical forests, in the regional environmental fates of organic pollutants in a
watershed with co-existing land uses/covers has not been adequately examined using multimedia modeling. In addition to this, assessments on the impacts of climate change on the long-term fates and associated health impacts of select organic contaminants in such watersheds, as well as on the roles played by tropical forests in governing these fates and health impacts, are limited. In keeping with the framework outlined in Fig. 1, the development of appropriate public health and environmental policies depend on such environmental fates data for organic pollutants.

6. CONCLUSION

PBDEs, dioxins, furans and current-use pesticides are among those organic pollutants that are now ubiquitous, associated with adverse effects and, therefore, have raised global concern. However, in order to manage and develop policies for these chemicals, so that their environmental and health impacts can be mitigated and/or controlled, information on the associations between their emissions, fates and effects need to be established, as shown in Fig. 1. In this triangular concept for the associations (emission-fate-effect), detailed research on each of these vertices is necessary. Whereas there are studies on the fates and influential factors in temperate areas, there are critical knowledge gaps for tropical regions, especially in developing countries. Among these critical knowledge gaps are:

- The influence of tropical forests on the long-term environmental fates and health impacts of the select organic contaminants in a multi-use watershed;
- The effects of climate change on the long-term environmental fates of the organic chemicals in a tropical region;
- The effects of climate change on the influence of tropical forests on the environmental fates and hazard potential of the contaminants;

To conclude this review, it is being purported that once these knowledge gaps are filled, through relevant and well-designed research, the environmental management of these organic pollutants in tropical regions, especially tropical developing countries, can be improved.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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