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Review article

Critical assessment of the ubiquitous occurrence and fate of the insect repellent *N*,*N*-diethyl-*m*-toluamide in water

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ABSTRACT

The insect repellent diethyltoluamide (DEET) is among the most frequently detected organic chemical contaminants in water across a wide range of geographies from around the world. These observations are raising critical questions and increasing concerns regarding potential environmental relevance, particularly when the emergence of severe neurological conditions attributed to the Zika virus has increased the use of insect repellents. After dermal application, DEET is washed from the skin when bathing and enters the municipal sewer system before discharge into the environment. Mainly measured by gas chromatography or liquid chromatography coupled to mass spectrometry (GC-MS or LC-MS), more than 200 peer-reviewed publications have already reported concentrations of DEET ranging ng/L to mg/L in several water matrices from North America, Europe, Asia, Oceania, and more recently Africa and South America. While conventional wastewater treatment technology has limited capacity of removal, advanced technologies are capable of better attenuation and could lower the environmental discharge of organic contaminants, including DEET. For instance, adsorption on activated carbon, desalinating membrane processes (nanofiltration and reverse osmosis), ozonation, and advanced oxidation processes can achieve 50% to essentially 100% DEET attenuation. Despite the abundant literature on the topic, the ubiquity of DEET in the environment still raises questions due to the apparent lack of obvious spatio-temporal trends in concentrations measured in surface water, which does not fit the expected usage pattern of insect repellents. Moreover, two recent studies showed discrepancies between the concentrations obtained by GC-MS and LC-MS analyses. While the occurrence of DEET in the environment is well established, the concentrations reported should be interpreted cautiously, considering the disparities in methodologies applied and occurrence patterns observed. Therefore, this manuscript provides a critical overview of the origin of DEET in the environment, the relevant analytical methods, the occurrence reported in peer-reviewed literature, and the attenuation efficacy of water treatment processes.

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1. Introduction

The constant progress in analytical chemistry over the last decades, and particularly the increasing sensitivity, dramatically improved the detection of trace organic contaminants (TOrCs) even within matrices as complex as wastewater. For instance, with ultra-high performance liquid chromatography and tandem mass spectrometry (UHPLC-MS/MS), many laboratories routinely and reliably quantify dozens of pharmaceuticals and personal care products (PPCPs) in various water qualities at concentrations below 1 ng/L (Anumol et al., 2013). Consequently, multiple monitoring studies have now been published, providing a global view as to TOrC occurrence in aqueous environments.

Among the TOrCs commonly monitored in water, the insect repellent diethyltoluamide (DEET) became of particular interest when the first reconnaissance study of wastewater contaminants in US streams revealed its occurrence in 74% of the samples analyzed (Kolpin et al., 2002). Since then, subsequent research confirmed its occurrence in various water matrices in Europe (Loos et al., 2010, 2013a; Sgroi et al., 2016), Asia (Anumol et al., 2016; Hanamoto et al., 2013; Heeb et al., 2012), Oceania (Costanzo et al., 2007; Smith et al., 2012), and more recently South America (De Gerónimo et al., 2014) and Africa (Sorensen et al., 2015). Such ubiquity of DEET in environmental waters and its detection in drinking water (Benotti et al., 2009a; Padhye et al., 2014) have raised concerns regarding public health and the efficacy of water treatment for attenuation. In the recent years, the continuous water stress and the growing interest for potable water reuse have further stimulated research on the removal of various wastewater contaminants, including DEET, by conventional and advanced treatment processes. These include biological, adsorption, membrane, and oxidative technologies.

The rising number of publications over the last decade has increased knowledge and awareness on the occurrence and fate of DEET. However, the ubiquitous detection of this chemical, even in some geographic areas and seasons where insect repellents are not expected to be heavily used, also raised some interrogation regarding a potential mimic in the environment or analytical interference (Merel et al., 2015a). Therefore, the first objective of this critical review is to provide a comprehensive understanding of the sources of DEET and its occurrence in all types of water matrices, including landfill leachate, wastewater, surface water, groundwater and drinking water, through data published from 1990 to 2015. A second objective of this review is the description of analytical procedures found in the literature for the analysis of DEET, with particular emphasis on their respective strength and limitation. More particularly, potential interferences that could induce a bias in the quantification of DEET will be discussed. Finally, this review aims at assessing the attenuation of DEET by a wide number of water treatment processes and natural processes with an overview of relevant transformation products.

2. Uses, commercialization, and consumption of DEET

The short and popular name DEET refers to the chemical diethyltoluamide, also sometimes called detamid, dieltamid, *N*,*N*-diethyl-*m*-toluamide or *N*,*N*-diethyl-3-methylbenzamide (Fig. 1). This chemical enters the composition of several products serving different needs. Therefore, this section intends to provide an overview of the different uses of DEET while examining its commercial distribution and assessing its consumption.

2.1. Main uses of DEET

DEET was developed by the US army in 1946 to protect troops deployed in certain areas from mosquito bites and related diseases (Kitchen et al., 2009; Schoenig and Osimitz, 2001). In 1957, DEET was made available to public and quickly became widely used as the active component of many commercial insect repellents. Accessible in different forms such as spray, liquid, or lotion for skin application, DEET may also be incorporated in products like clothes or mosquito nets for an increased protection against some diseases transmitted by vector bites in endemic areas (Kitchen et al., 2009; Pennetier et al., 2010). In addition, DEET is also used as an insect repellent on some animals. For example, the US EPA DEET reregistration eligibility decision factsheet from 1998 (EPA, 1998) includes application to cats, dogs, horses and pet living/sleeping quarters in the DEET use profile.

Potential uses of DEET other than those mentioned by the US EPA are possible, but specific information is scarce. For instance, DEET might also be used to protect animals other than pets against mosquito bites, as suggested by a study that attributed the detection of DEET in the air of a rural area to livestock sprayed with insect repellent (Aronson et al., 2012; Cheng et al., 2006). Moreover, another publication demonstrates the efficacy of DEET as antifeedant (Lee et al., 2010), which suggests a potential use in agriculture to repel and prevent insects from feeding on crops or fruits. The pesticide database from the pesticide action network (PAN, 2014) also documents the use of DEET in California for landscaping, structural pest control, outdoor flower nursery and greenhouse plants. Due to its ability to penetrate through the skin, DEET has also been considered as a potential dermal penetration enhancer to improve the dermal and transdermal delivery of drugs (Windheuser et al., 1982). Finally, a recent study also reported DEET being used for chemical applications such as resin solvent, surface plasticizer as well as carrier for flame retardants or dyes (Aronson et al., 2012). However, despite these references to other potential uses of DEET, its incorporation as the active ingredient of commercial insect repellent should remain largely predominant.

2.2. Commercialization of DEET-containing products

DEET exists at different concentrations within formulation of various commercial products, mainly as an active component of insect repellents. While gathering information on DEET-containing products commercially available worldwide is an overwhelming task, it can be easily done in USA where commercial insect repellents are registered by the environmental protection agency (EPA) which makes the list readily accessible (EPA, 2013). According to the last update from April 2013, a total of 63, 114, 182 and 63 registration numbers were reported respectively for products containing DEET at 0–10%, 10–20%, 20–30%

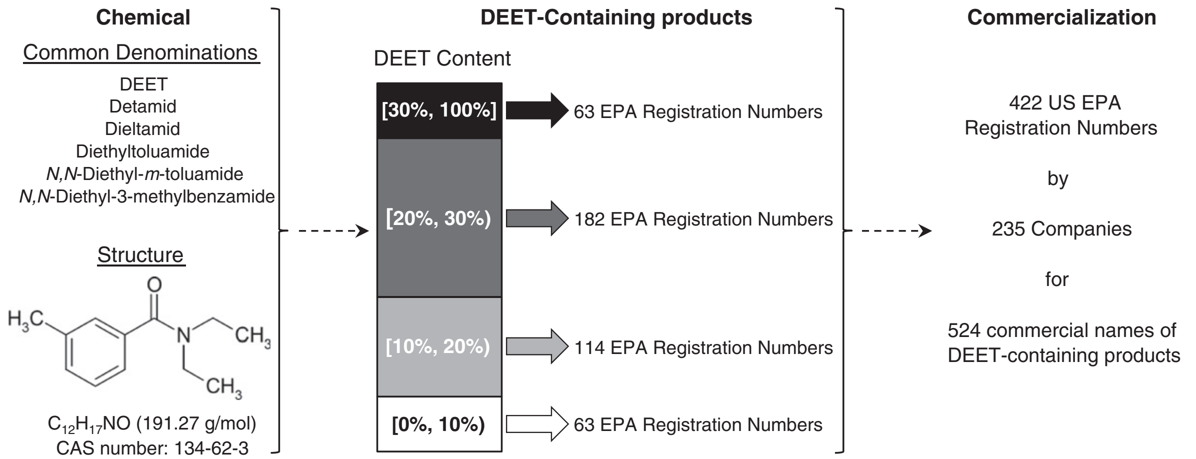


Fig. 1. Chemical names and structure of DEET with US EPA statistics on commercial DEET-containing products.

and 30–100% (Fig. 1). Overall, the EPA website counts a total of 422 registration numbers held by 235 companies for 524 commercial names of DEET-containing products. In comparison, the amount of registration numbers for other common active components of insect repellents is much more limited. For example, picaridin, *p*-menthane-3,8-diol, catnip oil and citronella respectively count 25, 5, 4 and 3 registration numbers. While these values might not be directly related to the relative consumption of each compound, they tend to designate DEET as the largely predominant active chemical. However, this EPA database is limited to insect repellents and does not account for other potential uses of DEET.

2.3. Annual consumption of DEET

The annual consumption of DEET worldwide or per country remains largely unknown, and its assessment represent a major challenge for multiple reasons. A first and usual approach to assess the consumption of DEET would consider the production volumes of the manufacturing companies. However, identifying these manufacturing corporations remains challenging since they could be implanted in virtually any country then export the native DEET to the USA and elsewhere where other companies incorporate it in the formulation of their own registered commercial products. Moreover, given the very high number of companies holding US EPA registration numbers for DEET-containing products, identifying for each of them their DEET provider is unrealistic. Another approach would consider the sales volume. Since DEET is widely available to consumers in numerous brands of products, a plethora of potential distribution points exist and sales volume remain unknown. Yet another approach would rely on the detection of DEET in wastewater and deduce an average consumption, as previously done with drugs of abuse (Meyer et al., 2015; Rodríguez-Álvarez et al., 2015). Nevertheless, a recent study showed a strong seasonal variability of DEET concentration in the wastewater influent which was no longer reflected in the wastewater effluent (Merel et al., 2015a). Therefore, any assessment of DEET consumption through wastewater analysis should be performed on raw sewage with diurnal and flow-proportional composite sampling over a full year but such studies are relatively rare.

The lack of data regarding DEET consumption often results in citing the approximate value of 1.8 million kg per year in USA as a reference (Chen et al., 2012; Merel et al., 2015a). However, such consumption approximation should be considered with caution since it dates back to the reregistration eligibility of DEET, in 1998, and was itself derived from pesticide usage information for the year 1990. Therefore, more than two decades later, such approximation might no longer be accurate. Moreover, it should not be extrapolated to other areas with different socio-economic level or where the prevalence of diseases transmitted by mosquitoes is higher. For instance, in areas where mosquitoes transmit the dengue virus and are likely to cause outbreaks of dengue fever, insect repellents are used for public health and the application of DEET is expected to be higher than in other areas where insect repellents are mostly used for comfort during outdoor activities. In addition, with the recent emergence of infections by Zika virus causing severe neurological complications in the French Polynesia and potentially associated with cases of microcephaly in Brazil (Musso and Gubler, 2016), the use of insect repellent is expected to significantly increase in affected areas. Consequently, much research remains necessary to estimate the magnitude of DEET consumption and to establish a parallel with concentrations reported in the aqueous environment.

3. Overview of literature available on DEET

3.1. Distribution of literature between DEET and other active compounds

Several active chemicals can be used in the formulation of commercial insect repellents. However, the peer-reviewed literature available regarding these active compounds is largely dominated by publications focusing on DEET (Fig. 2). Searching the Thomson Reuters Web of Science database, as described in a previous bibliometric study (Merel et al., 2013), illustrates this pattern. For instance, searching for publications associated with the topic “DEET” AND “insect repellent” resulted in finding 1,165 articles. Searching for publications associated with the topic “Picaridin” AND “insect repellent” allowed finding only 86 articles. A similar search for other active compounds of insect repellents, such as IR3535, nepetalactone (catnip oil), *p*-menthane-3,8-diol or the more recent SS220 allowed finding even fewer documents.

3.2. Overall distribution of literature on DEET

Among the 1,165 publications on the topic “DEET” AND “insect repellent” found previously through the Thomson Reuters Web of Science database, the first article was published in 1957, but the number of publications per year remained limited until 1980. In contrast, since 1980 the amount of literature available has continuously increased, reaching a total of 1,165 at the end of December 2015. These publications mostly cover the fields of entomology (efficacy to repel different mosquito species), chemistry (synthesis and reactivity), and toxicology, but studies on environmental occurrence were limited. However, this apparent disproportionality mostly results from the search method. Indeed, searching for the topic “DEET” AND “insect repellent” in the Thomson Reuters Web of Science database only provides publications that include each of these three terms in their title, abstract or keywords. Thus, while

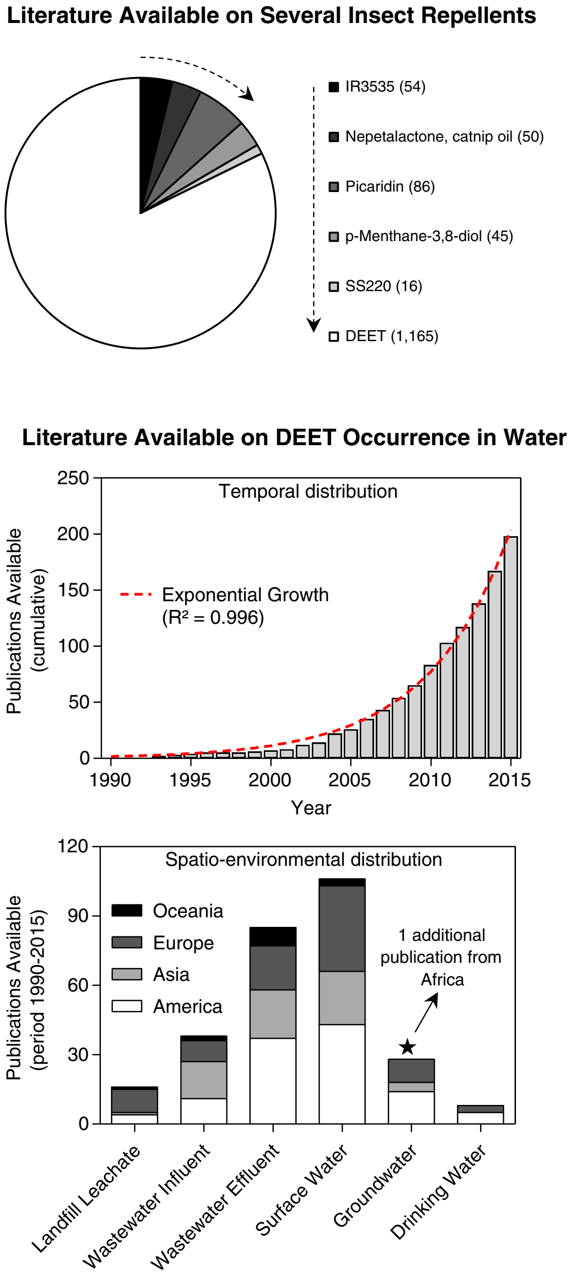


Fig. 2. Distribution of literature on insect repellents and the occurrence of DEET in water.

chemical and toxicological studies that usually focus on a single compound are likely to fulfill these criteria and are easily found, occurrence studies that usually detect DEET among tens or hundreds of other contaminants are not and require less specific search criteria.

3.3. Literature on DEET occurrence in water

Occurrence studies on DEET in water were initially found searching for the topic "DEET" AND "water" in the Thomson Reuters Web of Science database. Additionally, a few other studies were also found when repeating the same search but replacing "DEET" successively by all the other names of the chemical (Fig. 1). An additional search was also performed through the ScienceDirect database and the journal "Environmental Science & Technology" that allowed finding publications including DEET or one of its synonyms (Fig. 1) anywhere in the text.

Among the occurrence studies, the first to report the detection of DEET in the aqueous environment was published in 1993 and identified the insect repellent in samples of Swedish municipal landfill leachate collected in 1990 (Öman and Hynning, 1993). In the same year, a second study also reported the detection of DEET in a different geographic area and water matrix by identifying the insect repellent in samples from the Mississippi river (USA) collected in 1991–1992 (Pereira and Hostettler, 1993). Since then, the amount of publications reporting the occurrence of DEET in water has grown exponentially, reaching a total of 198 at the end of 2015 (Fig. 2). Even though most of the literature focuses on surface water and wastewater effluent, occurrence data for landfill leachate, wastewater influent, ground water and drinking water also have been reported (Fig. 3).

Reports on DEET occurrence are becoming increasingly global. While a large portion of publications still comes from America and Europe (Fig. 2), the number of reports from Asia and Oceania significantly increased in the last few years. However, when plotted on a map, these occurrence studies remain unevenly distributed. For example, almost all the publications from America actually come from USA, with only a single one from South America (De Gerónimo et al., 2014). Similarly, European occurrences studies are limited to Western countries, and Asian reports are largely dominated by China despite some high concentrations of DEET also reported in Indonesia (Dsikowitzky et al., 2014). Finally, only a single study reporting the occurrence of DEET in African waters could be found (Sorensen et al., 2015).

4. Origin of DEET occurrence in the aqueous environment

The occurrence of DEET in the aqueous environment involves multiple and potentially complex routes (Fig. 4). While a primary source of environmental occurrence of DEET could be the discharge of contaminated process effluents, either from manufactures of DEET or manufactures using this chemical, the strict law enforcement regarding environmental protection strongly hinders such hypothesis. In addition, such point source pollution would mostly result in high concentrations of DEET in limited areas, which is not consistent with actual environmental occurrence data showing a widespread but more diffuse contamination. However, despite the fact that no publication (including the grey literature) could be found to sustain it, such possibility should not be totally neglected in countries where industrial constraints regarding environmental protection are limited.

Although often neglected in the past due to their low individual contribution, households are now expected to be the major source of DEET in the environment. For example, following dermal application, DEET is removed from the skin when bathing and then collected in the wastewater system. While households in emerging countries along with remote and older houses in industrialized nations might generate a diffuse contamination through direct discharge of wastewater into the environment or a leaching septic tank, most urban households in industrialized nations are connected to a central wastewater treatment plant. However, with conventional treatment providing limited removal (see section on the fate of DEET), DEET is partially discharged with wastewater effluent into surface water or even groundwater in areas implementing aquifer recharge. Such receiving waters being often used as drinking water sources, DEET can be detected in water intended for domestic and human consumption (Benotti et al., 2009b; Padhye et al., 2014). Moreover, with an average log K_d of 1.91 (Hyland et al., 2012), the fraction of DEET retained in wastewater sludge (Chari and Halden, 2012) could reach other environmental compartments when such sludge is spread on agricultural soil or buried in landfills.

Landfills, and particularly landfill leachates, represent another source of DEET in the environment that remains poorly considered. As mentioned earlier, DEET could enter landfills through the burial of

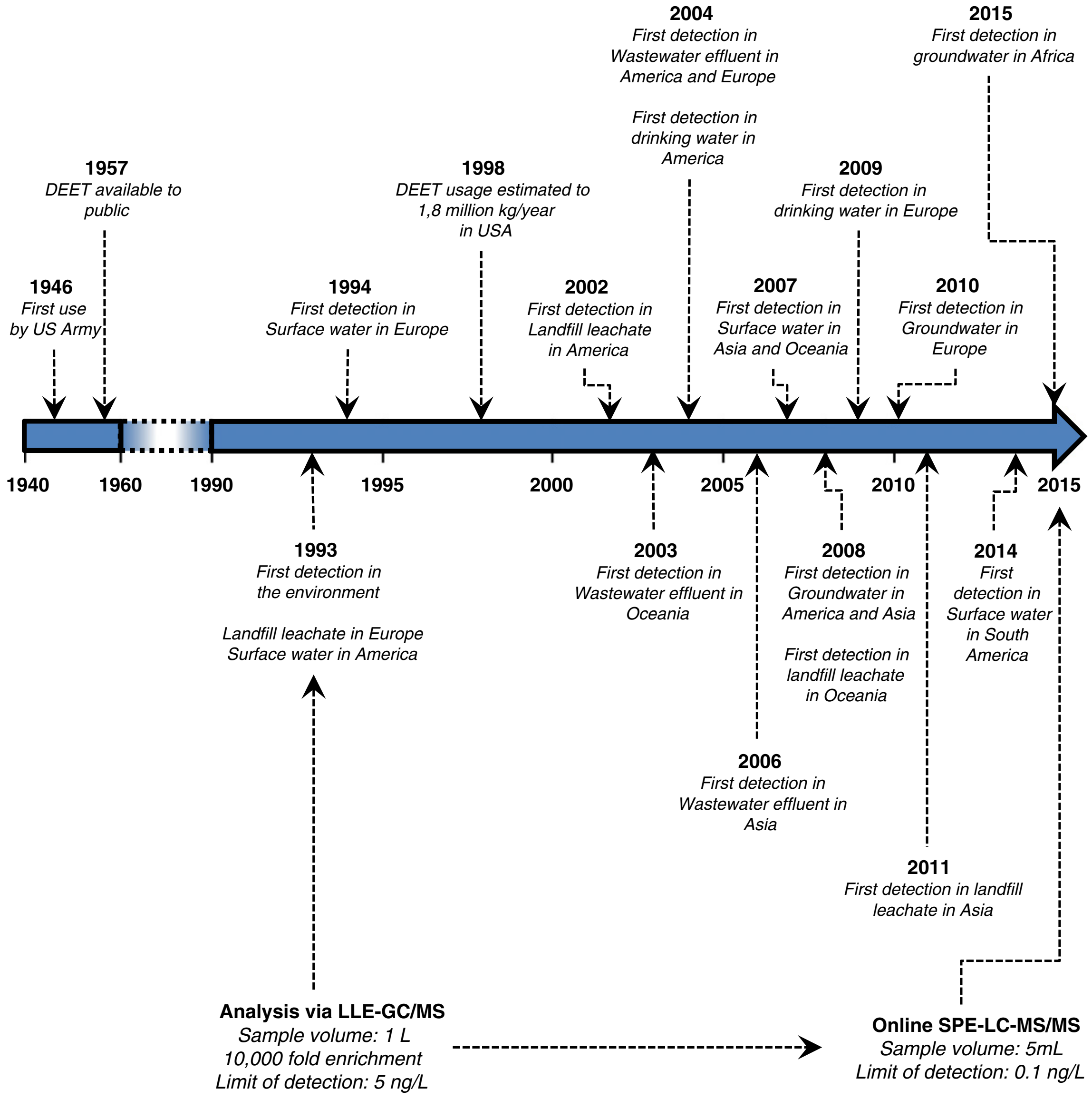


Fig. 3. Chronology of DEET detection in different water matrices and world regions.

wastewater treatment sludge or through domestic disposal of expired/unused DEET-containing products. Then, DEET accumulates in leachates which are either infiltrated into the ground or collected and sent to the local wastewater treatment plant. Even though relatively few studies have characterized the occurrence of DEET in landfill leachates (Fig. 2) the concentrations detected can be orders of magnitude higher than those reported in wastewater, sometimes even reaching the mg/L range (Clarke et al., 2015).

Alternative and potentially limited sources of DEET in the environment should also be considered even though the lack of relevant peer-reviewed research does not allow their accurate characterization. For instance, while DEET is mostly used as dermal application of insect repellent, direct transfer to water bodies used for recreational activities (swimming, waterskiing, etc.) is also a plausible source of environmental contamination, particularly in touristic areas. Similarly, the use of sludge and/or reclaimed water containing DEET in agriculture might lead to soil contamination or river contamination via runoff (Fig. 4). Finally, atmospheric transport of DEET should also be considered since a limited number of studies have reported its occurrence in aerosols from indoor and outdoor sampling campaigns in urban as well as rural areas (Balducci et al., 2012; Cheng et al., 2006; Dall'Osto et al., 2007).

5. Analytical methods for the quantification of DEET in water samples

Accurate identification and quantification of chemicals in the environment relies on appropriate procedures for sampling, sample transportation, sample preservation, chromatographic separation, and detection. Therefore, this section will consider each of these aspects with respect to the analysis of DEET.

5.1. Sample collection, transportation, and preservation

Monitoring reliably DEET and other environmental contaminants implies developing a sampling strategy that ensures collecting a representative sample. For instance, a river sample should be representative of the width, depth and flux, as described in the US Geological Survey field manual (USGS, 2015). Similarly, a wastewater sample should consider the intraday fluctuation of the input in terms of both the flowrate and the concentration of chemicals. Moreover, when comparing the removal provided by wastewater treatment plants, the sampling strategy should also consider the hydraulic retention time (Gerrity et al., 2011a). Such considerations tend to exclude the collection of grab samples,

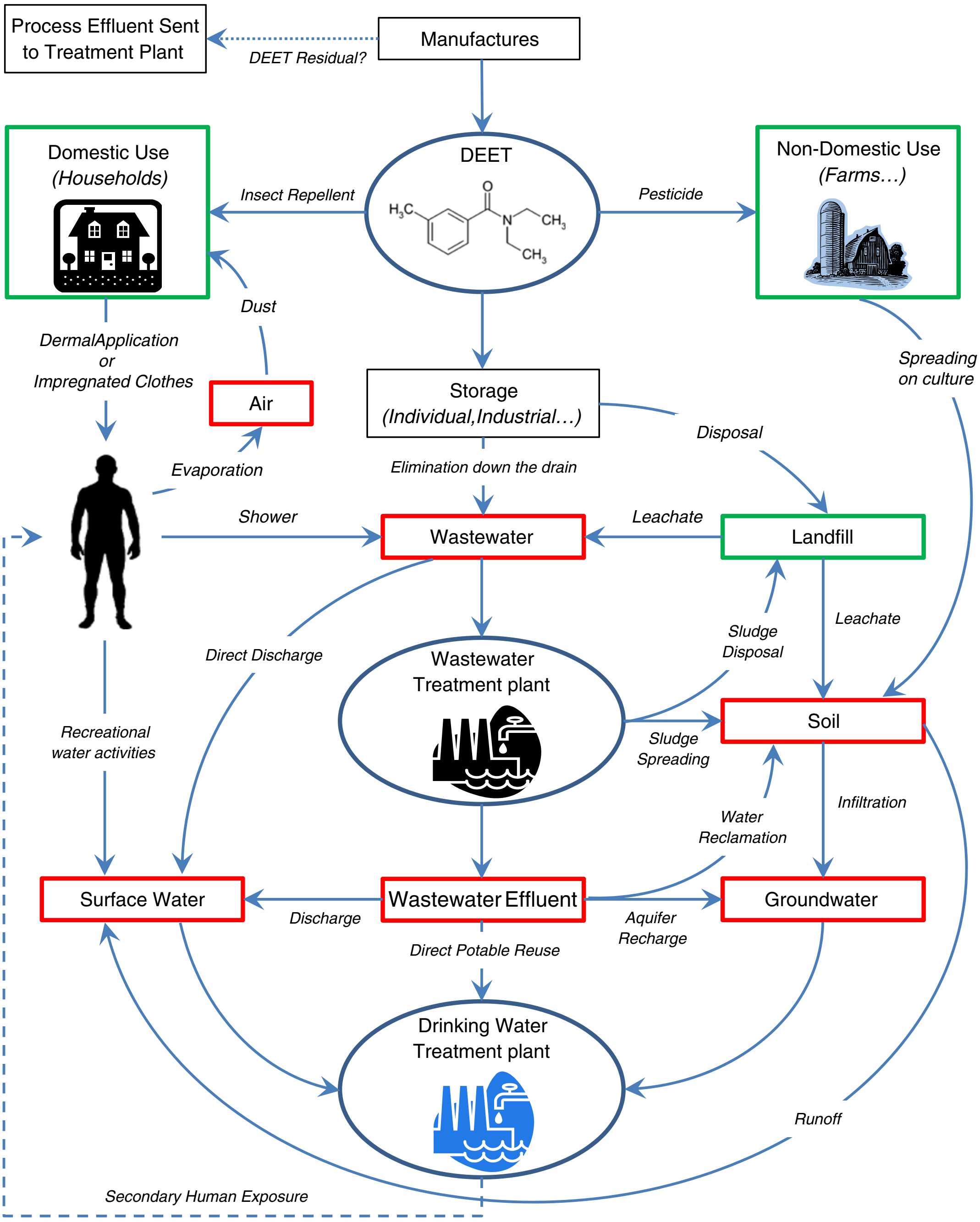


Fig. 4. Sources and routes of DEET in the environment.

giving priority to composite samples. However, most of the studies reporting the occurrence of DEET in water were carried out with single to triplicate grab samples per sampling point, which is enough to confirm the ubiquity of the insect repellent but insufficient to accurately characterize the magnitude of water contamination (potential diurnal variation). Only in the last few years composite samples became more popular for wastewater analysis while several studies reported surface water contamination using passive samplers (Alvarez et al., 2014a, b; Bargar et al., 2013; Bartelt-Hunt et al., 2009; Bidwell et al., 2010; Coes et al., 2014; Writer et al., 2010; Zenobio et al., 2015). Polar organic chemical integrative samplers (POCIS) deployed in water for a specific period offer a time-integrated semi-quantitative average of DEET occurrence. While converting values obtained from POCIS to a concentration of chemical in water remains difficult, POCIS and other types of continuous passive samplers are particularly useful for toxicological studies by providing a good assessment of exposure of biota to trace organic contaminants.

After determining the sampling strategy, a major concern is the potential contamination of water samples during or after collection. For instance, even though no peer-reviewed study showing evidence of sample contamination by direct transfer from the skin could be found, several studies published by the US Geological Survey clearly mention that using insect repellent was discouraged during sample collection and processing (Barnes et al., 2008; Focazio et al., 2008; Kolpin et al., 2002, 2004; Sandstrom et al., 2005). In addition, to account for any contamination resulting from sampling procedure, personnel, device or glassware, it is crucial to collect field blanks, which involves transporting and transferring previously characterized (DEET-free) water to the actual sampling location and transferring into a sample bottle. The concentration of the analyte in the field blank is then either mentioned in the report or subtracted from the concentration measured in the water samples.

Once samples have been collected (preferably avoiding headspace in the container), transport to the laboratory should occur with the minimum delay while keeping the samples cool and in darkness. The analysis usually being performed within 14 days, sample preservation is critical. Typical procedures reported in the literature involves simple storage at 4 °C, acidification (pH 2) with sulfuric acid (Trenholm et al., 2006), addition of sodium azide to prevent bacterial degradation (Anumol et al., 2013; Anumol and Snyder, 2015). While such procedures are effective for most common trace organic contaminants, a recent preservation study showed that with and without the addition of acid or azide, sample storage at 4 °C was not recommended for DEET since compound recovery was higher than 115% (Vanderford et al., 2011). However, the same study demonstrated that simple storage at −20 °C or simultaneous addition of acid and azide provided adequate sample preservation up to 28 days.

5.2. Sample extraction and analysis

Analysis of trace organic contaminants in water samples is often preceded by the enrichment of the analyte, either through liquid-liquid extraction (LLE) or solid phase extraction (SPE). Over the last two decades LLE has been progressively replaced by SPE which can be easily automated and adapted to extract a larger amount of contaminants. In the most recent years, the development of online SPE reduced the sampling volume from several liters to a few milliliters (Anumol and Snyder, 2015; Wang and Gardinali, 2012, 2013; Wert et al., 2011). However, with the constant development of instruments with increasing sensitivity, some studies even avoid sample enrichment for wastewater effluent and proceed with direct injection of 80–100 μL sample (Anumol et al., 2015a).

The quantification of DEET is mostly performed by gas chromatography (GC) or liquid chromatography (LC) coupled to mass spectrometry (MS). Currently, LC-MS is more prevalent (Fig. 5). In practice, LC-MS is often associated with electrospray ionization which mostly generates the molecular ion of DEET through hydrogen or sodium adducts (Fig. 6). While several compounds with a similar mass might result in the same mass spectrum, tandem mass spectrometry (MS/MS) is often performed. In this case DEET undergoes collision induced dissociation to generate four major specific product ions, each with a specific relative abundance (Fig. 6). Hence, monitoring two or more of these fragment ions and their relative abundance instead of the molecular ion of DEET decreases the chance of interference. On the contrary, GC-MS with electron impact ionization already generates several fragments of DEET with a specific relative abundance (Fig. 6). Therefore, while most studies relying on GC-MS don't perform tandem mass spectrometry, increasing method specificity by monitoring several of many fragments resulting from collision induced dissociation (Fig. 6) remains possible, particularly with most recent instruments (Merel et al., 2015a).

The direct comparison of different combinations of extraction procedures and analytical techniques is not straightforward. Indeed, the detection limits are highly dependent on the matrix analyzed, the concentration factor applied during the extraction procedure, the volume of extract injected in the instrument, and the model as well as the type of instrument used. For instance, drinking water often provides better detection limit than wastewater effluent due to the inherent lower matrix effect. Similarly, time of flight (TOF or QTOF) mass spectrometers are commonly less sensitive than triple quadrupole (QQQ) mass spectrometers, but this is most likely to be the opposite when comparing a recent TOF instrument with a QQQ instrument that is several years older. Such details and even the limit of detection itself are not always provided in the literature, adding to the difficulty of comparing analytical methods and occurrence data. Nevertheless, a trend can be observed where LLE-GC-MS, initially allowing detection down to 5 ng/L after 10,000 fold enrichment of 1 L sample (Pereira and Hostettler, 1993), is progressively replaced by SPE or online SPE-LC-MS/MS, usually allowing detection down to 0.1 ng/L with less than 2 mL sample (Anumol and Snyder, 2015). Such shift in detection limit and sample volume requirement accurately reflects the progress in the field of analytical chemistry over the last two decades. However, more than a sensitivity difference, the tendency to prefer LC-MS/MS most likely originates from the ease of automation when compared to GC-MS for which LLE or SPE are an absolute requirement (to avoid injecting water) but often without possibility of direct interface.

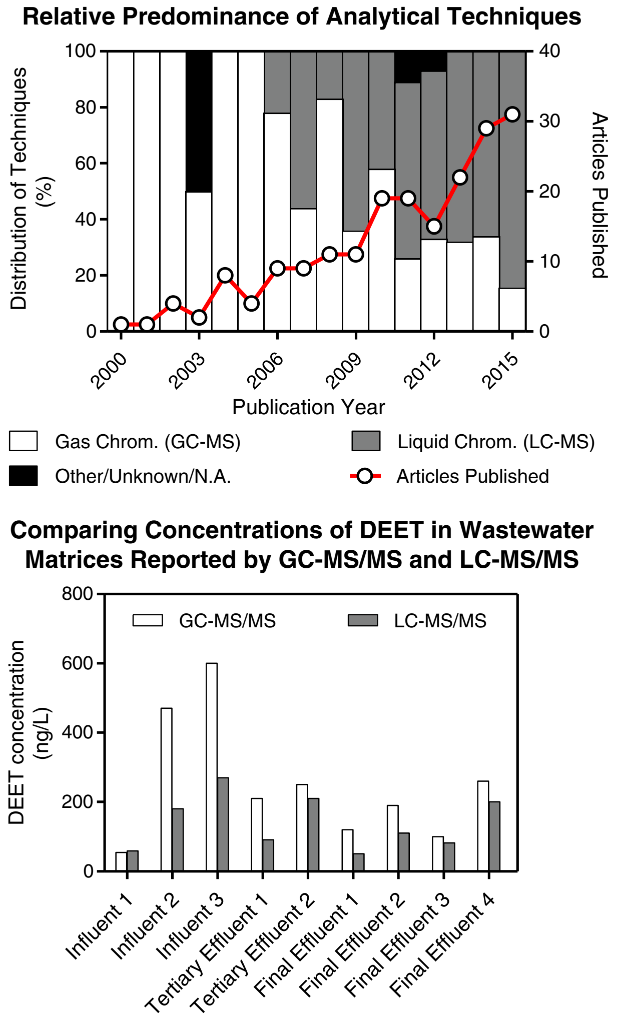


Fig. 5. Relative predominance of two analytical techniques for the analysis of DEET and comparison of the results reported in wastewater samples (Trenholm et al., 2008).

5.3. Potential analytical interferences

The analysis of trace organic contaminants by GC-MS and LC-MS/MS as described previously is known to provide a high compound specificity. However, within the last few years and the ubiquitous detection of DEET in the aqueous environment even in arid areas and during winter period, some concerns have been raised regarding a potential analytical interference. Even though such theory has not been unequivocally confirmed yet, several documented facts might tend to sustain it. A first fact is the large number of studies reporting the detection of DEET in laboratory blanks and field blanks (Anumol et al., 2013; Coes et al., 2014; Fox et al., 2010; Hunt et al., 2010; Katz and Griffin, 2008; Kolpin et al., 2013; Lee and Rasmussen, 2006; Phillips et al., 2015; Stiles et al., 2008;

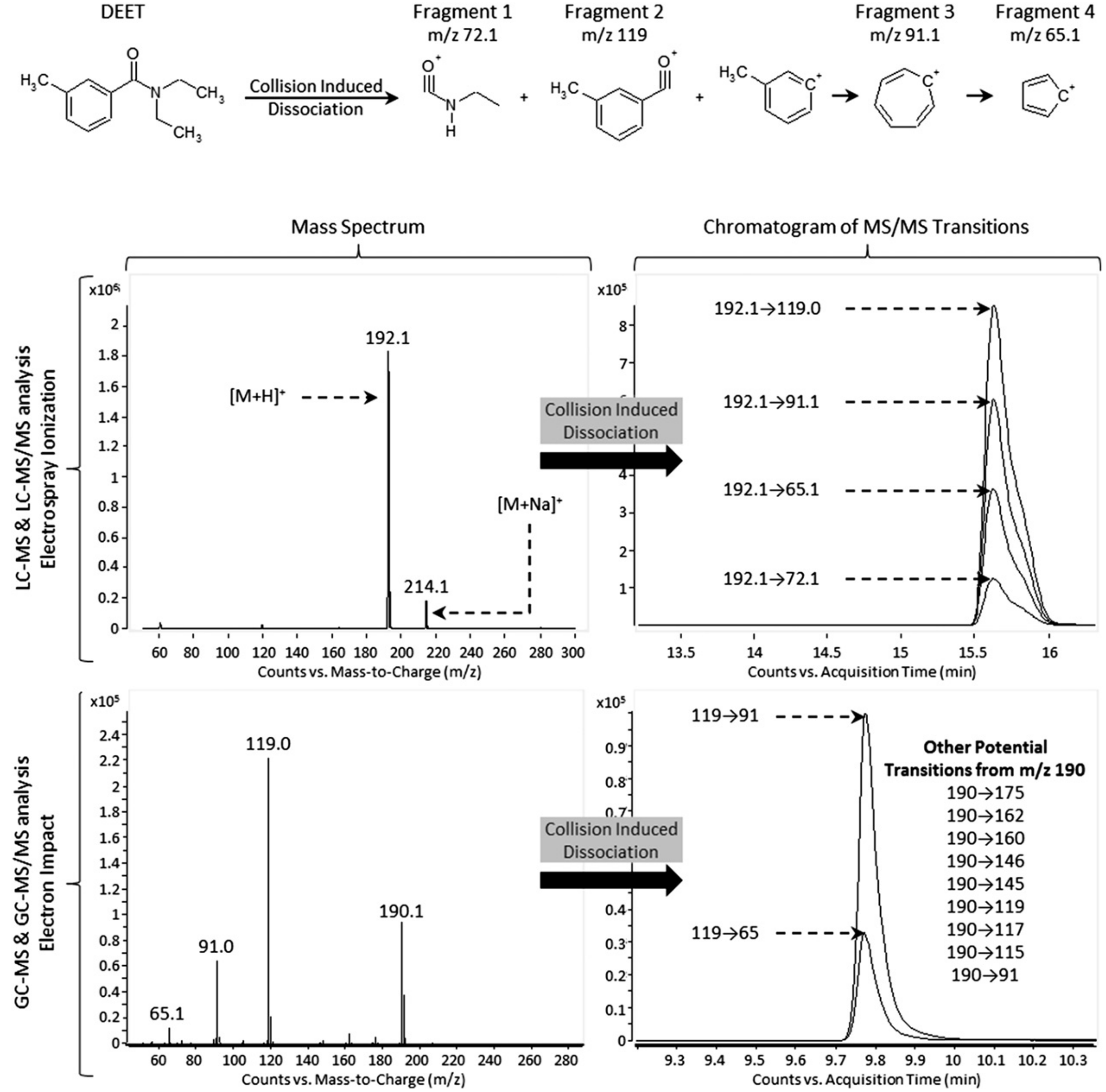


Fig. 6. MS spectrum and MS/MS transitions of DEET in GC-MS and LC-MS (Merel et al., 2015a). Data acquired in the Snyder Research Laboratories at the University of Arizona using an Agilent 7000 GC–MS/MS mass spectrometer, an Agilent 6460 LC–MS/MS mass spectrometer, and a DEET standard procured from Sigma-Aldrich.

Trenholm et al., 2008; Wode et al., 2015; Wu et al., 2014) that could lead to the exclusion of DEET from the study (Ferguson et al., 2013). This observation is confirmed and explained by a recent study showing the occurrence of DEET in LC-MS grade solvents used for sample extraction and analysis, which causes an accumulation of DEET on the analytical column during the equilibration time (Merel et al., 2015a). A second fact is the discrepancy between concentration of DEET reported by GC-MS and LC-MS (Fig. 5) observed in samples from three wastewater treatment plants (Trenholm et al., 2008) and further documented in a recent study (Merel et al., 2015a). Finally, a third fact is the report of a co-eluting compound that prevented the GC-MS quantification of DEET in sewer samples from Norway (Weigel et al., 2004a). This observation was also further confirmed in a recent study demonstrating the potential for two compounds to co-elute with DEET while sharing the same precursor and product ions, therefore inducing a slight overestimation of the concentration of insect repellent in samples analyzed by LC-MS/MS (Merel et al., 2015a).

The concentration of DEET reported in a sample could also be affected without the occurrence of mimic. For instance, LC-MS analysis is well known to suffer from matrix effect, particularly in complex matrix such as wastewater effluent rich in dissolved organic matter. In this case, co-eluting compounds that could be discriminated from DEET by mass spectrometry compete for the charges in electrospray ionization. This phenomenon usually results in the partial ionization of DEET, decreasing proportionally the intensity of the signal and underestimating the concentration of the insect repellent (Merel et al., 2015a; Wickramasekara et al., 2012). In some of the most recent studies this phenomenon is accounted for by spiking a stable isotope of DEET before sample extraction (surrogate) or before sample analysis (internal standard) to assess the loss of signal and correct the concentration of insect repellent accordingly. However, when using such method, the recovery of the stable isotope should always be specified along with the corrected concentration of DEET to allow a critical interpretation by the reader. Indeed, while a high recovery (above 80%) results in a minor and usually acceptable correction, extremely low recovery (below 20%) could result in the overcorrection of DEET concentration and the overestimation of insect repellent in the sample.

6. Overview of DEET occurrence in water samples

A total of 198 studies reporting the occurrence of DEET in water could be found, as already described in the overview of the literature. These reports of DEET occurrence were then examined by discriminating five different global world regions (Africa, America, Asia, Europe and Oceania) and six water matrices (landfill leachate, sewage, wastewater effluent, surface water, ground water and drinking water). In each global world region and water matrix, the occurrence of DEET was characterized according to the detection frequency and the maximum concentrations reported (Fig. 7 and Table 1).

6.1. Magnitude of contamination through detection frequency

The overall detection frequency was calculated only from studies that specified the total number of samples analyzed and those in which the insect repellent was actually detected. Assuming that excluding a few studies that did not provide the relevant information does not influence the overall assessment of DEET detection frequency, it quickly appears that landfill leachate, wastewater influent and wastewater effluent are the most largely contaminated matrices with detection frequency generally higher than 90% in all geographic areas. In America and Europe, the extent of DEET contamination significantly decreases in surface water with a detection frequency close to 60%. For these two geographic areas DEET contamination further decreases in groundwater with a detection frequency lower than 30%. With a very low number of samples analyzed the contamination of drinking water shows a high variability between geographic areas and should not be compared to other water matrices. Another major observation reflected by the overall DEET detection frequency is the larger contamination of surface water and groundwater in Asia compared to other geographic areas, which might characterize a lower ratio of the population connected to a sewer system and a wastewater treatment plant. In addition, this might also reflect a higher consumption of DEET in Asia. Indeed, in Europe, commercial formulations of insect repellent have progressively replaced DEET by other active ingredients (Knepper, 2004). Moreover, Asia is an area at risk of dengue transmission while Europe and North America are not (WHO, 2009). However, even though using DEET and other insect repellents is usually recommended to prevent infections transmitted by mosquitoes (dengue, malaria, yellow fever, etc.), statistics comparing the consumption of insect repellent per capita and per country with the risk of contracting diseases associated with mosquito bites is scarce or inexistent, most likely due to the already mentioned lack of data on sales volume. Such correlation would prove particularly useful for the accurate interpretation of environmental contamination by DEET and other active substances. Moreover such statistical analysis would also result highly valuable for epidemiological studies and the development of prevention campaign, particularly with the severe neurological disorders associated with the recent emergence of the Zika virus in South America.

6.2. Level of contamination through maximal concentrations

The assessment of the level of DEET contamination was challenging since studies report either median, mean or extreme concentration values. Therefore, this paper examines the level of DEET contamination according to the maximum concentrations reported by different studies because it is the only value that is not influenced by the size of the sampling campaign (number of samples collected and timeframe). According to this criterion, landfill leachate appears to be the most highly contaminated water matrix in all geographic areas with concentrations in the μg/L and even reaching higher than 1 mg/L (Clarke et al., 2015). Overall, the level of contamination was the following: landfill leachate ≫ wastewater influent ≥ wastewater effluent > surface water ≥ groundwater ≫ drinking water. Such trend illustrates the partial removal of DEET by wastewater treatment as well as the dilution of the contamination with the discharge of treated effluent in receiving waters. Finally, concentrations seem to further decrease in groundwater and particularly drinking water, showing further DEET removal during infiltration and drinking water treatment. Indeed, in drinking water the maximum concentration of DEET which was reported in USA did

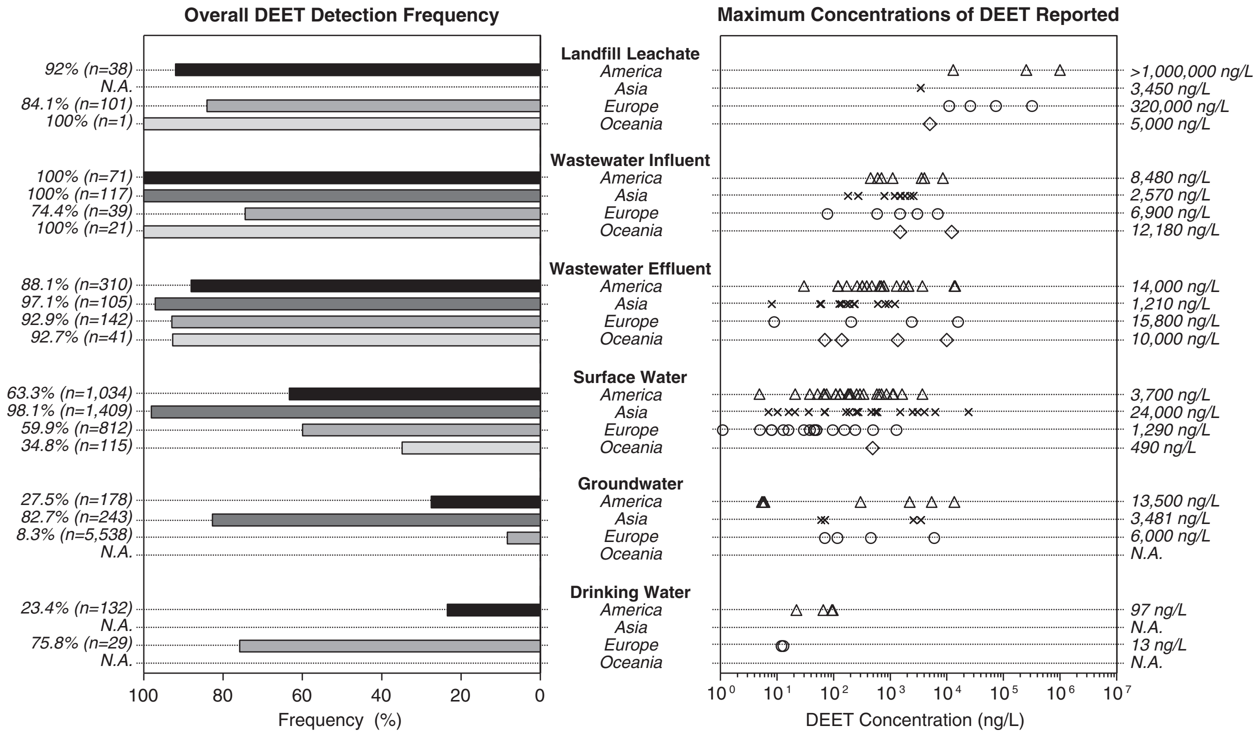


Fig. 7. Overview of DEET detection frequency and maximum concentrations reported according to global world regions and water matrices.

Table 1
Survey of literature on DEET occurrence in different water matrices and geographic areas.

Water type	Geographic area	References found	Overall detection frequency[a]	Highest concentration reported
Landfill leachate	America	4 (A)	92% ($n = 38$)	>1,000,000 ng/L
	Asia	1 (B)	–	3,450 ng/L
	Europe	10 (C)	84.1% ($n = 101$)	320,000 ng/L
	Oceania	1 (D)	100% ($\mathrm{n} = 1$)	5,000 ng/L
Wastewater influent	America	11 (E)	100% ($n = 71$)	8,480 ng/L
	Asia	16 (F)	100% ($n = 117$)	2,570 ng/L
	Europe	9 (G)	74.4% ($n = 39$)	6,900 ng/L
	Oceania	2 (H)	100% ($n = 21$)	12,180 ng/L
Wastewater effluent	America	37 (I)	88.1% ($n = 310$)	14,000 ng/L
	Asia	21 (J)	97.1% ($n = 105$)	1,210 ng/L
	Europe	19 (K)	92.9% ($n = 142$)	15,800 ng/L
	Oceania	8 (L)	92.7% ($n = 41$)	10,000 ng/L
Surface water	America	43 (M)	63.3% ($n = 1{,}034$)	3,700 ng/L
	Asia	23 (N)	98.1% ($n = 1{,}409$)	24,000 ng/L
	Europe	37 (O)	59.9% ($n = 812$)	1,290 ng/L
	Oceania	3 (P)	34.8% ($\mathrm{n} = 115$)	490 ng/L
Groundwater	America	14 (Q)	27.5% ($\mathrm{n} = 178$)	13,500 ng/L
	Asia	4 (R)	82.7% ($\mathrm{n} = 243$)	3,481 ng/L
	Europe	10 (S)	8.3% ($\mathrm{n} = 5{,}538$)	6,000 ng/L
	Oceania	–	–	–
Drinking water	America	5 (T)	23.4% ($n = 132$)	97 ng/L
	Asia	–	–	–
	Europe	3 (U)	75.8% ($\mathrm{n} = 29$)	13 ng/L
	Oceania	–	–	–

A) (Barnes et al., 2004; Clarke et al., 2015; Masoner et al., 2014; Nanny and Ratasuk, 2002).
B) (Wu et al., 2011).
C) (Baderna et al., 2011; Eggen et al., 2010; Jernberg et al., 2013b; Müller et al., 2011; Öman and Hynning, 1993; Paxéus, 2000; Pitarch et al., 2010; Portolés et al., 2011; Schwarzbauer et al., 2002, 2006).
D) (Benbow et al., 2008).
E) (Anumol et al., 2013; Anumol and Snyder, 2015; Bisceglia et al., 2010; Gerrity et al., 2011a; Holloway et al., 2014; Merel et al., 2015a; Snyder et al., 2006, 2007; Trenholm et al., 2006, 2008; Yang et al., 2011).
F) (Chen et al., 2012; Heeb et al., 2012; Nakada et al., 2006, 2007b, 2010; Okuda et al., 2008; Qi et al., 2015; Ryu et al., 2014; Sui et al., 2009, 2010, 2011; Tran et al., 2013a, 2014; Wang et al., 2014; Xue et al., 2010; Zhu and Chen, 2014).
G) (Bernhard et al., 2006; Celano et al., 2014; Knepper, 2004; Margot et al., 2013; Rodil and Moeder, 2008; Rodil et al., 2009, 2012; Smital et al., 2011; Terzić et al., 2008).
H) (Costanzo et al., 2007; Phan et al., 2015).
I) (Anumol et al., 2013, 2015b; Anumol and Snyder, 2015; Barber et al., 2007, 2013; Bartelt-Hunt et al., 2009; Bisceglia et al., 2010; Cordy et al., 2004; Dickenson et al., 2011; Dong et al., 2015; Gerrity et al., 2011a, b, 2012; Glassmeyer et al., 2005; Hope et al., 2012; Karpuzcu et al., 2014; Laws et al., 2011; Lee and Rasmussen, 2006; Loraine and Pettigrove, 2006; Merel et al., 2015b, a; Nanaboina and Korshin, 2010; Nelson et al., 2011; Oppenheimer et al., 2011; Snyder et al., 2006; Trenholm et al., 2006, 2008; Vidal-Dorsch et al., 2013a, b; Wang and Gardinali, 2012, 2013; Wert et al., 2009, 2011; Wu et al., 2014; Yang et al., 2011; Yu et al., 2015; Zenobio et al., 2015).
J) (Alidina et al., 2014; Chen et al., 2014b, 2012; Heeb et al., 2012; Kim et al., 2009a, 2007; Nakada et al., 2006, 2007a, b, 2010; Narumiya et al., 2013; Qi et al., 2015; Ryu et al., 2011, 2014; Sui et al., 2009, 2010, 2011; Trenholm et al., 2006; Wang et al., 2014, 2015b; Zhu and Chen, 2014).
K) (Celano et al., 2014; Dsikowitzky et al., 2004; Gómez et al., 2009, 2010, 2012; Hernández et al., 2015; Hollender et al., 2009; Jernberg et al., 2013a; Knepper, 2004; Loos et al., 2013a; Margot et al., 2013; Pintado-Herrera et al., 2014; Reemtsma et al., 2006; Rodil and Moeder, 2008; Rodil et al., 2009, 2012; Schymanski et al., 2014; Smital et al., 2011; Weigel et al., 2004a).
L) (Allinson et al., 2012; Chapman, 2003; Costanzo et al., 2007; Escher et al., 2013; French et al., 2015; Hawker et al., 2011; Leusch et al., 2014; Reungoat et al., 2010).
M) (Alvarez et al., 2014a, 2005, 2012, 2014b; Anumol et al., 2013; Anumol and Snyder, 2015; Barber et al., 2013; Bargar et al., 2013; Bartelt-Hunt et al., 2009; Benotti et al., 2009a; Bernot et al., 2013; Bidwell et al., 2010; Coes et al., 2014; Corwin and Summers, 2010; De Gerónimo et al., 2014; Dong et al., 2015; Fairbairn et al., 2015; Ferrer and Thurman, 2007; Focazio et al., 2008; Garcia-Ac et al., 2009; Glassmeyer et al., 2005; Haggard et al., 2006; Karpuzcu et al., 2014; Klosterhaus et al., 2013; Kolpin et al., 2002, 2004, 2013; Lee and Rasmussen, 2006; Loraine and Pettigrove, 2006; Merel et al., 2015a; Oppenheimer et al., 2011; Oros et al., 2003; Padhye et al., 2014; Pereira and Hostettler, 1993; Regnery et al., 2015; Sandstrom et al., 2005; Sengupta et al., 2014; Singh et al., 2010; Stackelberg et al., 2004, 2007; Veach and Bernot, 2011; Writer et al., 2010; Zenobio et al., 2015).
N) (Chen et al., 2015, 2014b; Dai et al., 2015; Dsikowitzky et al., 2011, 2014; Hanamoto et al., 2013; Heeb et al., 2012; Kim et al., 2007; Liu et al., 2015; et ala; Nakada et al., 2008; Qi et al., 2014; Ryu et al., 2014; Sun et al., 2015; Tanoue et al., 2015; Tran et al., 2013a, 2014; Wang et al., 2015a, b; Yoon et al., 2010; You et al., 2015; Zhang et al., 2015; Zhu et al., 2013).
O) (Almeida et al., 2014; Calza et al., 2011, 2013; Celano et al., 2014; Deroux et al., 1996; Dsikowitzky et al., 2004, 2015; Franke et al., 1995; Geerdink et al., 1999; Gómez et al., 2009, 2010, 2012; Hendriks et al., 1994; Hernández et al., 2015; Jernberg et al., 2013a; Knepper, 2004; Langford and Thomas, 2008; Loos et al., 2013b; Moschet et al., 2013, 2014; Neale et al., 2015; Pintado-Herrera et al., 2014; Quednow and Püttmann, 2009; Rasmussen et al., 2011; Robles-Molina et al., 2014; Rodil and Moeder, 2008; Rodil et al., 2009, 2012; Ruff et al., 2015; Schwarzbauer and Heim, 2005; Schwarzbauer and Ricking, 2010; Stuart et al., 2014; Weigel et al., 2001, 2002, 2004a, b; Wode et al., 2015).
P) (Costanzo et al., 2007; Kennedy et al., 2012; Smith et al., 2012).
Q) (Anumol et al., 2013; Anumol and Snyder, 2015; Barnes et al., 2008; Del Rosario et al., 2014; Focazio et al., 2008; Fox et al., 2010; Hunt et al., 2010; Katz and Griffin, 2008; Katz et al., 2009; Laws et al., 2011; Phillips et al., 2015; Regnery et al., 2015; Schaider et al., 2014; Stiles et al., 2008).
R) (Kuroda et al., 2012; Nakada et al., 2008; Tran et al., 2013a, 2014).
S) (Almeida et al., 2014; Hernández et al., 2015; Lapworth et al., 2015; Loos et al., 2010; Lopez et al., 2015; Portolés et al., 2014; Stuart et al., 2012, 2014; ter Laak et al., 2012; Wode et al., 2015).
T) (Benotti et al., 2009a; Padhye et al., 2014; Stackelberg et al., 2004, 2007; Stiles et al., 2008).
U) (Almeida et al., 2014; Rodil et al., 2009, 2012).

[a] Overall detection frequency calculated using only publications identifying clearly the number of samples collected and those where DEET was detected.

not exceed 97 ng/L. However, studies in such water matrix are very limited and these low values should be considered with caution.

6.3. Seasonal variation

Seasonal variation in DEET concentration is strongly expected since the insect repellent should be used almost exclusively in the summer period but, as already mentioned, the detection of DEET in the winter period of temperate regions is drawing attention. Among the occurrence studies found, only a few compared both seasons and reported lower concentrations in winter (Bernot et al., 2013; Calza et al., 2013; Hope et al., 2012; Knepper, 2004; Loos et al., 2013b; Nakada et al., 2006; Quednow and Püttmann, 2009; Sui et al., 2011; Yang et al., 2011). However, most of these studies rely on a few sampling dates. A more recent study based on a weekly sampling over a six months period confirms the high seasonal variation in DEET usage by measuring the insect repellent in the wastewater influent, but revealed that such pattern was no longer reflected in the effluent (Merel et al., 2015a). Therefore, while the seasonal variation of DEET concentration is observed by several studies, it does not disprove the hypothesis of a potential interference discussed previously. Indeed, the occurrence of an interfering compound at a constant concentration over the year would still perfectly explain the higher concentration of DEET in summer (DEET + interfering compound) and the lower but yet unexpected concentration of DEET in winter (interfering compound only).

7. Attenuation and fate of DEET in water treatment and rivers

The ubiquitous detection of DEET in the environment is largely attributed to the discharge of wastewater effluent in receiving waters, as explained previously. Therefore, in order to restrict the annual load of insect repellent and lower its concentration in the aqueous environment, the efficacy of wastewater treatment plants must be improved, either through the alteration of operating conditions or through the incorporation of new processes.

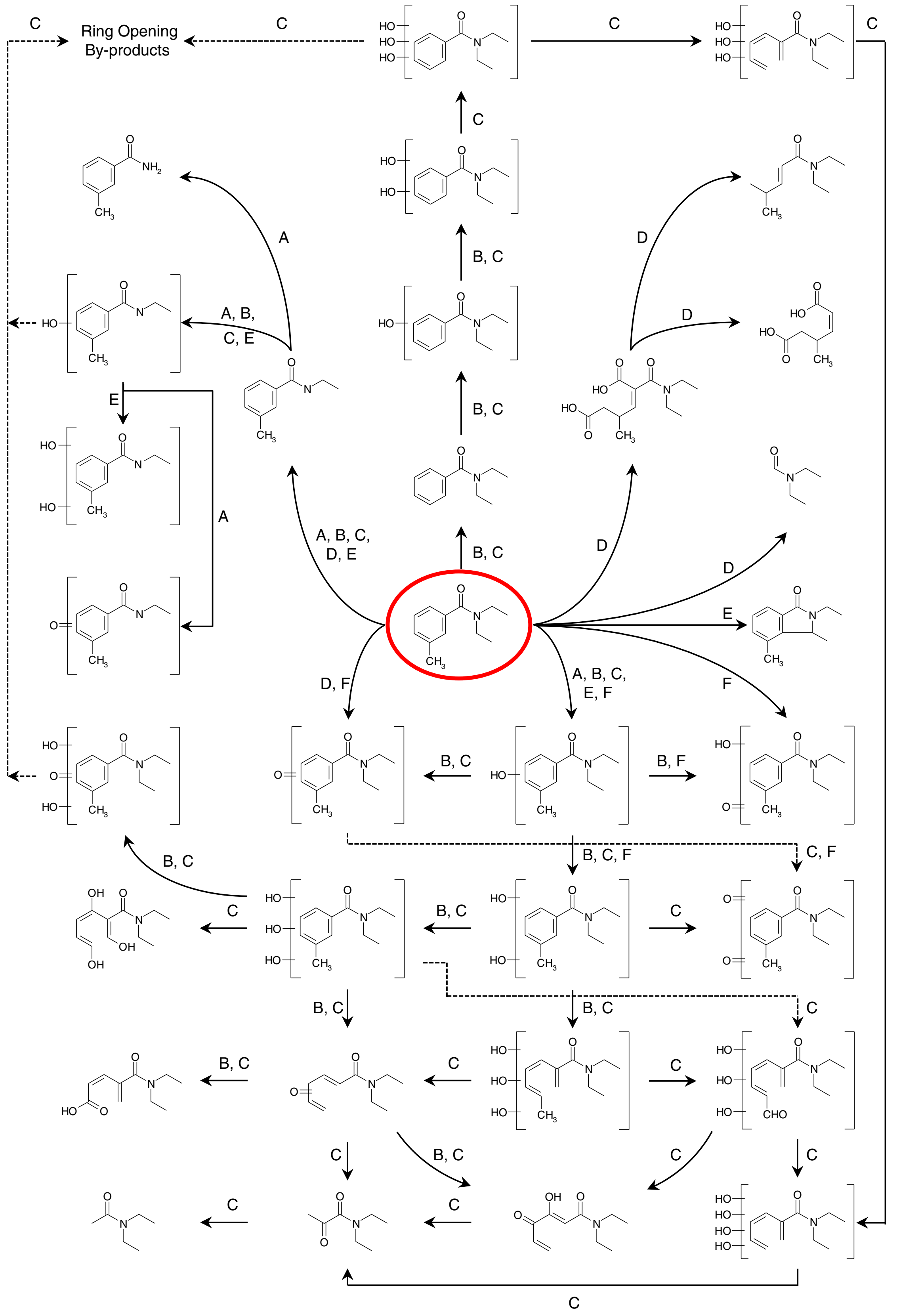
7.1. Attenuation efficiency in wastewater treatment

With over a hundred references, the literature reporting the fate of DEET with different treatment processes is rather abundant and synthesized in Table 2. There seems to be consensus that DEET is not well attenuated by conventional treatments. Several studies showed largely different removal efficiency for wastewater treatment by activated sludge and membrane bioreactor. Indeed the attenuation of DEET was reported in the range 10–90%, with membrane bioreactor providing overall better removal. In fact, some studies showed that the high variation in the attenuation of DEET was a consequence of operating conditions and particularly the sludge retention time (Bernhard et al., 2006; Tadkaew et al., 2011). The attenuation was shown to increase with sludge retention time which should be kept at a minimum of 20 days. Other treatment processes allowing the retention of DEET include membrane filtration and activated carbon. While the rejection of trace organic contaminants always depends on the selection of the membrane, multiple studies agreed on the expected low removal (below 50%) of DEET by ultrafiltration and microfiltration, the high removal (above 80%) by nanofiltration and the very high removal (above 90%)

Table 2
Survey of literature on the fate of DEET in water treatment and river water.

Removal/degradation process	Articles found	References	Overall Impact on DEET
Coagulation	4	A	No removal
Activated sludge	8	B	High removal variation (commonly 0–50% but up to 90%)
Membrane bioreactor (MBR)	12	C	High removal variation (10–90%)
Ultrafiltration (UF)	7	D	Overall, no or low removal (0–50%)
Microfiltration (MF)	3	E	No or low removal (0–25%)
Nanofiltration (NF)	15	F	High to very high removal (80–100%)
Reverse & forward osmosis (RO & FO)	12	G	Very high removal (mostly above 90%)
Powdered activated carbon (PAC)	3	H	Low to very high removal (21–98%)
Granular activated carbon (GAC)	7	I	Low to high removal (15–85%)
Chlorination	4	J	No or very low removal (below 10%)
Ozonation	25	K	Moderate to high and very high removal (50–90%)
UV	8	L	Low removal (0–20%)
O_3/H_2O_2	8	M	Moderate improvement compared to ozonation (10% better)
O_3/UV	2	N	Moderate improvement compared to ozonation and similar to O_3/H_2O_2
UV/H_2O_2	10	O	Major improvement compared to UV alone (up to 90% removal reported)
Fenton oxidation	7	P	High to very high removal (up to 100% removal reported)
TiO_2 photocatalysis	8	Q	Low to very high removal (30–90%)
Soil filtration and aquifer recharge	6	R	Low to very high removal (25–90%)
Other treatment process/combination	26	S	–
Biodegradation/photocatalysis in river	3	T	Low to very high attenuation (20–99%)

A) (Stackelberg et al., 2007; Wert et al., 2011; Westerhoff et al., 2005; Wu et al., 2011).
B) (Bernhard et al., 2006; Hyland et al., 2012; Lee and Rasmussen, 2006; Nakada et al., 2006, 2007b, 2010; Sui et al., 2010; Yang et al., 2011).
C) (Alturki et al., 2012, 2010; Bernhard et al., 2006; Holloway et al., 2014; Monsalvo et al., 2014; Phan et al., 2015; Pisarenko et al., 2012; Snyder et al., 2007; Sui et al., 2011; Tadkaew et al., 2011; Wijekoon et al., 2015; Xue et al., 2010).
D) (Acero et al., 2015; Comerton et al., 2007; Snyder et al., 2007; Sui et al., 2010; Yang et al., 2011; Yoon et al., 2006, 2007).
E) (Escher et al., 2013; Snyder et al., 2007; Sui et al., 2010).
F) (Acero et al., 2015; Alturki et al., 2010; Bellona and Drewes, 2007; Bellona et al., 2010, 2012; Comerton et al., 2007, 2008; Fujioka et al., 2014; Shahmansouri and Bellona, 2013; Simon et al., 2013; Steinle-Darling et al., 2010; Sudhakaran et al., 2013a, b; Yoon et al., 2006, 2007).
G) (Alturki et al., 2010; Bellona et al., 2012; Comerton et al., 2007, 2008; Escher et al., 2013; Fujioka et al., 2015; Holloway et al., 2014; Huang et al., 2011; Snyder et al., 2007; Sudhakaran et al., 2013a, b; Sui et al., 2010).
H) (Margot et al., 2013; Snyder et al., 2007; Westerhoff et al., 2005).
I) (Anumol et al., 2015b; Corwin and Summers, 2010; Rossner et al., 2009; Stackelberg et al., 2007; Sudhakaran et al., 2013a, b; Yang et al., 2011).
J) (Acero et al., 2013; Benotti et al., 2009a; Stackelberg et al., 2007; Westerhoff et al., 2005).
K) (Benitez et al., 2013b, 2009a; Dickenson et al., 2009; Escher et al., 2013; Gerrity et al., 2012; Hollender et al., 2009; Kim et al., 2008; Lee et al., 2012, 2013; Lei and Snyder, 2007; Margot et al., 2013; Masten et al., 2001; Nakada et al., 2007b; Nanaboina and Korshin, 2010; Padhye et al., 2014; Snyder et al., 2006; Sudhakaran et al., 2012, 2013a, b; Sui et al., 2010; Tay et al., 2009; Wert et al., 2009, 2011; Westerhoff et al., 2005; Yang et al., 2011).
L) (Benitez et al., 2013a, b, c; Kim and Tanaka, 2009; Kim et al., 2009a, b, 2008; Merel et al., 2015b).
M) (Benitez et al., 2013b; Dickenson et al., 2009; Gerrity et al., 2011b; Kim et al., 2008; Snyder et al., 2006; Sudhakaran et al., 2012, 2013a, b).
N) (Benitez et al., 2013b; Kim et al., 2008).
O) (Benitez et al., 2013a, b, c; Kim et al., 2009a, b, 2008; Merel et al., 2015a; Sudhakaran et al., 2013a, b; Yu et al., 2015).
P) (Benitez et al., 2013b, c; Li et al., 2012, 2013; Wu et al., 2011; Zhang and Lemley, 2006, 2007).
Q) (Adams and Impellitteri, 2009; Antonopoulou et al., 2013; Antonopoulou and Konstantinou, 2013, 2014; Benotti et al., 2009b; López Fernández et al., 2014; Medana et al., 2011; Westerhoff et al., 2009).
R) (Cordy et al., 2004; Laws et al., 2011; Sudhakaran et al., 2013a, b; Teerlink et al., 2012; Yoon et al., 2013).
S) (Acero et al., 2015; Chapman, 2003; Chen et al., 2014a; Farhat et al., 2015; Garcia-Segura et al., 2015; Gerrity et al., 2011b; Helbling et al., 2010a, b; Huang et al., 2011, 2015; Leblebici et al., 2015; Mena et al., 2015; Narumiya et al., 2013; Okuda et al., 2008; Oulton et al., 2015; Peller et al., 2011; Rattier et al., 2014; Reinhold et al., 2010; Reungoat et al., 2011; Rivera-Cancel et al., 2007; Seo et al., 2005; Song et al., 2009; Stevens-Garmon et al., 2011; Tran et al., 2013b; Wang et al., 2014; Zhu and Chen, 2014).
T) (Calza et al., 2011, 2013; Hanamoto et al., 2013).



A: UV photolysis in ultrapure water (Benitez et al., 2013a)
B: Photolysis in river water (Calza et al., 2011)
C: TiO_2 photocatalysis (Antonopoulou et al., 2013; Medana et al., 2011)
D: Ozonation (Tay et al., 2009)
E: Free radicals (Song et al., 2009)
F: Anodic Fenton process (Zhang and Lemley, 2006)

Fig. 8. Synthesis of DEET transformation products from physicochemical oxidation.

by reverse osmosis. However, the attenuation of DEET by activated carbon was found highly variable. Other treatment processes based on oxidation were also studied. While UV irradiation and chlorination where usually shown to result in a limited attenuation of DEET, ozonation was proven more efficient with an attenuation of DEET in the range 50–90%. The efficiency of ozone varies with the operating conditions and particularly the ratio O_3/DOC which should be kept at a minimum of 1 to 1.5 (Lee et al., 2013; Wert et al., 2009). Combining ozonation with the addition of hydrogen peroxide or UV irradiation usually resulted in the improvement of DEET attenuation by 10% while Fenton oxidation was even more efficient. Other emerging treatment processes relying on titanium dioxide photocatalysis or electrochemistry were also investigated in the literature but the attenuation achieved was highly variable depending on the operating conditions.

7.2. Fate of DEET in the environment

Even though DEET commonly reaches surface water through the discharge of wastewater effluent, only a limited amount of studies have investigated its fate in the environment. According to peer-reviewed literature (Antonopoulou et al., 2015; Calza et al., 2011), DEET tend to undergo photolytic degradation in river water. A recent publication reported the enhanced photolytic transformation of DEET in the presence of dissolved organic matter and simulated solar irradiation, indicating the undirect photolysis of the insect repellent via the formation of dissolved organic matter in excited state. The findings of laboratory photolysis experiments were confirmed in the field, via samples taken from the Po River in Italy (Calza et al., 2011).

Biotransformation is also a potential degradation pathway of DEET in river water. For instance, while biodegradation of DEET by microorganisms from activated sludge has already been reported (Helbling et al., 2010b; Rivera-Cancel et al., 2007), it cannot be neglected that adapted microorganisms downstream of wastewater treatment plants could also biodegrade the insect repellent. In fact, the fungi *C. elegans* and the bacteria *Pseudomonas putida* were identified as microorganisms able to use DEET as carbon source (Rivera-Cancel et al., 2007; Seo et al., 2005). However, further research is still required to demonstrate that biodegradation of DEET does occur in river water.

After being discharged in surface water, DEET can also potentially accumulate in sediments. For instance, at least two peer-reviewed publications have reported the occurrence of DEET in sediments from USA (Wauhob et al., 2007) and Australia (Magnusson et al., 2013). However, sediment contamination will not be further developed since it is not in the scope of this review.

7.3. Transformation products of DEET

The attenuation of DEET by oxidation in water treatment or photocatalysis and biodegradation in rivers results in the formation of multiple transformation products. While these transformation products have been investigated by a limited number of studies (Antonopoulou et al., 2013; Benitez et al., 2013a; Calza et al., 2011, 2013; Medana et al., 2011; Song et al., 2009; Tay et al., 2009; Zhang and Lemley, 2006), the respective physicochemical degradation pathways of DEET are summarized in Fig. 8. With the production of several intermediates, a first possible route of DEET transformation involves a ring demethylation followed by three successive hydroxylations leading to a ring opening. The transformation of DEET could also occur through several hydroxylations and formation of ketone with ring demethylation and ring opening occurring later. Finally, some degradation pathways suggested a direct ring opening or attack on the tertiary amine.

Transformation products resulting from the biodegradation of DEET have also been characterized in three peer-reviewed articles (Helbling et al., 2010b; Rivera-Cancel et al., 2007; Seo et al., 2005) and are summarized in Fig. 9. The soil fungi *C. elegans* was shown to transform DEET via formation of an N-oxide, with or without previous N-dealkylation. Similarly, biodegradation from sludge microorganisms was shown to form the N-dealkylated DEET but without

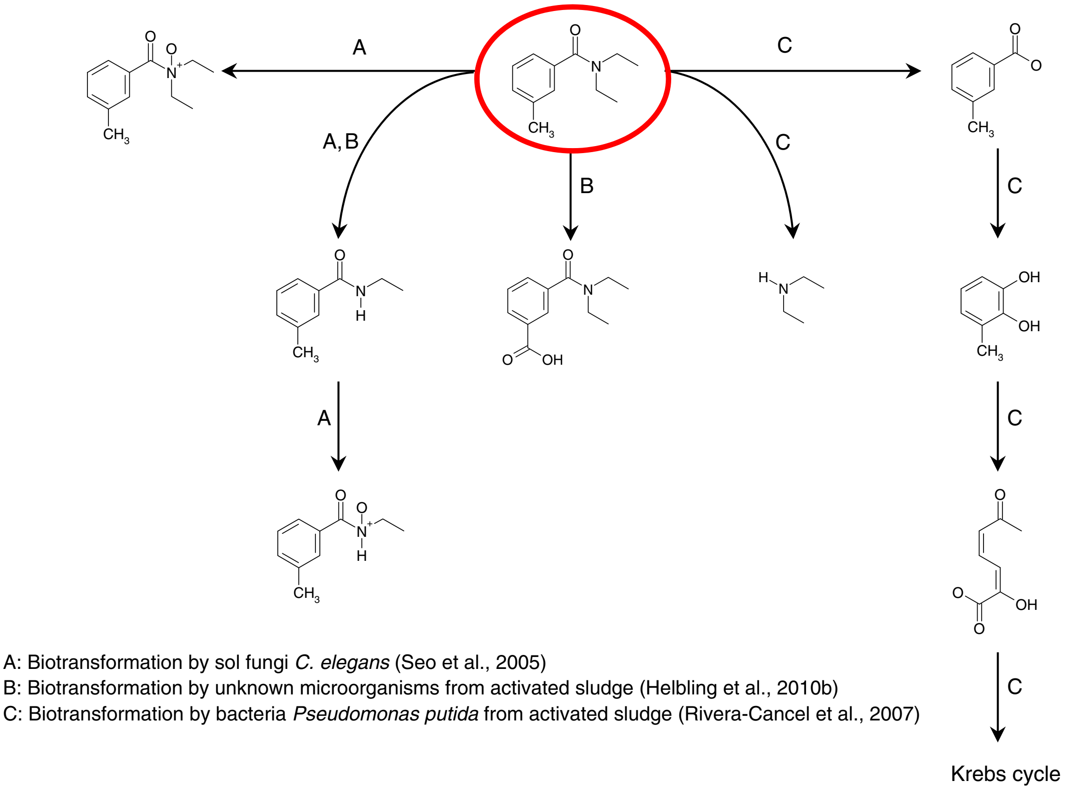


Fig. 9. Synthesis of DEET biotransformation products.

further N-oxide formation. However, another transformation product was identified, which consist in the formation of a carboxylic acid instead of the methyl group of the aromatic ring of DEET (Helbling et al., 2010b). Finally, a third biodegradation pathway of DEET by *Pseudomonas putida* isolated from activated sludge resulted on the formation of diethylamine along with 3-methylbenzoate which undergoes further transformations in order to finally enter the Krebs cycle (Rivera-Cancel et al., 2007).

8. Relevance of DEET ubiquity in the aqueous environment

The occurrence of DEET in the aqueous environment is well documented with nearly all water matrices being affected and concentrations ranging from ng/L to mg/L. Therefore, the relevance of such ubiquitous occurrence should be considered.

The toxicity of DEET to aquatic species still remains poorly assessed in peer-reviewed publications. Nevertheless, acute toxicity studies tend to indicate that DEET would not have significant toxicity at environmental concentration reported in Table 1, with an LC_{50} higher than 100 mg/L for *Daphnia magna*, fathead minnow, rainbow trout and birds (Costanzo et al., 2007). A more recent study also confirmed that deleterious effect of DEET on caddisflies were only observed at levels above environmentally relevant concentrations, but DEET exposure yet decreased carbohydrate contents in *Sericostoma vittatum* (Campos et al., 2016).

The insect repellent DEET has been designed for direct application on the skin and therefore exposure to environmental concentrations should be benign for human health. In fact, the Australian guideline for water recycling suggests a maximum of 2.5 mg/L for DEET (Leusch et al., 2014), a value even higher than the largest concentrations reported in raw sewage. However, no official guideline is available for drinking water, but it is expected that exposure through water consumption is negligible compared to the dermal application of DEET-containing insect repellent. In fact, a peer-reviewed publication (Blanset et al., 2007) confirms this assumption by comparing the 90th percentile of daily drinking water exposure to DEET (17 ng/kg/day) to the average daily exposure of DEET users (10–12 mg/kg/day) which appears to be itself largely superior to the acceptable daily intake (100 μg/kg/day) derived from a NOAEL of 100 mg/kg/day determined after chronic administration to rats and dogs. Therefore, while the occurrence of DEET in drinking water should not be of concern to human health according to toxicological data available to date, further studies are still required to assess the impact of long term exposure to low doses along with a mixture of other trace organic contaminants.

The occurrence of DEET at concentrations typically found in surface water is not likely to be of concern for neither human health nor environmental health, according to the previously mentioned acceptable daily intake. Nevertheless, such assumption should be confirmed through ecotoxicity tests, particularly in fish population already affected by endocrine disruptors downstream of wastewater treatment plants.

9. Conclusion

The insect repellent DEET is largely detected in all types of environmental water matrices, including landfill leachate, wastewater influent and effluent, surface water and groundwater. Moreover, DEET has been reported in America, Europe, Asia, Oceania and more recently in Africa. This environmental contamination mostly results from the low removal of DEET by conventional wastewater treatment plants and the discharge of still concentrated effluents. Furthermore, the amount of peer-reviewed publications detecting the insect repellent in the aqueous environment is growing exponentially. Current occurrence data do not clearly support a seasonal variation in the concentration of DEET in surface water, however, this review draws attention to the potential occurrence of compounds that could mimic DEET and lead to an overestimation of the DEET concentration in water samples. Therefore, the concentrations of DEET reported in multiple studies should be considered carefully and critically according to the following important points: type of analysis (GC-MS vs. LC-MS), detection of DEET in laboratory blanks or field blanks, correction of DEET concentration with a stable isotope, recovery of such labeled isotope.

The upgrade of conventional wastewater treatment with the implementation of advanced treatment processes usually considered to enhance the attenuation of common trace organic contaminants should also result in a better attenuation of DEET. However, with the implementation of such processes, further studies should be performed in order to provide a better characterization of the transformation products of DEET, particularly with respect to their toxicity at low concentration and within a mixture of trace organic contaminants.

Finally, for DEET as well as for other chemicals, the difficulty to obtain data regarding the amount produced or the sales volume is a strong limitation when assessing their environmental fate. Indeed, such information is necessary to perform a mass balance and detect potential inconsistency, particularly for those ubiquitous compounds associated with significant environmental concentrations. Data availability regarding the use of chemicals and their respective sales volume would also significantly improve the identification of water contaminants through non-target screening approaches.

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