
Emerging Contaminants: The Issue, Case Studies, and Bioremediation Options Part II

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Abstract

This second part of the review on “Emerging Contaminants” presents and discusses a series of other four contaminants such as: polybrominated diphenyl ethers (PBDEs), alkylphenols ethoxylates, methyl tert-butyl ether (MTBE), and 1,2-dichloroethane (1,2-DCA). Information related to the properties, uses, production, as well as different criteria that qualifies such contaminants as emerging is provided and discussed. Finally, a brief overview on different emerging technologies (including biotechnologies) available for remediation of persistent organic pollutants will be presented. The aim of these minireviews series is to increase the awareness related to emerging contamination and encourage further studies to develop and apply remedial technologies, especially biotechnological methods.

Keywords: polybrominated diphenyl ethers (PBDEs), alkylphenols ethoxylates, methyl tert-butyl ether (MTBE), and 1,2-dichloroethane (1,2-DCA), sources, production, properties.

Introduction

Technological progress comes at a price. Often, the release of a large variety of pollutants with unexpected and hard to predict impact may seriously affect both human health and the environment. Today, the complexity of overall environmental contamination is reflected by the increasing number of contaminants that are considered emergent. The initial concept associating emergent contamination with new chemicals released in the environment has dramatically changed while our improved understanding and awareness of different factors and conditions that could render a compound hazardous has evolved, along with an increased capacity to detect contaminants at tiny levels. Contamination is often not obvious, but its environmental impact could be serious and irreversible.

This article is the continuation of a previous study sharing information on emerging contaminants of recent and high environmental concern in California and all over the U.S. and worldwide. While in our previous study, following a brief discussion on emerging criteria, information related to four emerging contaminants (1,4-dioxane, N-nitrosodimethylamine, 1,2,3-trichloro propane, and perchlorate) commonly associated with military ranges was presented, this current article will point out some key information related to other four

emerging contaminants: polybrominated diphenyl ethers – PBDEs, alkylphenols etoxylates, MTBE and 1,2-DCA.

Polybrominated Diphenyl Ethers (PBDEs)

Polybrominated diphenyl ethers (PDBEs) – commonly used as flame retardants – represent an example of emerging contaminants that are classified as emerging based on their increased detection in humans. Thus, in 1998, researchers in Sweden reported something alarming: PDBEs have been detected at increasing concentrations in breast milk. Practically, their concentration has doubled every two to five years for the past 25 years. Moreover, due to the current environmental legislation banning the use of PCBs, PDBEs concentration is constantly increasing while PCBs concentration is decreasing. The propensity to accumulate in fat tissues of living organisms, along with their persistence in environment and toxicity classify PDBEs as emerging contaminants and as the possible PCBs of tomorrow. More information on PBDEs can be found on various websites.

At the same time, PBDEs may be consider emerging due to other causes such as toxicological data linking them to liver, brain and thyroid problems in laboratory rodents [1, 2]. Their health effects on humans are still poor understood. Yet, the increasing concentrations in water supplies and human body, among which the highest concentrations may soon trigger the laboratory effects observed so far in animals at levels just slightly higher than the highest human reported levels.

As more and more of the common consumer products containing PBDEs as flame retardant are dumped into landfills, the risk for contaminating the water supplies increases. Luross & al. [3] noted an 100-fold increase in PBDEs in Lake Ontario trout between 1978 and 1998, while recent research by Schecter & al. [4] found levels of PBDEs in U.S. women to be 10-100 times higher than reported levels in European women.

Properties

Similar with PCBs, there are 209 PBDE congeners named in analogy to PCBs. From these, only 7 congeners comprise about 95% of PBDEs. Being similar in chemical structure, they have similar properties with PCBs, so they are highly lipophilic with tendency to bioaccumulate in fat tissues. They are persistent in the environment and their resistance to degradation increases as bromination increases (penta-BDE>>Octa-BDE>Deca-BDE).

There are three common commercial products of PBDEs: penta-BDE, octa-BDE and deca-BDE (deca comprising 80% of the worldwide use). Penta-BDE is the most bioaccumulative form, with the highest production in North America.

Production and Uses

PBDEs are used as flame retardants in a large variety of commercial products including textiles, foam products, lubricants, electrical equipment, building materials and transportation. Examples of common products containing PBDEs are: textiles (clothing, carpets, curtains, tents, interior for offices and public buildings, seating for amphitheatres, stadiums, glass fiber products) and furniture, as well as circuit boards in consumer electronics, like TVs and computers (about 40% of PBDEs are used in the outer casings of computers,

printers and TVs, with the highest PBDE in the mixture as deca-BDE), or electronic switches, relays and molding filers. PBDEs are also connected with transportation, being present in textiles, seat cushion and covers, printed circuit board assemblies, transformers, capacitors and other components of passenger cars, buses, trucks, aircraft, subways and ships. The plastic parts in coffee makers of hair dryers, some medical and laboratory equipment, some building materials (insulation materials, translucent and glass fiber panels, switches, cables, capacitors, etc.), and paints (flame retardant paints and flame retardant joint fillers) also contain PBDEs. Their presence in the housings of printers, fax machines, photocopiers, keyboards and modems is also common.

The global production of PBDEs is over 40,000 tons a year. Great Lakes Chemical Company is the sole U.S. manufacturer that has announced that they will phase out some formulations by 2005.

Alkylphenol Etoxylates

Although chemically different, alkylphenol ethoxylates have similar environmental fate with PBDEs. Thus, chemicals of this group are persistent, bioaccumulative and toxic. Recently, there have been many concerns regarding the environmental safety of alkylphenol ethoxylate surfactants, particularly regarding whether their metabolites are able to mimic hormones. Alkylphenol ethoxylates may be biodegraded to a series of metabolites, such as alkylphenols, that are generally more toxic and tend to accumulate in living organisms and interfere with human hormones. By instance, alkylphenols were first found to be oestrogenic in the 1930s, but the implications of these effects were only highlighted in recent research by Warhurst [5]. Nonylphenols are well-known endocrine disruptors that can make cancer cells proliferate, particularly those cells sensitive to oestrogen. Thus, the growth of cultured human breast cancer cells is affected by nonylphenol at concentrations of only 1 μM (220 ppb), and by octylphenol at 0.1 μM (20 ppb). Oestrogenic effects have also been shown on rainbow trout hepatocytes, chicken embryo fibroblasts and a mouse oestrogen receptor. In an 1995 Report by Warhurst [5], it was concluded that the current environmental levels of alkylphenolic compounds are probably high enough to be affecting the hormonal control systems of some organisms, with possible effect on human health, too. This conclusion was based on the environmental concentrations, bioconcentration factors and *in vitro* oestrogenic effect levels of alkylphenol ethoxylates. Still, little is known on the potential health effects of these compounds. The hormonal effects are best evaluated probably by using cell culture, while more research is needed to establish other potential toxic mechanisms that may involve the immune or the nervous system.

The use patterns of these surfactants, which range from individual household type applications to large-scale industrial processes, result in their discharge to the environment via sewage treatment plants as well as directly from untreated effluents. There is recent alarming data related to their occurrence into indoor air of U.S. homes. Overall, alkylphenol ethoxylates are a group of chemicals that do not degrade adequately in swage treatment, persist in environment, breakdown to more toxic, more persistent and more lipophilic intermediates that bioaccumulate and may have oestrogenic effects at current environmental concentrations. All these are clear traits of emerging contaminants.

Properties

Alkylphenol ethoxylates are non-ionic surfactants, consisting of a branched-chain alkylphenol which has been reacted with ethylene oxide, resulting in an ethoxylate chain. The length of the ethoxylate chain varies between 1 to 50 ethoxy units, depending on their application. Commercial formulations are complex mixtures of homologues, oligomers and isomers. According to White & al. [6], the main commercial alkylphenols are nonylphenol ethoxylates (NPnEO – 80% of the world market) and octylphenol ethoxylates (OPnEO – the remaining of about 20% world market).

Alkylphenol ethoxylates are more or less lipophilic compounds [7], This leads to their bioconcentration into fat tissues. In the higher organisms, the bioconcentration levels may include biomagnification through the food chain, and also uptake of contaminated sediment (with higher levels than the water) [8].

Alkylphenol ethoxylates may be biodegraded by removal of the ethoxy groups, producing less biodegradable products such as: alkylphenol mono- and di-ethoxylates, alkylphenoxy acetic and alkylphenoxy polyethoxy acetic acids, as well as alkylphenols. These metabolites frequently persist through swage treatment and in rivers, while anaerobic conditions generally lead to alkylphenols accumulation. Also, these metabolites of alkylphenol ethoxylates are generally more toxic than the original compounds and accumulate in organisms, with bioconcentration factors varying from ten to several thousands, depending on metabolite, species and organ.

Production and Uses

For over 50 years, they were used in a variety of industrial (including wool washing) and domestic applications [9] as cleaning agents and surfactants.

Their main use today is as industrial surfactants, though they still have some domestic uses, by instance the spermicide nonoxynol-9 according to Soto & al. [10]. They are important to a number of industrial processes including: pulp and paper manufacture, textile manufacture, antioxidants/stabilizers for polymers, emulsion polymers, coatings, agricultural pesticides, lube oils and fuels, laundry detergents, hard surface cleaners, packaging materials, refrigerator lining, thermoplastic polymers, water-based paints, tire and rubber manufacture, household cleaners, adhesives, foam control agents, resins, institutional cleaners (<http://www.aperc.org/productinfo.htm>). Industrial application comprises 55% of market, while industrial and institutional cleaning products comprises about 30%, and household cleaning products 15%, with less than 1% for other uses.

Alkylphenol ethoxylates, in particular nonylphenol and octylphenol ethoxylates, are widely used non-ionic surfactants. Worldwide, the annual production in 1997 was reported to be in excess of 500 ktons [11], with one tenth of this produced in China. The annual production of approximately 200,000 tonnes was recorded in the United States in 1988.

MTBE

MTBE is a gasoline additive used in order to reduce air pollution by contributing to a more complete combustion and improve engine performance. It was first commercially used in gasoline in Italy in 1973. Later on, in 1979 MTBE started to be added to gasoline in the U.S. to replace the lead and prevent knocking (usually between 1-8% by vol.). The use of MTBE as gasoline additive increased in time as a result of more strict air regulations. By instance, the U.S. Clean Air Act of 1990 required the use of oxygenates in reformulated gasoline (2.7% oxygen by weight equivalent to 15% MTBE) [12]. Premium gasoline typically contains more MTBE than the regular one and, in total MTBE accounts for approximately 4%

by vol. of all gasoline in the U.S. [13]. Today, MTBE is used worldwide with an annual consumption of approx. 24 billion liters, the U.S. using approx. 60% of the MTBE produced, Europe about 15%, and the remaining 25% are used in the rest of the world. Apart from its presence in gasoline, MTBE is also present in fuel oil, diesel, kerosene and other middle distillates, due to the transportation of the petroleum products in the same pipelines and trucks [14, 15].

However, leaking petroleum storage tanks with associated piping and spillages have caused MTBE to get into the groundwater where it was detected all over USA and worldwide. The first major contamination event, which brought public attention to MTBE happened in Santa Monica, CA. Thus, in 1996, the city learned that two of its drinking water well fields were contaminated with MTBE at levels as high as 610 ppb, and had to shut down the two wells accounting for 50% of city's drinking water. At present, more than 20 public drinking water wells in California have ceased water production for similar reason.

Once in the groundwater, it takes only little amount of MTBE to render the water unusable for drinking because of the added taste and smell. MTBE increasing detection in water wells, associated with its resistance to biodegradation and offensive taste and odor at low levels, were raising many concerns related to the true benefits of using MTBE in fuels and classify it as emergent contaminant, sparking the national controversy between the need to reduce air pollution and the necessity to safeguard precious water sources from contamination(<http://www.epa.gov/mtbe/faq.htm>).

The health effects of MTBE are only partially understood. Thus, the majority of human health researches were focusing on effects caused by the inhalation of the chemical and have not concluded about the imminent public health threat posed by MTBE-oxygenated gasoline. There is limited data on MTBE effects caused by ingestion and on a long-term basis. EPA's Office of Water has concluded that available data are not adequate to estimate the potential risks of low-exposure to MTBE in drinking water. However, the existing data support the conclusion that MTBE is a potential human carcinogen at high doses. Although it is less toxic than other gasoline constituents, the short and long-term effects of MTBE exposure in humans are not known.

Properties

MTBE is a colorless, flammable organic liquid with a strong taste and smell noticeable at only 15 ppm level. MTBE has a relatively high vapor pressure (so a high volatility) and water solubility. It does not adsorb to soil particles and thus may travel through soil fast and contaminate the groundwater. It is much more soluble in water than most other petroleum constituents and subsequently will travel long distances with groundwater, getting further from the source area than other petroleum compounds. MTBE has a specific gravity lower than 1, thus will float on the water. When exposed to air, MTBE evaporates readily. MTBE, as other gasoline constituents, has a vapor density greater than air, and thus will tend to sink and accumulate in low areas (such as basement or utility trenches) in the absence of pressure or temperature gradients.

MTBE is generally resistant to biodegradation. Yet, there is one well-documented study of natural MTBE biodegradation under iron-reducing conditions (see Landmeyer et al., [16]), but the removal rate, although statistically significant, was very slow and probably limited by the supply of biologically available iron in the aquifer sediment. There is no indication, to date, of MTBE degradation in sediments under sulfate-reducing conditions. In general, the research rather shows MTBE resistance to biodegradation in most environmental

conditions. It is generally considered that the rate of natural bioattenuation of MTBE is much slower than the rate of benzene bioattenuation, in the case the MTBE was shown to degrade at all.

Production and Uses

Since 1979, MTBE was produced for use as gasoline additive (in place of lead) designed to reduce the air pollution, by reducing the carbon monoxide and ozone emissions (increasing burning efficiency) and boosting the Octane ratings of gasoline. It helps gasoline meet the government's clean-air regulations that restrict vehicle emissions, especially in warmer weather climates.

Other minor uses of MTBE include its medical use to dissolve gallstones in humans by injection, with no toxic injuries of the patients reported so far. MTBE has also been used in analytical laboratories as an extractant and to a minor extent, in chemical synthesis (e.g., hydrolyzed to produce tert butyl alcohol - TBA). MTBE can be thermally decomposed into methanol and isobutene and in the past has been used to produce isobutene.

Available statistics show a dramatic increase in MTBE production from the 1980s to the early 1990s and a slower, but steady, increase in production during the 1990s. In the U.S., four states (California, Louisiana, Michigan, and Texas) account for over 50% of the total number of reporting MTBE disposal facilities.

In the U.S., annual MTBE production estimates for 1991 were 9.57 billion pounds (4.34 billion kg), and estimates for 1992 were at 10.86 billion pounds (4.92 billion kg). The estimated production capacity during 1995 was 240,100 barrels (about 62.2 million pounds or 28.2 million kg) per day. Since some companies operate plants that make large contributions to the total annual U.S. production of MTBE, production estimate statistics may exclude figures from certain companies. In fact, such private providers of production information as SRI International no longer provide any MTBE production quantities for facilities or companies. Only limited quantitative estimates on annual import and export levels for MTBE are available for years prior to 1994. For 1994, the level of MTBE imports was 1.753 billion kg (3.865 billion pounds); the level of exports for 1994 was estimated as 0.405 billion kg (0.893 billion pounds). The annual production volume of MTBE in the year 1997 in the EU was 3 030 000 tonnes.

1,2-Dichloroethane (1,2-DCA)

1,2-DCA also referred to as "lead scavenger" (to encourage greater volatility in the lead combustion by-products to be exhausted from the vehicle) was additive of leaded gasoline until the late 1980s when leaded gasoline was phased out. Although it has not been used as additive to gasoline since more than a decade, recent studies indicate the possibility that this compound may persist in the environment and affect drinking water supplies. And, since it has had and still has other uses, the source of lead scavengers in the environment remains unclear. 1,2-DCA has been found increasingly in groundwater and soil near landfills and industries using large chemical quantities. Thus, the risk for contaminating drinking water supplies is high. Moreover, it is present in homes in some cleaning solvents, pesticides, glues, varnishes, and strippers, with the risk to contaminate the indoor-air also high.

Considering its health effects, EPA has found that 1,2-DCA can cause central nervous system disorders, adverse lung, kidney, liver, circulatory and gastrointestinal effects on a short-term exposure. The long-term exposure has the potential to cause cancer from a lifetime exposure at levels above the MCL. Animal studies to date suggest that 1,2-DCA causes stomach, lung, breast and other types of cancer. EPA currently classifies 1,2-DCA as a

“probable human carcinogen”. With a widespread occurrence and persistence in the environment, along with potential carcinogenic effects as well as other effects on human health, 1,2-DCA may certainly be added to the list of emerging contaminants.

Properties

1,2-DCA is a colorless, oily, organic liquid with a sweet, chloroform-like odor, that does not occur naturally in environment. It evaporates very quickly (at normal temperatures) from water and soil releases into the atmosphere, where it is diluted and carried long distances. It may be broken down into less-toxic compounds in the atmosphere. It can leach into groundwater rapidly where it is likely to persist for a very long time. If the compound reaches deeper soils or groundwater where it does not break down, it can travel long distances and can enter drinking water wells. It can be detected by odor in water at about 20 ppm, and in air at about 10-50 ppm.

It is generally resistant to biodegradation, but not expected to accumulate in fish. However, evidence of biodegradation exists and, as in the case of the other emerging contaminants presented, we are still limited by our knowledge in understanding the true potential for biodegradation of such compounds and the right environments in which this may occur. Thus, 1,2-DCA biodegradation was successfully put in evidence by carbon isotope fractionation among substrate, inorganic carbon, and biomass during an aerobic mineralization by *Xanthobacter autotrophicus* according to Hunkeler and Aravena [17]. The stimulated anaerobic biodegradation of both 1,2-DCA and MTBE was also demonstrated by Kinder Morgan and ARCADIS at a field scale, at a large storage terminal along the Houston Ship Channel, with the greatest reduction of 1,2-DCA recorded after 124 days and a complete biodegradation after 190 days.

Production and Uses

1,2-DCA was used as gasoline additive being a lead scavenger until late 1980s when leaded gasoline was phased out. It was also used as an industrial solvent.

Currently, one of the main use is to make vinyl chloride monomer and in the production of chlorinated solvents such as TCE, PCE, TCA, and vinylidene chloride. These chemicals are involved in plastics, rubber and synthetic textile fibers. Other uses include: as a solvent for resins and fats, photography, photocopying, drugs, cosmetics, as well as a fumigant for grains and orchards

(http://www.epa.gov/safewater/contaminants/dw_contamfs/12-dichl.html).

The production of 1,2-DCA was 18 billion lbs. in 1993. It is generally produced in volumes much greater than most other chlorinated compounds. According to the Toxic Release Inventory, from 1987 to 1993, releases to water and land totaled over 455,000 lbs., with the largest releases observed in New Jersey and Louisiana.

Emerging technologies for removal from contaminated environments of Persistent Organic Pollutants (POPs)

The emerging technologies for treatment of environments contaminated with Persistent Organic Pollutants are classified in biological, physico-chemical and thermal technologies.

According Eduardo Gonzalez-Valencial & al. [18] a classification of established and innovative and emerging remediation technologies is as follow:

Table 1. Some criteria to assess established remediation technologies

Remediation technology	Overall cost* (USD/ton)	Cean-up time	Reliability and maintenance (level)
Physico-chemical			
Landfill cap system (in or ex)	N.A.	-	Varies
Vapour extraction (in)	S	M to L	Average
Vapour extraction (ex)	S	M to L	High r. and Low m.
Thermal technologies			
Combustion systems (ex)	M to L	S to M	Average
Thermal desorption (in or ex)	S to M	S to M	Average
Pyrolysis (ex)	M to L	S	Average
Biological technologies			
Bioventing (in)	M	M to L	Low r. and High m.
Composting (ex)	M		Average
Biopiles (ex)	S	S	Average
Land farming (ex)	S	M to L	High r. and Low m.

Table 2. Some criteria to assess innovative and emerging remediation technologies

Remediation technology	Overall cost* (USD/ton)	Cean-up time	Reliability and maintenance (level)
Physico-chemical			
Base catalyzed dechlorination (ex)	M to L	S	Average
Electrochemical oxidation (in)	M to L	S to M	Low r. and High m.
Solvent extraction (ex)	M to L	S to M	Average
Solvated electron (ex)	L	S	Average
Supercritical water oxidation (ex)	S to M	S	Average
Solar detoxification (ex)	N.A.	S to M	N.A.
Gas phase chemical reduction (ex)	L	S to M	High m
Catalytic hydrogenation (ex)	N.A.	S	High m and.Low r.
Thermal technologies			
T.D. – Catalyzed dehalog. (ex)	M to L	S	Low r. and High m.
T.D. – Pyrolysis (ex)	M to L	S to M	Average
Plasma ARC Systems (ex)	L	S	Average
Vitrification (in or ex)	M to L	S	High m and.Low r.
Biological technologies			
Bioslurry (ex)	M	S to M	Average
Enhanced bioremediation (in)	S	L	Low r. and High m.

Legend: (in) = in situ (ex) = ex situ; S = short term < 6 months, M = medium, 6 to 12 months, L = long, > 12 months.

*Cost (USD): S = less than \$150 to \$300, L = more than \$300;

r. = reliability; m. = maintenance. T.D. = thermal desorption. N.A. = not available.

To select the most proper technology several ratable and non-ratable criteria should be considered. Among non-ratable or relative criteria, are included public acceptability, risk and environmental impacts, which depend on the specific geographic site location. The ratable criteria may include the applicability of the method (in accordance with its development status), overall cost, minimum achievable concentration, clean-up, time required, reliability maintenance, post treatment cost and ability to use soil after treatment. Also, social, environmental, technical and economical criteria should be considered during the technology selection process. The applicability and availability of different treatment technologies depends on the location of treatment systems and whether the waste can be transported to treatment facility. In case of choosing more than one technology to treat a specific waste or soil, the limitations, impacts and risks due to the combined methods should be considered.

Conclusions

Emerging contaminants are a wide range of chemicals introduced into the environment more or less recently, with an increased detection in water wells, soils and air and with high potential for serious human health effects (including carcinogenic effects) on a long-term exposure basis. They are usually very mobile and persistent, and some could bioaccumulate to a high extent. They are generally resistant to biodegradation, and some may degrade into even more toxic metabolites. Whatever the case, most of these contaminants lack the appropriate regulation and represent a current and continuous threat to the environment and human society.

The current article is presenting four emerging contaminants along with an overview of emerging remediation techniques that may be applicable for these contaminants. The goal is to increase the awareness and point out the need for future research to better understand the environmental fate of such compounds. Two of the presented contaminants PBDEs and alkylphenols ethoxylates are emerging mainly due to their persistence, bioaccumulation and toxicity characteristics. Their detection in human bodies is increasing while their long-term health effects remain unpredictable, with potential for cancer and endocrine disruption (in the case of alkylphenol ethoxylates). The other two emerging contaminants discussed here, MTBE and 1,2-DCA, are associated with fuel additives and are highly mobile once released in the environment, being also resistant to biodegradation. They easily reach the groundwater and may travel long distances without degrading. They may also be carcinogenic on a long-term exposure basis.

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