LEARNING TOXICOLOGY
THROUGH OPEN EDUCATIONAL

PERSISTENT ORGANIC POLLUTANTS INTRODUCTION

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INTRODUCTION

People living nowadays inhabited a world where persistent organic pollutants (POPs) such as DDT, Dieldrin, PCP, HCH, PAH, etc., produced and used in agriculture and industry during 1920-1950s, are stil present everywhere, including the tissues of living organisms on Earth, presenting harmful effects for humans and environment.

POPs have been found on every continent and in every major climatic zone, including the world's most remote regions, such as the open ocean and deserts, and in every wildlife species and humans (Ritter et al. 1997; AMAP, 2014). POPs are chemical substances that persist in the environment, bio-accumulate, and pose a risk of causing adverse effects to human health and/or the environment. These pollutants are transported across international boundaries, far from their sources, even to regions where they have never been produced or used. The ecosystems and indigenous people of the Arctic are particularly at risk because of the long-range environmental transportation and biomagnification of these substances. The Baltic and the Alpine regions are also cited as examples of EU sinks of POPs.POPs pose a threat to the environment and/or to human health all over the globe. International action is necessary to reduce and eliminate production, use, releases of these compounds.

POPs are both natural and anthropogenic, halogenated organic compounds resistant to chemical, photolytic and biological degradation, characteristics leading to their accumulation in terrestrial and aquatic ecosystems. Humans can be exposed to POPs through: diet, occupational accidents and the environment (including indoor). Exposure to POPs, either acute or chronic, can be associated with a wide range of adverse health effects, including illness and death. Some of POPs such as PCBs), may persist in the environment for

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periods of years and bioconcentrate by factors of up to 70,000 (Ritter et al. 1997; Stockholm 2007; AMAP, 2014; WHO 2010; WHO, 2016).

PROPERTIES AND ENVIRONMENTAL BEHAVIOUR OF POPs

The behaviour of chemicals/POPs in the environment is determined by:

- Their chemical structure, chemical/physical properties, nature of the environment.
- Organic compounds which have very low persistence, low toxicity and immobilty present no risk to the environment and to human health.
- *POPs are organic compounds that are persistent, mobile and toxic*.
- Relatively few substances possess properties corresponding to POPs.
- *The Persistence* is the length of time the compound will remain in the environment before being broken down or degraded into other, less hazardous substances.
- Very persistent in environment are POPs with half-lives greater than 6 months.
- *Dissipation* is a combination of at least two processes, degradation and mobility, and represents the disappearance of a substance from environment.
- *Semi-volatility* is an important property of POPs which confers a degree of mobility through the atmosphere, sufficient to allow relatively great amounts to enter the atmosphere and be transported over long distances.
- Semi-volatile substances are usually highly halogenated, have a molecular weight of 200 to 500 and a vapour pressure lower than 1000 Pa. These substances may volatilize from hot regions and condense and tend to remain in colder regions.
- *Lipophilicity* is tendency of POPs to preferentially dissolve in fats rather than water

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- High lipophilicity of POPs determine their bioconcentration from the surrounding medium into the living organisms. Combined with environmental persistence and the resistance to biological degradation, lipophilicity also results in biomagnification through the food chain (Ritter et al.1997; Stockholm 2007; UNEP, 2010; Ding,2013).

CHEMISTRY AND TOXICOLOGY OF POPs

CHEMISTRY

- By definition, POPs are organic compounds, often halogenated/chlorinated.
- POPs are highly resistant to biological, photolytic or chemical degradation.
- The carbon-chlorine bond is very stable towards hydrolysis and, the greater the number of chlorine substitutions and/or functional groups, the greater the resistance to biological and photolytic degradation. Chlorine attached to an aromatic (benzene) ring is more stable to hydrolysis than chlorine in aliphatic structures.
- POPs with high degree of halogenation, have very low water solubility and high lipid solubility and can to pass readily through the phospholipid structure of the biological membranes and accumulate in fat deposits.
- Halogenated hydrocarbons are a major group of POPs and organochlorines are the most important group (dioxins, furans, PCBs, hexachlorobenzene, mirex, toxaphene, heptachlor, chlordane and DDT).
- POPs are are characterized by low water solubility, high lipid solubility and environmental persistence, long half-lives and their potential to bioaccumulate and biomagnify in organisms once dispersed into the environment.
- Although some natural sources (from bacteria, fungi, plants, marine organisms, insects, etc.) of organochlorines are known to exist, most POPs originate almost entirely from *anthropogenic sources* associated

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with the manufacture and use of organic chemicals. In contrast, HCB, dioxins and furans *formed unintentionally* in a wide range of manufacturing and combustion processes.

- *POPs are typically semi-volatile compounds*, characteristic that favours their long-range transport over great distances through the atmosphere and the resistance to biological and photolytic degradation.Volatilisation may occur from plant and soil surfaces by application of POPs as pesticides.
- Halogenated /chlorinated organic compounds have been utilized by the chemical industry in production of millions of tonnes per year of PVC, solvents, pesticides) and speciality chemicals and pharmaceuticals.
- In addition, both anthropogenic and non-anthropogenic sources also lead to production of undesirable by-products and emissions, characterized by their *persistence and resistance to breakdown* (such as chlorinated dioxins, furans, etc).
- Due to their physicochemical properties POPs can be transformed in environment in a a variety of microbial, chemical, photochemical processes. The efficiency of these environmental processes are largely dependent on the properties of the specific compound and characteristics of the environment.
- Cyclic, aromatic, cyclodiene-type chlorinated hydrocarbon compounds, such as some chlorinated pesticides (DDT, chlordane, lindane, heptachlor, dieldrin, aldrin, etc.), with molecular weights greater than 236 g/mol have the ability to accumulate in biological tissues and are known for their persistence in the environment.
- The lower molecular weight chlorinated hydrocarbons (less than 236 g/mol) may include alkanes and alkenes (dichloromethane,chloropicrin, chloroform) and are often associated with little acute toxicity, reversible toxicological effects and relatively short environmental and biological half-lives.

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- *Bioavailability* (the proportion of the total concentration of a chemical available for uptake by a particular organism), is controlled by a combination of chemical properties of the compound including the ambient environment and morphological, biochemical and physiological properties of organism.
- Generally, excretion of organic pollutants is facilitated through the metabolic conversion to more polar compounds. Because of their resistance to degradation and breakdown, the POPs are not easily excreted and those pollutants (e.g. toxaphene, PCBs etc.) most resistant to metabolism tend to accumulate in organisms and through the food chain. Some organic pollutants may be converted to more persistent metabolites (for exemple, the metabolic conversion of DDT to DDE and of aldrin to its persistent metabolite dieldrin). (Ritter et al. 1997; Ding et al. 2013; WHO, 2010).

TOXICOLOGY

Environment

- POPs have been associated with significant environmental impact in a wide range of species and at virtually all trophic levels. Acute effects of POPs intoxication have been well examined and adverse effects were associated with chronic low level exposure in the environment.
- The long biological half life of POPs in living organisms facilitate accumulation of small unit concentrations over extended periods of time. For some POPs, there is experimental evidence that cumulative low level exposures may be associated with chronic non-lethal effects (immunotoxicity, dermal effects and carcinogenicity).
- Studies demonstrated that immune dysfunction is cause for increased mortality among marine mammals and also demonstrated that consumption of POPs contaminated diets may lead to thyroid deficiencies and susceptibility to microbial infections and reproductive disorders in marine mamals.

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- Immunodeficiency has been induced in a variety of wildlife species by POPs (TCDD's, PCBs, chlordane, HCB, toxaphene, DDT).
- Wildlife, with reported high incidence of tumours contained high concentrations of PCBs mirex, chlordane and toxaphene (AMAP, 2014; WHO, 2010; WHO, 2016).

Human health

- Scientific studies suggests that some POPs have the potential to cause significant adverse effects to human health, at the local, regional and global levels through long-range transport (LRT).
- For some POPs, occupational and accidental high-level exposure presents great risk for workers, especially in developing countries where the use of POPs in agriculture resulted in a large number of deaths.
- For example, a study in the Philippines showed that in 1990, endosulfan became the number one cause of pesticide-related acute poisoning among farmers.
- Worker exposure to POPs during waste management is a significant source of occupational risk in many countries. Short-term exposure to high concentrations of certain POPs results in illness and death.
- Occupational, bystander and near-field exposure to toxic chemicals is often difficult to minimize, especially in developing countries.
- Problems in managing workplace exposure are caused by lack of training and of safety equipment and substandard working conditions.
- Earliest reports of exposure to POPs related to human health impact include an episode of HCB poisoning of food in south-east Turkey, resulting in the death of 90% of those affected and, in other exposure, related hepatic cirrhosis, porphyria and neurological disorders.
- Acute incident in Italy in 1976, release of 2,3,7,8-TCDD to the environment resulted in an increase of chloracne.
- The US EPA is currently reviewing dioxin related health effects especially for the non-carcinogenic endpoints (immunotoxicity, reproductive disorders and neurotoxicity).

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- Such effects are not common in the case of exposure to lower concentrations derived from the environment and the food chain.
- Laboratory and field observations on animals, as well as clinical /epidemiological studies in humans, and studies on cell cultures, demonstrated that overexposure to certain POPs may be associated with a wide range of biological effects.
- These adverse effects may include immune dysfunction, neurological deficits, reproductive anomalies, behavioural abnormalities and carcinogenesis.
- The scientific evidence demonstrating a link between chronic exposure to sublethal concentrations of POPs (which occur as a result of long-range transport) and human health impacts is difficult to establish.
- Studies reported the dietary intake of PCBs, dioxins and furans can be linked to reductions in of lymphocytes, and suggested that children with POPs dietary intake present rates of infection 10-15 times higher than children with lower levels.
- The developing fetus and neonate are particularly vulnerable to POPs exposure due to transplacental / lactational transfer of maternal burdens.
- Residents of the Canadian Arctic having PCB levels in excess of the acceptable daily intake, are at special risk for reproductive and developmental effects.
- Carcinogenesis studies associated with occupational exposure to 2, 3, 7, 8-TCDD indicate that high-level exposures of human populations increase cancer incidence.
- Laboratory studies provide evidence that selected organochlorine chemicals (dioxins and furans) may have carcinogenic effects and act as strong tumour promoters.(Ritter et al. 1997; Dewailly et al. 2000; UNEP, 2010; WHO, 2016).

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ENVIRONMENTAL TRANSPORT of POPs

- Persistence can be reduced by environmental transformation processes: (a) biotransformation; (b) abiotic oxidation; (c) hydrolysis and photolysis.
- The importance of these processes depends on the rates at which they occur under natural environmental conditions.These rates are dependent on the chemical structure and properties of the substance and its distribution in environment.
- Environmental factors have little effect on the breakdown/transformation of POPs.
- Factors that might have some effect are less effective in polar regions. Given the continued use and release of POPs in other parts of the globe, the result of this is a net accumulation of POPs in the polar regions. Some of the physical properties are strongly dependent on environmental conditions.
- Temperature affects vapour pressure, water solubility, Henry's law constant.
- The net exchange direction for substances in the open ocean also reflects differences in surface water temperature and atmospheric concentration.
- For example, net movement of POPs in the Bay of Bengal in the Indian Ocean is from the ocean to the atmosphere, while that in polar regions is the reverse
- Temperature may also affect deposition in other locations. The distribution of POPs is inversely related to vapour pressure, and thus to temperature.
- Lower temperatures favour greater partitioning of these compounds from the vapour phase to particles suspended in the atmosphere, increasing their removal and transport to the surface of the earth by rain and snow.
- Countries in the tropics have higher year-round temperatures than countries in the temperate and polar regions of the globe.

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- The practice of using pesticides in tropical agriculture during the warmer wetter growing season may facilitate the rapid dissipation of POPs through air and water.
- These and other observations suggest that inputs of POPs to tropical coastal water through river discharge are less significant than in temperate zones.
- The residence time in the tropical aquatic environment is short and transfer to the atmosphere is greater in these areas. The short residence time of POPs in the tropical water can be considered as favourable for local organisms, but have implications for the global environment, because volatilized residues from the tropics disperse through global atmosphere.
- The distribution of POPs in the oceans is correlated with a major change in distribution during the last decades. Until 1980s, there were higher concentrations of POPs (DDT, PCBs in the oceans of the northern hemisphere, due to the large usage in developed countries (Japan, Europe, and North America).This distribution has not been seen in the recent samples.
- Atmospheric transport and accumulation of POPs (PCBs, DDT, HCHs, chlordane) in the polar regions has been extensively documented. Accumulation in polar regions is partly the result of global distillation followed by cold condensation of compounds within the volatility range of PCBs and pesticides.
- The contaminants are continually deposited and reevaporated and fractionate according to their volatilities. The result is relatively rapid transport and deposition of POPs having intermediate volatility (such as HCB), and slower migration of less volatile substances (such as DDT).
- The characteristics of polar ecosystems intensify the contamination with POPs. The colder climate, reduced biological activity and relatively small incidence of sunlight increase the persistence of the POPs.

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- Inspection of data regarding concentrations of POPs in samples from Arctic and the Antarctic regions showed declines in concentrations since some of these POPs were banned or restricted. The maintenance of a central database of all analytical data on the POPs would greatly aid in determining spatial and temporal trends in the data and linking these to changes in use pattern of POPs (EMEP, 2008; EMEP, 2014; AMAP, 2014).

THE 12 INITIAL POPS UNDER THE STOCKHOLM CONVENTION

The Stockholm Convention on Persistent Organic Pollutants (May 2001) focuses on reducing and eliminating releases of 12 POPs (named the "Dirty Dozen"by the United Nations Environment Programme (UNEP). These 12 chemicals include:

- eight pesticides (aldrin, chlordane, DDT, dieldrin, endrin, heptachlor, mirex, toxaphene)
- two industrial chemicals (polychlorinated biphenyls (PCBs) and hexachloro-benzene (HCB);
- two unintended by-products, dioxins (PCDD) and furans (PCDF).

The 12 POPs are used in or arise from industry, agriculture and disease vector control.

In 1970s, all of 9 pesticides and PCBs had been banned or subjected to severe use restrictions in many countries. Some of these POPs are still in use in parts of the world where they are considered as essential for ensuring public health

In order to further reduce their use in these countries, it is important to understand what countries are using POPs, and how they are applied POPs.

Aldrin

Aldrin is an pesticide applied to soils to kill termites, grasshoppers, corn rootworm, and other insect pests. It is also used to protect wooden structures from termites. Aldrin readily converts to dieldrin in plants and animals and,

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therefore, residues of these chemicals are usually found in small amounts. Aldrin has low toxicity to plants, but has adverse effects on aquatic invertebrates, particularly insects. Acute exposure to aldrin has caused death in aquatic animals, birds, fish and humans. The International Programme on Chemical Safety (IPCS) estimates the fatal dose for humans to be 5 grams. The most common human exposure pathway is via food, particularly dairy products and meat. Aldrin has been banned or severely restricted in many countries (UNEP, 2010; WHO, 2010).

Chlordane

Chlordane is a broad-spectrum agricultural insecticide used on crops (vegetables, grains, potatoes, fruits, cotton, etc). It is also used to combat termites. Chlordane remains in the soil for a long time (half-life of one year). Chlordane can kill aquatic invertebrates, fish and birds. Chlordane is suspected of damaging the human immune system and is listed as a possible human carcinogen. The most common human exposure pathway is through the air, particularly from indoor environments. Chlordane has been detected in the indoor air of residences (US, Japan) and has been banned in many countries (WHO, 2016).

DDT

DDT was widely used during World War II to protect soldiers and civilians from malaria, typhus, and other diseases spread by insects. After the war, DDT continued to be used to control diseases (malaria, etc.), and sprayed on agricultural crops, especially cotton. Due to its stability and persistence (over 50% can remain in the soil 10-15 years after application), residues of DDT have been detected in the Arctic. Food contaminated with DDT is the greatest source of exposure for population. The long-term exposures have been associated with chronic health effects (immune system depression and estrogen alterations). DDT is listed as possible human carcinogen. Although its use had been banned in many countries, DDT has been detected in food and

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breast milk all over the world. To date, 34 countries have banned DDT and 34 have severely restricted its use (Stockholm Convention 2007; EMEP, 2008; UNEP, 2010; WHO, 2010).

Dieldrin

Dieldrin was used to control termites, insect-borne diseases and insects living in agricultural soils. Its half-life in soil is 5 years. Aldrin rapidly converts to dieldrin, so concentrations of dieldrin in the environment are higher than dieldrin use indicate.Dieldrin is highly toxic to fish and other aquatic animals and affects the human immune system. Dieldrin residues have been found in air, water, soil, fish, birds, and mammals, including humans, exposed to dieldrin through food (mainly dairy products and animal meats). Dieldrin was the second most common pesticide detected in a US survey of pasteurized milk (UNEP, 2010, WHO, 2016).

Endrin

Endrin is an insecticide used against pests of cotton, rice and corn. It has also been used as a rodenticide against mices and rats. Animals can metabolize endrin, which does not accumulate in their fatty tissue as other POPs. Endrin is toxic to fish and other aquatic organisms and has a long half-life, persisting in soil 12 years. Endrin is suspected of suppressing the human immune system. As many of other POPs, humans are exposed to endrin through food, although intake levels are usually very low (Stockholm Convention 2007; UNEP, 2010).

Heptachlor

Heptachlor is an insecticide used to combat soil and crop pests (particularly of cotton), termites, grasshoppers, ants, and mosquitoes (to control malaria). Heptachlor is metabolized to heptachlor epoxide, which has a similar toxicity level to heptachlor. Residues have been detected in the blood of cattle from the U.S. and Australia. Heptachlor is toxic to wildlife even at low concentrations. In birds (from Canada and U.S.), exposure to heptachlor induced behavioural

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changes, reduced reproducibility and increased mortality. Food is the major source of exposure for humans. Heptachlor is listed as a possible human carcinogen. Several countries have banned or restricted the use of heptachlor (Stockholm Convention 2007; EMEP, 2008; WHO, 2010).

Mirex

The insecticide Mirex was used against ants, termites (U.S, South America, Africa).

- Mirex is also used as a fire retardant in plastics, rubber, paint and electrical goods.
- Mirex is one of the most stable and persistent POPs with a half life up to 10 years.
- Mirex is toxic to plants, aquatic organisms (crustaceans, fish) and birds.
- Humans are exposed to mirex through their diets (meat, fish, wild animals).

Toxaphene

- Toxaphene is an insecticide used to protect cotton, cereals fruits, nuts, vegetables.
- Toxaphene (a mixture of up to 670 chemicals) was the most widely used pesticide in U.S. in 1975. 50% of toxaphene released can persist in soils for up to 12 years.
- Toxaphene is non-toxic to plants but highly toxic to fish and to birds.
- Humans are exposed to toxaphene through diets, levels in food are usually low.
- Toxaphene has been listed as a possible human carcinogen (UNEP, 2010; WHO, 2010) .

Polychlorinated biphenyls PCBs

- **PCBs** were first manufactured in 1929 and used in industry as heat exchange fluids, in electric transformers and capacitors (where continue

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to be used), as additives in paints, adhesives and plastics. PCBs are by-products of incomplete combustion and of some industrial processes.

- Of the 209 different types of PCBs, 13 exhibit a dioxin-like toxicity.
- Their persistence in the environment depend on the degree of chlorination, half-lives can vary from 10 days to one-and-a-half year.
- Many of the countries ceased production in the 1970s, however, PCBs remain in the environment for decades, are available for uptake and bioaccumulation in organisms. PCBs are toxic to aquatic organisms, fish and wildlife species.
- Chronic exposure can cause alteration to liver enzymes, developmental, mental and behavioural problems, immunosuppression, and possibly cancer.
- Humans are exposed to PCBs through their diet. Vegetable oils and milk, fish and marine mammals, rich in fat are at risk of PCB contamination.
- The WHO International Agency for Research on Cancer ranks PCBs as a probable human carcinogen (WHO 2016; UNEP, 2010; Ding et. al. 2013; WHO 2016).

Hexachlorobenzene (HCB)

- HCB was first manufactured in 1945 for seeds treatment (especially wheat.)
- By the mid 1980s most nations ceased its manufacture.
- HCB is generated as unintentional by-product from the manufacture of pesticides, organic chemicals (solvents, dyes), wood preservatives.
- HCB results from incomplete combustion (burning of municipal waste, fuels).
- The acute exposure to HCB of humans, animals, fish, birds, determine kidney and liver damage, central nervous effects, respiratory neurological and metabolic disorders, death. Foods (dairy products, animal meat), all over the world, contain HCB, which also has been found in Arctic air, snow, seawater, vegetation, biota. 100% of HCB contaminant in in

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pesticides (simazine, atrazine) is released into the air, posing a health risk to applicators. (Ding et. al. 2013, WHO, 2016).

Polychlorinated dibenzo-p-dioxins (PCDD)

- PCDD are produced unintentionally by incomplete combustion (burning of hospital, municipal and hazardous waste, coal, peat, wood), during the manufacture of pesticides and other chlorinated substances.
- Dioxins were found in soils 10 - 12 years after the first exposure.
- Due to their lipophilicity, dioxins accumulate in humans and wildlife tissue.
- Small amounts in contaminated water can bio-concentrate up the food chain to dangerous levels. Food (from animals) is the major source of exposure for humans.
- Dioxines can cause effects in reproductive development, immune system damage, thyroid disorders, nervous system disorders, diabetes and are teratogens, mutagens, carcinogens. Dioxins are found in all humans, with higher levels in persons living in industrialized countries.The estimated elimination half-life for dioxins in humans is 7.8 -132 years. Ding et. al. 2013, WHO, 2010, WHO, 2016).

Polychlorinated dibenzofurans (PCDF)

- PCDF/ furans are produced unintentionally from the same processes that produce dioxins and PCBs, being detected in emissions from waste incinerators and automobiles.
- Furans are structurally similar to dioxins and share many of their toxic effects.
- The 135 different PCDFs have varied toxicity and persist in environment long time.
- Food, (particularly fat animal products) is the major source of exposure for humans.

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- Furans are classified as possible human carcinogens .(Ding et. al. 2013; WHO, 2016).

ADDITIONAL POPs

Polybrominated diphenyl ethers (PBDEs)

- PBDEs are flame retardants used in production of plastic and textiles materials, computers, furniture, cars. PBDEs were banned in Europe in 2005 and US in 2003.
- PBDEs have been found at high levels in indoor dust and accumulate in human blood, fat tissue and breast milk (in US levels 40 times higher than in Europe).
- Marine mammals (dolphins, whales, etc), birds, and fish (salmon, tuna, etc) have been found to be contaminated with PBDEs. Research linked some PBDEs to effects on thyroid function, brain function, male fertility, ovarian and embryonic nervous system development (UNEP,2010; POPs Fach Sheets).

Perflorinated compounds (PFCs)

PFCs are used as industrial and commercial surfactants. In contrast to the majority of POPs that are deposited in fatty tissue, PFCs circulate in the blood and accumulate primarily in the liver. PFCs bioaccumulate and are extremely resistant to physical degradation. Studies demonstrated links between chemicals such as phthalates, bisphenol A, PFCs found in consumer products (baby toys, cosmetics,etc.) and reproductive disorders. (UNEP, 2010, POPs Fach Sheets).

ALTERNATIVES TO POPs

Regarding alternatives to POPs, there are many barriers to adaptation of new, clean technologies, especially in developing countries. Some alternatives are more costly, both in price and resources required to apply than older, most hazardous POPs. Alternatives can be more toxic to applicator than POPs,

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adding human health costs. Other problems to adoption include education and training, on both the older and new compounds, for everyone in the production chain, individual users and vendors.The infrastructure regulations needed to manage the use of alternatives to POPs and to educate and train individuals is not fully developed in all countries (UNEP, 2010; WHO, 2016).

CONCLUSIONS

The POPs are characterized by lipophilicity, persistence and semi-volatility, characteristics that pre-dispose POPs to long environmental persistence and long range transport and accumulation in the polar regions of the world, far from any source of use. POPs are also known for their ability to biomagnify and bioconcentrate under typical environmental conditions, potentially achieving toxicological concentrations. POPs have been implicated in a broad range of adverse human health and environmental effects such as reproduction and endocrine dysfunction and immunosuppression. In many cases, POPs are considered as possible human carcinogens by the International Agency for Research (IARC) on Cancer. Due to their toxic characteristics, POPs represent a threat to humans and environment. Therefore, in recent years, the international community called for urgent global action to reduce and eliminate the release of POPs and to identify their possible risk to human health and environment (UNEP, 2010; EMEP, 2014; WHO, 2016).

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