Persistent Organic Pollutants in a global changing climate

Climate change is not only altering the structure and function of natural and human systems, but is also impacting on less visible and less apparent aspects. There is a growing body of evidence that climate change will have broad negative impacts on the behaviour and fate of environmental contaminants. This paper briefly examines the information reported in the scientific literature to provide a short review of the current state of knowledge on the environmental behaviour of persistent organic pollutants (POPs) and the potential impacts of climate change on environmental dynamics of POPs. Reference is provided to the global efforts undertaken with the Stockholm Convention to reduce environmental and human exposure to POPs and to the risk that global warming could undermine such efforts

Pasquale Spezzano

Gli inquinanti organici persistenti e il cambiamento climatico globale

I cambiamenti climatici non solo stanno modificando la struttura e la funzione dei sistemi naturali e umani, ma hanno effetti anche su aspetti meno visibili ed evidenti. Numerose evidenze indicano che i cambiamenti climatici avranno notevoli ripercussioni sul comportamento e sul destino dei contaminanti ambientali. Il presente articolo esamina brevemente le informazioni riportate in letteratura per fornire una breve rassegna dello stato attuale delle conoscenze sul comportamento ambientale degli inquinanti organici persistenti (POPs) ed i potenziali effetti dei cambiamenti climatici sulla dinamica ambientale dei POPs. Viene inoltre fatto riferimento all'impegno globale assunto con la Convenzione di Stoccolma per ridurre l'esposizione ambientale ed umana ai POPs ed al rischio che il riscaldamento globale possa compromettere questi sforzi

Persistent Organic Pollutants (POPs)

Persistent organic pollutants (POPs) are chemical substances that, by definition, have three primary attributes: environmental persistence, tendency to bioaccumulate in the fatty tissue of living organisms, and toxicity. POPs are among the most toxic chemicals known to both humans and other organisms. Specific effects associated with POPs include endocrine di-

■ Pasquale Spezzano
ENEA, Technical Unit for Environmental Technologies

sruption, immunotoxicity, neurotoxicity, reproductive toxicity, mutagenicity and carcinogenicity. These effects may occur in exposed individuals as well as in their offspring. [*]

An important property of POPs is that of semi-volatility, a feature that permits these compounds to occur either in the vapour phase or adsorbed/absorbed on environmental surfaces. Some POPs cycle in the global environment for many decades because of re-volatilization from environmental compartments contaminated in the past. These chemicals can undergo seasonal cycles of evaporation from warm regions and subsequent deposition in colder regions (grass-hopping)^[1]. POPs in air may be degraded by photochemical re-



actions and via reactions with hydroxyl radical. Wet and dry deposition of the particle-bound fraction of POPs is, on average, the fastest and most effective of the removal processes from the atmosphere. The persistence of POPs in other environmental media (water, soil, and sediment) is significantly longer. Over long periods of time (years to decades) these compounds eventually degrade or are sequestered in deep soils and sediments.

As a result of releases to the environment over the past decades, POPs are widespread and are found ubiquitously in the environment, including regions, such as the Arctic, far from the place in which they were used and released. In response to this global problem, international agreements have been ratified, such as the POPs Protocol under the UN-ECE Convention on Long Range Transboundary Air Pollution (LRTAP)[2] and the Stockholm Convention (SC) on POPs^[3].

The legally-binding SC was signed in 2001 and entered into force in 2004 with the overall objective of protecting human health and the environment from POPs. Since then, 178 countries have ratified the treaty. Initially, twelve chlorinated organic chemicals (the socalled dirty dozen) were listed under the SC. Subsequently, ten new substances have been added (Table 1). Currently, five chemicals are being considered for listing¹.

A characteristic of most chemicals classified as POPs under the SC is that in the past they were high production volume chemicals. In addition, many of these productions were conducted according to the state-of-

the-art and legal framework of the time. The identification of POP substances has just begun. Brown and Wania^[4] used a data set of more than 100,000 industrial chemicals, subjected it to screening models and identified 120 chemicals which could be classified as POPs. Recently, applying screening criteria to a set of 93,144 organic chemicals, 510 substances were found which can be considered as POPs[5]. Presently, ten of these substances are high production volume chemicals, and 249 are pre-registered in the EU under the REACH Regulation².

POPs fall into three categories: chlorinated pesticides, industrial chemicals and unintentional by-products. Historically, many POPs were used as pesticides. Although the use of many POP pesticides has been banned or restricted in the industrialized countries since the '70s, their presence still remains of concern. The problem is particularly severe in many developing countries and countries with economies in transition, because of stockpiles and uncontrolled dumpsites. In Africa, it is estimated that 20% of the over 27,000 tons of obsolete pesticide stockpiles consists of POPs that have been banned under the SC^[6].

Some POPs have been used in industrial processes and in the production of a range of goods. For example, polychlorinated biphenyls (PCBs) have been widely used since 1930 as dielectrics in transformers and large capacitors, heat exchange fluids, paint additives, in carbonless copy paper, and in plastics. In 1985, the use and marketing of PCBs in the European Community were heavily restricted and, successively,

Annex A (elimination)

Aldrin*, chlordane*, chlordecone*, dieldrin*, endrin*, heptachlor*, hexabromobiphenyl#, hexabromodiphenyl ether and heptabromodiphenyl ether#, hexachlorobenzene (HCB)*#, alpha hexachlorocyclohexane*, beta hexachlorocyclohexane*, lindane*, mirex*, pentachlorobenzene*#, polychlorinated biphenyls (PCBs)#, technical endosulfan and its related isomers*, tetrabromodiphenyl ether and pentabromodiphenyl ether#, toxaphene*.

Annex B (restriction)

DDT*, perfluorooctane sulfonic acid, its salts and perfluorooctane sulfonyl fluoride#.

Annex C (unintentional production)

Polychlorinated dibenzo-p-dioxins (PCDD), polychlorinated dibenzofurans (PCDF), hexachlorobenzene (HCB), polychlorinated biphenyls (PCBs), pentachlorobenzene

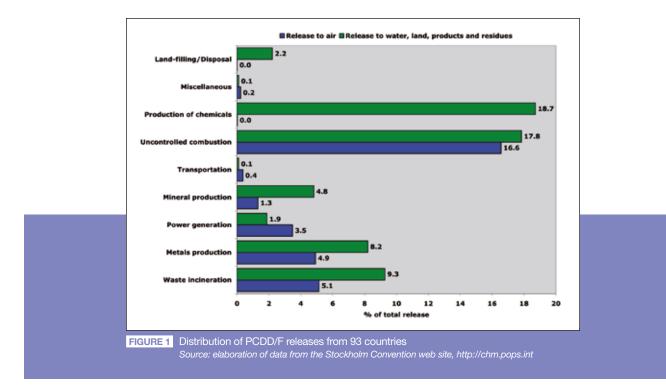
* Pesticide; # Industrial chemical

TABLE 1 Listing of POPs in the Stockholm Convention Source: Stockholm Convention web site, http://chm.pops.int production, marketing, and use were completely banned. However, about 30% of the estimated 700,000 tons of PCBs produced in Europe has already been released into the environment. Several brominated flame retardants (BFRs) have characteristics that qualify them as POPs. After manufacture, the treated products can release BFRs during use and as waste after disposal. Thus, although several BFRs-POPs such as polybrominated diphenyl ethers (PBDEs) have been banned or phased out, the long life-times of the products which they are incorporated in will lead to their continued release for several decades to come^[7]. Today, disposal and recycling of POP-containing applications (such as electronic equipment) are increasingly relocated to developing countries.

Unintentionally produced POPs (U-POPs) such as dioxins and furans (PCDD/F) are formed as by-products during combustion and manufacturing processes. It is thought that global releases of U-POPs have remained the same over the last decades, or even increased, due to increased global combustion and production. In addition, there has been a significant shift in the emission of these substances from developed to developing countries. This suggests that, given the expected increased energy and goods demands in developing countries, global releases of U-POPs may increase in coming years.

The development of global emission inventories for U-POPs is challenging. The SC request Parties to establish national release inventories. The preliminary inventories were generally made according to the methodology recommended in the UNEP Standardized Toolkit[8]. From a selection of PCDD/F inventories for 93 countries (excluding the EU), covering a wide range in terms of geographic distribution, size, population and industrial development, the total annual PCDD/F releases to the five main vectors (air, water, land, product, and residue) can be calculated in 92.8 kg TEQ3 per year. The atmosphere receives 34% of the total release (31.6 kg TEQ per year) (Figure 1). It has been estimated that in the EU-25 some 21 kg TEQ of PCDD/F are released per year, of which around 5 kg to air, and 16 kg as waste^[9].

Available inventories show that in developed countries,





the main PCDD/F sources are the ferrous and nonferrous metal industries and waste incineration, whereas the predominant sources in developing countries are releases from open burning processes as well as forest fires, and pre- and post-harvest burning in agriculture. In Europe, the contribution of domestic sources to U-POP emissions is becoming increasingly important in relative terms. It was estimated that these sources may contribute with as much as 45% of the total emissions of PCDD/F to air[9].

However, the largest sources of PCDD/F release to the environment are probably related to past events of formation and releases[10]. PCDD/F contamination from pesticide use between 1950 and 1998 has been estimated at 460 kg TEQ in Japan alone. Timber treated with pentachlorophenol (PCP) and similar compounds resulted in an estimated total of 205-250 kg TEQ incorporated in timber in Sweden. Similarly, 378 kg TEQ of PCDD/F were released from a single factory producing pesticides in Hamburg, and more than 366 kg TEQ were released from spraying of defoliants during the Vietnam War^[11]. The accident at the Hoffmann-La Roche subsidiary ICMESA in Seveso, in the summer of 1976, released anything from hundreds of grams to 34 kilograms of 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD)[12, 13].

Impact of climate change on the environmental fate of POPs

Climate change induced by anthropogenic activities is one of the major global problems, with significant social, economic, ecological, and health related impacts. The growing concern about global warming has led to the establishment of international agreements such as the Kyoto Protocol and the Intergovernmental Panel on Climate Change (IPCC). Climate variability forecasted by climate change scenarios presented by the 4th IPCC Assessment Report[14] shows that the atmospheric temperature is expected to increase by 1.8-4.0 °C by the end of the century. In addition, climate change will affect the atmospheric and oceanic circulation patterns, and the precipitation rate.

As the environmental behaviour of chemicals is governed by environmental factors, one of the consequences of climate change is its effect on the environmental distribution of chemical pollutants[15]. The Arctic Monitoring and Assessment Programme (AMAP) has conducted several activities in recent years documenting the link between climate change and the environmental transport and fate of contaminants, including POPs^[16]. A joint UNEP/AMAP expert group reported on the implications of climate change on POPs and found that climate change is likely to increase exposure to POPs in some regions[17].

The distribution of an organic compound between air, water, sediment, soil and biota depends on complex interactions between different factors within an open environmental system, and is largely dependent on some key equilibrium parameters which include vapour pressure (P), water solubility (S), Henry's law constant (H), partition coefficient octanol/air (KOA), and partition coefficient octanol/water (K_{OW}). Vapour pressure affects the volatility of a chemical from various substrates. It governs, through the Henry's law constant, the exchange rate of a chemical across an air-water interface. KOA is used to describe partitioning from air to aerosols, vegetation and soils while Kow is used to describe the uptake to aquatic organisms from water. The effects of global warming on the environmental behaviour of POPs can be predicted by considering temperature-driven changes in these partitioning constants[18].

Temperature is an important factor in determining the environmental behaviour and fate of POPs, as it has a direct influence on vapour pressure[19, 20]. An increase in temperature of 1°C increases the volatility of a typical POP by 10-15%. At the local level, atmospheric temperatures can, however, increase much more: for instance, an increase from 10°C to 15°C doubles vapour pressure of PCB-153[21]. Global warming will therefore lead to enhanced volatilization of POPs from contaminated environments, stockpiles and open applications.

The effect of higher temperatures on the increase of secondary emissions from contaminated environments is supported by several experimental evidences[19, 22-24]. On the other hand, an increase in temperature could accelerate the atmospheric degradation of POPs, offsetting their increase in atmospheric concentrations[1, ^{25, 26]}. These processes respond in opposite directions



to changes in temperature and the net result is difficult to predict. Additionally, degradation of POPs often includes the formation of products that are structurally similar to the parent compound and may also be similarly toxic and persistent^[27].

Winds are the most important factors for the atmospheric transport of POPs. Modified wind patterns and higher wind speeds, as expected in the future, will lead to faster and more efficient atmospheric long-range transport of POPs. Desertification induced by climate change might also lead to enhanced distribution of POPs through dust transport associated with altered wind fields^[16].

Climate change will also lead to changes in precipitation patterns. Precipitation projections in a changing climate differ from region to region, indicating both decreasing and increasing trends^[14]. A decrease in the precipitation rate will lead to enhanced volatilization of POPs to the atmosphere, while an increase in intensity and frequency of rain events will lead to an enhanced wet deposition of airborne POPs^[15]. Snow melting is important in POPs cycling^[28]. Abundant snow deposition may lead to a large contaminant release during snowmelt, with the potential to impact drinking and agricultural water supplies^[29].

The IPCC^[14] reports that extreme precipitation events are expected to become more frequent, widespread, and intense. The impact of extreme events on the remobilization and redistribution of POPs has been documented^[30]. As storms and rainfall events become more intense and frequent, increasing amounts of POPs bounded to soil particles could be transported by erosion and transferred to rivers, lakes and oceans, making them available to the aquatic environments^[29]. Flooding events may also contribute to the dissemination and redistribution of POPs formerly stored in sediment and soils^[31,32].

Of particular concern is the melting of glaciers, which cover most of the Polar regions, Greenland and mountainous areas such as the Alps. Because of the global transport, the Arctic acts as a long-term sink for POPs. In addition, at low temperatures POPs degrade at a slower rate than in temperate regions. In some cases, POPs are present in aquatic and terrestrial ecosystems of the Arctic at levels similar to those in in-

dustrialised countries^[33]. Temperature increases due to climate change are more pronounced at higher latitudes^[14]. Undoubtedly the Arctic environment is the most affected and vulnerable area with respect to both global warming and POPs contamination.

Melting of polar ice caps as well as loss of permafrost result in the release of stored pollutants making them available for transfer to the atmosphere or to aquatic and terrestrial ecosystems^[18]. A recent study^[34] provides the first evidence that some POPs, previously stored in snow, ice, ocean, and presumably soil reservoirs, are being remobilized back into the Arctic atmosphere as a result of climate change. The atmospheric and oceanic circulation patterns could carry the released POPs to other parts of the globe. Release of POPs from mountain glaciers to Alpine lakes has already been observed^[35,36].

Polar ice melting and increased evaporation rates influence ocean salinity, which in turn affects the solubility of organic chemicals (POPs are less soluble in water if salinity is higher) and, consequently, air-water partitioning. Salinity and wind patterns influence oceanic currents, which are important in determining POPs cycling^[37]. These modified currents can provide an increased pollution in some regions of the globe. However, changes in ocean currents will mainly affect the transport of the more water-soluble POPs, such as HCHs and perfluorinated acids^[38].

In addition to the many abiotic factors that can influence the behaviour of contaminants, organic carbon cycling, lipid dynamics and food web structures in terrestrial and aquatic systems can be adversely affected by climate change, which will in turn alter POPs transfer in biota^[39, 40]. A result by IPCC^[14] showed that approximately 20-30% of plant and animal species assessed so far are likely at risk if increases in global average temperature are greater than 1.5°C-2.5°C. Climate change will affect ecosystem functions, biodiversity and population dynamics^[41-43] (Figure 2). Thus, climate change will impact on the transfer of POPs through the food chains, from the absorption of POPs in phytoplankton and zooplankton from water to the bioaccumulation and biomagnification in top predators[15, 18, 34, 43].

Migratory species, such as fish, birds, and marine

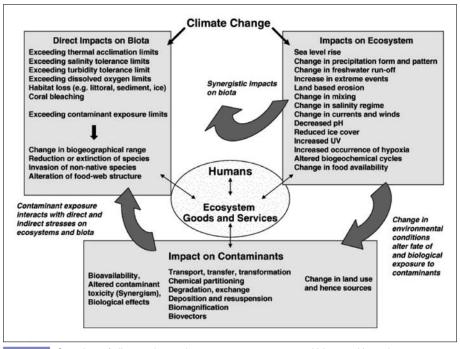


FIGURE 2 Overview of climate change impacts on ecosystems and biota and how they may interact with contaminants fate and effects Source: Schiedek et al., 2007

mammals, can assimilate POPs in one location and transport these contaminants to other locations[44]. This biotic transport may be similar in magnitude to the atmospheric and oceanic transport^[45]. Change in the species migration patterns related to climate change could be an important factor modulating the local and global transport of POPs[18,46].

Food is the main route of the potential background human exposure to POPs[47]. This dietary exposure will be affected by any changes in the structure of the relevant food webs associated with climate change. The groups of populations most at risk from exposure to POPs, and therefore more likely to be affected by climate-related influences are, in general, developing foetus, children and the elderly.

There is plenty of experimental and modelling evidence to suggest that climate change impacts the environmental fate of POPs^[48, 49]. Monitoring programs over long time periods performed under the AMAP indicate that both temporal and spatial patterns of POPs

in the Arctic air may already be affected by various processes driven by climate change^[50]. Changes in sea ice cover, temperature, precipitation rates, and primary production have been identified by modelling and sensitivity analyses as the factors that have the greatest impact on the transport and accumulation of POPs[20, 21, 28, 51-53]

Managing the problem

The only long-term solution to reduce the level of POPs in the environment is to prevent these substances from being released. Intentionally produced POPs currently listed in the SC are subject to a ban on production and use except where there are generic or specific exemptions. The production and use of DDT - a pesticide still used to control malaria and other vectors of disease in developing countries - is severely restricted. The introduction in the market and, therefore, into the environment, of new chemicals with POP characteristi-



cs could be prevented by more restrictive legislations. In Europe, the production or import of new POPs could be prevented by the REACH Regulation. Under this framework, companies that manufacture or import more than one tonne of a chemical per year are required to register it in a central database. Substances of very high concern, including persistent, bio-accumulating and toxic substances (PBT) and very persistent and very bio-accumulating substances (vPvB) require authorisations for particular uses. The European Chemicals Agency (ECHA) has the right to request further testing if it suspects that a substance might exhibit POP characteristics.

As a priority action, the SC requires the identification and safe management of stockpiles containing or consisting of POPs. Waste containing, consisting of, or contaminated with POPs should be disposed of in such a way that POPs are destroyed or irreversibly transformed. With regard to the identification and remediation of sites contaminated by POPs, the SC encourages Parties to develop strategies for identifying contaminated sites; if remediation is necessary then it must be done in an environmentally sound manner.

The U-POPs flow is characterised by relatively small amounts that are constantly formed and released. Remarkable stocks that need to be disposed of do not exist. The SC requires Parties to take measures to reduce the total releases of U-POPs. The crucial point in reducing future loading of U-POPs in the environment is to reduce their formation by applying Best Available Techniques (BAT) and Best Environmental Practices (BEP). These may include end-of-pipe solutions or the development of substitute or modified materials, products and processes that avoid the formation and release of U-POPs. Guidelines on BAT/BEP have been developed under the SC^[54]. Processes and technologies preventing POPs from being formed and transferred to air, water, soil and waste streams should be introduced.

The obligations and objectives of the SC are very ambitious. The issue is particularly severe in many developing countries and countries with economies in transition because of limited financial and technological resources. The SC recognises the particular needs of these countries, therefore the general obligations include provisions on technical assistance and streng-

thening of the national capabilities through the promotion of cooperation and exchange of information and technology transfer.

A range of activities focussed on assisting developing and transition economy countries to meet their obligations under the SC are currently being led by international organisations. In this context, ENEA is providing the United Nations Industrial Development Organization (UNIDO) with consultancy services in the development of demonstration projects aimed at promoting the uptake of BAT and BEP in industry, mainly the ferrous and non-ferrous metal industry, one of the most critical sector. These activities are targeted at avoiding the creation of a technology base inefficient and highly impacting on the environment and human health.

Conclusions

POPs that have been banned or regulated decades ago are sometimes referred to as 'legacy' POPs, because present day contamination is largely a 'legacy' of the past. Some POPs are preserved almost indefinitely in the environment. Most of these pollutants generated by our grandparents have been stored in environmental reservoirs such as soil, ocean water and glaciers over the past decades.

There is a growing body of evidence that climate change scenarios will result in a substantial release of POPs from their reservoirs and will affect the environmental fate of POPs at the global, regional and local scales. Whilst little can be done for pollutants released in the past and presently still 'hidden under the carpet', much can be done to avoid the production of new substances with POPs characteristics and to reduce the releases of unintentionally produced POPs. Yet, global warming could undermine the global efforts made to reduce the environmental and human exposure to POPs.

- notes
 - Hexabromocyclododecane, short-chained chlorinated paraffins, chlorinated naphthalenes, hexachlorobutadiene and pentachlorophenol.
 - Regulation (EC) 1907/2006 concerning the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH).
 - TEQ: Toxic EQuivalent. A system to calculate the total toxicity of the sum of several PCDD/F congeners by rating them against the most toxic one (2,3,7,8-tetrachlorodibenzo-p-dioxin).

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