Atmospheric Pollution Research 3 (2012) 466-476

Critical Literature Review

Atmospheric Pollution Research

www.atmospolres.com

Climate change effects on POPs' environmental behaviour: a scientific perspective for future regulatory actions

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ABSTRACT

Since the adoption of the United Nations Framework Convention on Climate Change, international efforts were aimed at limiting global change, and at managing and reducing its inevitable impacts. The growing concern on climate change related issues lead to create international agreements such as the Kyoto Protocol, and to establish the Intergovernmental Panel on Climate Change aimed at studying climate evolution and at defining common actions through the adoption of joint climate change mitigation and adaptation measures. From the time when international Task Forces, projects and programs were shared in order to deal with the reduction of environmental exposure to persistent organic pollutants (POPs), international organisations have also been committed to estimate how climate change may affect POPs' environmental behaviour and distribution. In this review paper, we report the track of POPs' regulation efforts driven towards decreasing POPs' environmental concentrations through reducing or banning POP emissions in the environment. We also report scientific studies on climate change related effects on POPs' environmental behaviour in order to feature how climate change is influencing POPs' fate and transport. Our final aim is to identify how POPs–related regulations may take into account climate change in managing current or future POPs sources. We find in several case studies on this topic that climate change is considered to contribute to enhance POPs' long–range transport and that remote areas are considered likely to be the most impacted by POPs' pollution under a climate change perspective. Our findings also consider that continuous monitoring programs oriented towards the observation of secondary POP sources and the enhancement of inventories reporting primary and secondary POP emissions are useful in dealing with POPs' exposure under climate change scenarios. We also suggest how communication between science and regulation should be driven towards considering climate change effects into chemicals' legislation.

Keywords:
Persistent organic pollutants
Climate change
Regulation
Monitoring programs
Multimedia modelling

Article History:
Received: 09 May 2012
Revised: 11 September 2012
Accepted: 13 September 2012

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doi: 10.5094/APR.2012.054

1. Introduction

1.1. Persistent organic pollutants (POPs)

Persistent organic pollutants (POPs) are chemical substances that resist to environmental, chemical, physical and biological degradation; because of their characteristic persistence, they are transported through air, water and migratory species across international boundaries and deposited far from their place of release, where they accumulate in terrestrial and aquatic ecosystems (UNEP, 2001). POP inputs in the environment can be distinguished in primary and secondary sources. Primary sources are those with direct fluxes into the environment and secondary sources are already contaminated environmental compartments that can release POPs in a time subsequent to their use or production (Hung et al., 2010; UNEP/AMAP, 2011). Due to their wide distribution, ability to bioaccumulate, and potential harmful effects such as immunotoxicity, neurotoxicity, developmental toxicity, carcinogenicity, mutagenicity, and endocrine disruption potentials, POPs have drawn scientific and political concern during the last decades (WHO, 2003).

POPs undergo long–range transport (LRT), meaning that they are transported to areas that are remote if compared to the source regions. In order to manage the harm posed by POPs, several conventions and initiatives were initiated in the past decades. The first national regulations and regional agreements, dealing with POPs' management were introduced in the late '70s and aimed at banning or limiting the use and production of some POPs (e.g. dichlorodiphenyltrichloroethane –DDT, polychlorinated biphenyls –PCBs), and in the same period some monitoring programmes were also established.

More recently, the European Parliament adopted a legislation on the Registration, Evaluation and Authorisation of Chemicals (REACH) (EU, 2006; EEA, 2007; ECHA, 2008), that was enforced in 2007. It aims at collecting information on the large amounts of chemicals that entered the European market without appropriate information on the hazards that they may pose to human health and the environment. This is done through the definition of new procedures for the registration of chemicals marketed in Europe and through the identification of a framework for assessing chemicals' impacts on human health and on the environment in order to minimize and avoid future harms caused by exposure to chemicals (ECHA, 2011). Under this framework, a procedure for special approach regarding Persistent, Bioaccumulative and Toxic chemicals (PBTs) and very Persistent and very Bioaccumulative chemicals (vPvBs) (including POPs) (ECHA, 2008) was proposed to provide assessment on persistence, bioaccumulation and toxicity of marketed chemicals. This procedure requires the characterisation of the substances and related emissions, the definition of human and environmental exposure and risks associated with the subsequent uses, collecting information on chemicals' persistence, bioaccumulation and toxicity, assessment of exposure and impact on population or environmental stocks, evaluation of the economic value of health and environmental impacts, and an analysis of the positive or negative changes on health or environmental effects (ECHA, 2011). REACH legislation is currently seen as the European contribution to the Strategic Approach to International Chemicals Management (SAICM). The adoption and implementation of the SAICM's goals are related with the linkages between chemicals management, environment, and sustainable development organized under the auspices of the United Nations Economic Commission for Europe (UNECE). The REACH strategy may lead to classification of a substance as either persistent or not, or it may indicate the need for further testing and represents a substantial improvement of testing strategies and interpretation of results from biodegradation tests, maximizing the use of existing data, using multimedia models to identify and prioritize the most important environmental compartments, using this information to help determining the types of persistence data that would be most useful.

Similarly, the implementation of international conventions and bodies such as the Mediterranean Action Plan for the Barcelona Convention (MAP) (UNEP, 1978) and the Arctic Monitoring and Assessment Programme (AMAP), clearly established that long–range atmospheric transport is one of the main routes by which POPs are deposited in remote regions (Arctic Council, 1991).

1.2. Climate change and its effects on POPs' environmental behaviour

In 1980, the World Climate Research Programme (WCRP) was established by the joint action of the International Council for Science (ICSU) and the World Meteorological Organization (WMO) with the goal to determine how predictable the climate is and to assess the effect of human activities on climate.

Several studies reported evidence of current and past climate warming from sea surface temperature (SST) and land surface temperature (LST) observations (Smith and Reynolds, 2005; Brohan et al., 2006; Hansen et al., 2010), showing a more rapid temperature increase at higher latitudes than in the global average air and ocean water (Solomon et al., 2007; Rummukainen et al., 2010). Some consequences related to the increasing temperature are the faster rate of ice melt in the Arctic Sea, in Greenland and Antarctica, and the reduced area covered by ice caps and glaciers (AMAP, 2011), and global sea level rise (Solomon et al., 2007; Rummukainen et al., 2010).

The IPCC Fourth Assessment Report (Solomon et al., 2007) addressed the scientific progress in climate science (Rummukainen et al., 2010; The Royal Society, 2010), and reported recent observations of particular relevance for the vulnerable regions, such as the Arctic (AMAP, 2011), Antarctic (Chen et al., 2009), Greenland and the Alps (EEA, 2007).

When dealing with observed climate change, anthropogenic forcings are taken into account (e.g. anthropogenic emissions of greenhouse gases, land use change) together with environmental factors contributing to climate change (e.g. solar variability and volcanic eruptions) (Forster et al., 2007; Rummukainen et al., 2010). Climate change induced faster changes from the Earth system response in several regions, consisting for instance in higher summer temperatures in the Arctic that are suggesting that the warming rate over the last 50 years is nearly twice that for the last 100 years (Solomon et al., 2007), and in the changes in net accumulation and melt of Greenland ice sheets (Jansen et al., 2007; AMAP, 2011).

Environmental variables such as temperature, wind speed, precipitation, and solar radiation influence directly or indirectly POPs' environmental fate and transport. Thus, climate warming is likely to influence the environmental behaviour of POPs by enhancing the volatilisation from primary and secondary sources, by influencing their partitioning between soil, sediment, water and atmosphere, including air–surface exchange, wet/dry deposition, and reaction rates (Noyes et al., 2009; Armitage et al., 2011). In addition, climate change also encompasses the alteration of other important processes that influence POPs' fate and transport, such as snow and ice melting (Bogdal et al., 2008), biota lipid dynamics (Matz et al., 2011), and organic carbon cycling (Nizzetto et al., 2010), changing chemicals' fugacity capacity in environmental compartments (Noyes et al., 2009; Armitage et al., 2011; Lamon et al., 2012). Moreover, climate variability may induce interannual variations of POPs' revolatilisation that could undermine global efforts aimed at reducing environmental and human exposure to POPs (Klanova et al., 2011; Ma et al., 2011).

Each climate variable is associated to its variability and to the uncertainty related to the projected climate change scenario (IPCC, 2005). This uncertainty needs to be taken into account in POPs' multimedia modelling together with the uncertainties related to physical–chemical properties.

In this paper, a review of the literature concerning POPs' environmental behaviour affected by climate change and some suggestions on possible future regulatory actions is presented in order to : (1) better understand how global warming could undermine global efforts to reduce environmental and human exposure to POPs; (2) highlight climate change effects increasing POPs' environmental mobilization, exposure and transfer; (3) emphasize how regulations could address new POPs, assuming that environmental processes driven by climate change may affect temporal and spatial exposure to POPs; and (4) highlight the importance of screening for new or potential POPs assuming that temporal and spatial patterns of POPs may be affected by various processes driven by climate change.

2. State of the Art on POPs' Regulation

POPs are the main focus of several national and international regulatory efforts aiming at reducing their emissions because of their high potential for LRT, persistence, and potential for bioaccumulation and their adverse effects on human health and the environment (AMAP, 1998; Breivik et al., 2002; NCP, 2003; Wania, 2006; TF-HTAP, 2010).

2.1. International treaties on POPs

Transboundary air pollution is one of the most concerning environmental issues that attracted International Communities' efforts in the last 40 years. POPs are considered a global concerning issue and several efforts were done in the past decades to eliminate their emissions through the adoption of international agreements and conventions, as we anticipated before.

High exposure to POPs in remote regions, observation of POPs' high concentrations in boreal biota from marine, freshwater and terrestrial ecosystems (Wania, 1999; MacDonald et al., 2002; Meyer and Wania, 2008) are examples of the high international concern on POPs' toxicological effects.

Past studies proved the partial effectiveness of control actions by POPs' regulations. In Japan, for instance, Kumar et al. (2005) highlighted the evidence that the reduction of some POPs is apparent since some were detected in both the organisms and in the environment, notwithstanding the ban of those substances 30 years ago. Following regulatory restrictions, the levels of hexachlorobenzene (HCB), hexachlorocyclohexane and chlordanes declined in most world regions, while some others, like alpha–endosulfan, chlorothalonil and trifluralin were reduced only for some regions such as Europe. However, some other regions, such as India, showed high concentrations of organochlorine pesticides.

The first internationally legally–binding instrument to deal with air pollution on a broad regional basis was the Convention on Long–range Transboundary Air Pollution (CLRTAP). Its main objective is to reduce and prevent long–range transboundary air pollution through the knowledge of transfer pathways between countries, promoting monitoring, modelling and prevention of pollutants' emissions into the environment (http://www.unece.org/env/lrtap/lrtap_h1.html).

Although CLRTAP was originally ratified in 1983 to address sulphur emissions in Western Europe, and its respective resulting acidification of the Scandinavian lakes, Canada´s Federal Government prepared and presented a report just in 1989 on POPs in the Arctic to the CLRTAP Working Group on Effects and the Convention's Executive Body. Following preliminary studies demonstrating clear evidence that actions to address POPs should be warranted, in 1998 the Aarhus Protocol was drafted and signed with the objective to control, reduce or eliminate discharges, emissions and losses of a list of 12 POPs, including pesticides, industrial chemicals, and by–products. The protocol banned the use of some of them, while others were scheduled for future elimination.

After the official recognition that POPs' emissions are affecting remote regions and that atmosphere is the dominant pathway, in 1995 the Governing Council of UNEP invited the Intergovernmental Forum on Chemical Safety and other international bodies to assess the 12 previously identified POPs. The "dirty dozen" were those chemicals that had almost certain adverse effects on human health and the environment (Noren and Meironyte, 2000; Schecter et al., 2003; Furst, 2006).

The resulting international legally–binding "Stockholm Convention" was finally signed in 2001 and entered into force in 2004, with the aim of addressing the identified list of POPs and of identifying additional candidate substances for future actions (UNEP, 2009). The first task of the UNEP Stockholm Convention since its adoption was consisted in proposing a list of criteria for the identification of other POPs' candidates. Following these criteria, the substance has to be persistent, bioaccumulative, toxic and have potential for long–range environmental transport (UNEP, 2009). When a substance fulfils these requirements, then a risk profile is required in order to evaluate the effects of chemicals' emissions. This step involves a hazard assessment for the identified endpoint(s), including the toxicological interaction with multiple chemicals, persistence and bio–accumulation, monitoring data, and the definition of environmental exposure. Finally, if the risk profile concludes that the substance is toxic and/or has environmental effects, a risk management evaluation is needed, reflecting socioeconomic considerations associated with possible control measures (UNEP, 2009). Amendments to the Stockholm Convention enlarged the list up to 22 POPs in 2011, as shown in Table 1.

Other POPs–related international efforts were carried out for monitoring biotic and abiotic environments over several decades such as the Convention for the Protection of the marine Environment of the North–East Atlantic (OSPAR) (Ministerial Meeting of the Oslo and Paris Commissions, 1998).

2.2. POPs' monitoring campaigns

Several attempts were made in the last two decades to monitor current emissions, to collect information for building emission inventories and to identify contributions of point and diffuse sources, in order to reduce the uncertainty related to model predictions (Halsall et al., 1994; IADN, 2000; King and Adeel, 2002; Pozo et al., 2006; Klanova, 2009a) both at regional and global scales.

The early approaches used to identify any particular substance and to reach a regional or global agreement for its elimination or restriction, stressed the importance of a programme for monitoring and modelling of long–range transport of air pollutants (UNECE/EMEP, 1977; UNECE/CLRTAP, 1979; AMAP, 1991; Shatalov et al., 2001). Pollutants emitted by primary sources have been commonly monitored (Breivik et al., 2002; AMAP, 2003), evaluated and modelled, with the aim of estimating temporal and spatial patterns, thanks to the global and regional monitoring programs reported in Table 2. Global monitoring programs are considered here as the ones covering areas in different continents or international waters. Monitoring programs are also reported in Figure 1.

Global POPs' monitoring programs. POPs' time trends have been investigated by collecting data through active sampling monitoring activities over long time periods, some lasting for more than a decade. For example, the Arctic Monitoring and Assessment Programme (AMAP) established in 1992, providing information on the status of the Arctic environment through data collection by high volume air samplers with sampling frequencies varying from site to site (from one sampling each year to one sampling each month). In addition, satellite stations were installed in the Arctic by AMAP, which were in operation at various times of periods ranging from 3 months to 2 years (Hung et al., 2010). OSPAR also carries out monitoring activities in the area of interest as it is shown in Figure 1, as POPs are listed under the OSPAR list of hazardous chemicals.

Unfortunately, due to the high operational cost of high volume active samplers, POPs' atmospheric monitoring programs have only been conducted at a reduced number of sites. To solve this problem, Passive Air Sampler (PAS) programs were initiated: the Global Atmospheric Passive Sampling (GAPS) network is a key global program for producing comparable global–scale data for POPs. This program started in December 2004 and considers three month sampling periods at more than fifty sites all over the world (Pozo et al., 2006; Gusev et al., 2009).

Regional POPs' monitoring programs. Several attempts to integrate the components of global and regional monitoring networks were undertaken. Regional programs such as the European Monitoring and Evaluation Programme (EMEP, http://www.emep.int/), the Integrated Atmospheric Deposition Network (IADN) and the Northern Contaminants Program (NCP) provided relevant monitoring data to global programs like AMAP.

EMEP was established in 1977 with the objective of collecting, analysing and reporting emission data, measurement data and integrated assessment results. Many POPs monitoring sites from EMEP were (some of them still are) operated in support of already existing programmes, such as the Helsinki Commission or Baltic Marine Environment Protection Commission (HELCOM) (The Helsinki Commission, 2000) that aimed at protecting the marine environment of the Baltic Sea Area including the water–body and the seabed, their living resources and other forms of marine life. Furthermore, the National Monitoring Network in Europe (MONET) was designed and operated by the Research Centre for Environmental Chemistry and Ecotoxicology (RECETOX), aiming at collecting monthly samples at a number of EMEP sites to support relevant efforts for improving data consistency on air quality in most of Central and Eastern Europe, through capacity building and transfer of technology under the Stockholm Convention on POPs, GAPS and MONET also in support of EMEP. The MONET program consists of monitoring stations in Central, Southern and Eastern Europe (MONET–CEEC) (Gasic et al., 2010; Klanova et al., 2011), MONET in Africa (Klanova et al., 2009b; Lammel et al., 2009) and MONET in Pacific Islands (MONET–PI) (Lal et al., 2009; Brebbia et al., 2011).

Table 1. *Listing of POPs in the Stockholm Convention and the Aarhus Protocol. List of POPs that are included in the Stockholm Convention aiming at eliminating (Annex A), restricting (Annex B) the production, use, release and unsafe disposal of POPs and reducing (Annex C) their unintentional releases (Annex C). The table includes POPs that are listed in the Aarhus Protocol that has the objective to control, reduce or eliminate discharges, emissions and losses of POPs, including pesticides, industrial chemicals, and by-products. The protocol scheduled some of them for elimination (Annex I), for restrictions on use (Annex II), and for reductions in annual emissions from the level of the emission in a reference year (Annex III). The table also shows the "dirty dozen" that were the initial POPs included before 2009 in both agreements*

	Stockholm Convention			UNECE POPs (Aarhus Protocol)		
Chemical	**Annex A**	**Annex B**	**Annex C**	**Annex I**	**Annex II**	**Annex III**
Pesticides						
Aldrin	X			X		
Chlordane	X			X		
DDT (dichlorodiphenyltrichloroethane)		X		X	X	
Dieldrin	X			X		
Endrin	X			X		
Heptachlor	X			X		
Mirex	X			X		
Toxaphene	X			X		
Chlordecone	X			X		
alpha- HCH (alpha-hexachlorocyclohexane)	X			X	X	
beta-HCH (beta- hexachlorocyclohexane)	X			X	X	
Lindane (gamma-hexachlorocyclohexane)	X			X	X	
Pentachlorobenzene [a, b]	X		X	X		
Endosulfan and its isomers	X					
Industrial-chemical						
HCB (hexachlorobenzene) [a, b]	X		X	X		X
PCB (polychlorinated biphenyl) [a]	X		X	X	X	X
HBBP (hexabromodiphenyl)	X			X		
hexa-BD (hexabromodiphenyl ether) and hepta-BD	X			X		
PFOS (perfluorooctane sulfonic acid, its salts:		X		X	X	
Tetrabromodiphenyl ether and pentabromodiphenyl	X			X		
Industrial by-products						
PCDD (polychlorinated dibenzo-p-dioxins)			X			X
PCDF (polychlorinated dibenzofurans)			X			X
PAH- benzo(a)pyrene						X
PAH- benzo(b)fluoranthene						X
PAH- benzo(k)fluoranthene						X
PAH- indeno(1,2,3-cd)pyrene						X
Hexachlorobutadiene				X		
PCN (polychlorinated naphtalenes)				X		
Short-chain chlorinated paraffins				X	X	

Notes: Annex A: chemicals production and use must be eliminated; annex B: restrictions for production and use; annex C: reduce the unintentional releases. Annex I: substances scheduled for elimination; Annex II: substances scheduled for restrictions on use; Annex III: substances scheduled for reduction in annual emissions from the level of the emission in a reference year.
[a] Pesticide and Industrial chemical
[b] by-product chemical
The "dirty dozen" are given in italics.

IADN was developed as a sampling network around the Great Lakes in North America as a collaborative effort between Canada and the United States that was in operation since 1990 and aims at (i) determining atmospheric loadings and trends of priority toxic chemicals, (ii) acquire measurements, and (iii) helping in identifying POP sources.

Another monitoring activity on POPs in Canada was provided by the NCP, established in 1991, with the aim of reducing and possibly eliminating contaminants from the Arctic environment. NCP was responsible also for activities related to POPs' modelling to identify, understand and illustrate the connection between POPs' sources in industrial and agricultural regions to the Northern Hemisphere and the contaminants' transport to remote regions of the Arctic (NCP, 2003; Hung et al., 2005; Macdonald et al., 2005).

In Asia in 1996 the project "Environmental Monitoring and Governance in the East Asian Hydrosphere –Monitoring of POPs" (EMGEAH) was launched as a joint collaboration between the United Nations University (UNU) and Shimadzu Corporation. It involves monitoring pollution from POPs' land–based sources in marine and coastal environments (King and Adeel, 2002; Iino et al., 2009; Wang et al., 2012). The EMGEAH network has monitored POPs in water, sediment, soil and biota. These monitoring data provided a general overview of the levels of POPs in the Eastern Asia (Iino et al., 2009), contributing to the effective implementation of Stockholm Convention towards the production of an inventory of priority pollutants (Wang et al., 2010), starting the process of environmental risk assessment in some areas of the country (Wang et al., 2011; Wang et al., 2012).

The Western Airborne Contaminants Assessment Project (WACAP) (http://www.nature.nps.gov/) which operated from 2002 to 2007 was designed to determine the risk from airborne contaminants to ecosystems and food webs in western national parks of the United States, through conducting analysis of the

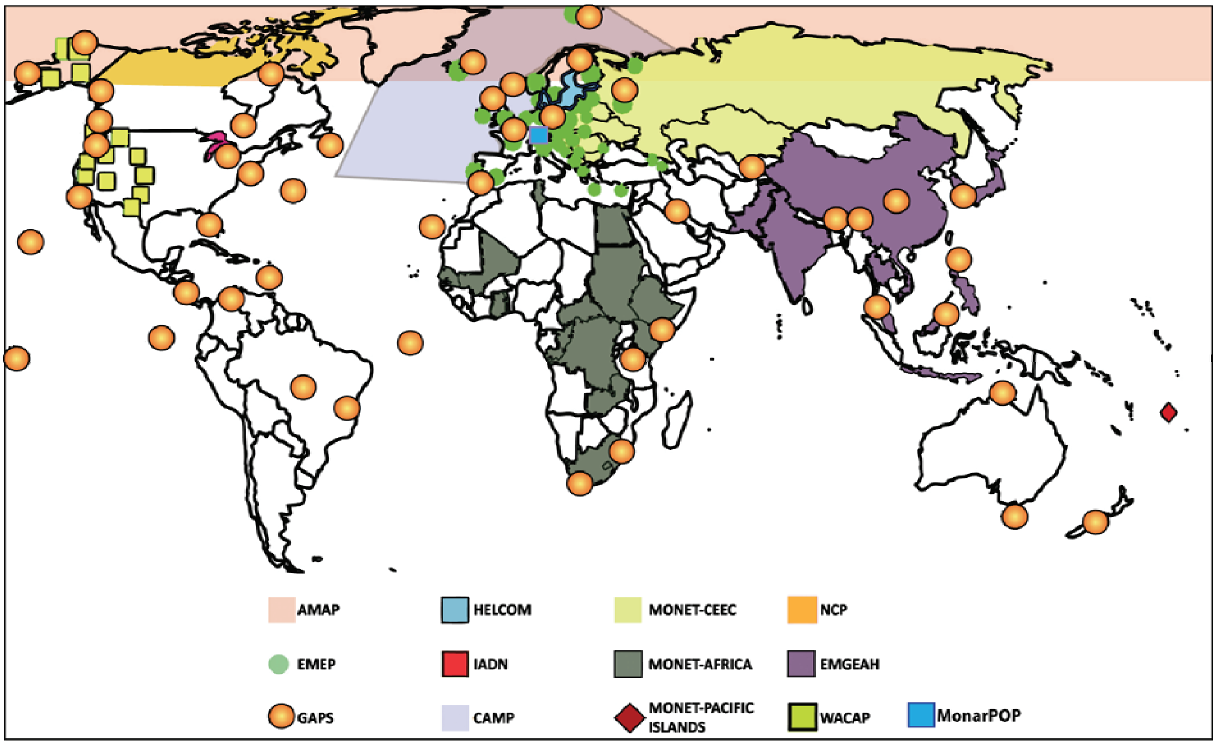

Figure 1. *Global and regional international monitoring programmes. Geographical distribution of POPs' global and regional monitoring campaigns. All the monitoring programs in the figure are also listed in Table 2. When possible, single sampling stations were reported.*

concentrations and biological effects related to exposure to POPs. POPs were monitored in air, snow, water, sediments, lichens, conifer needles, and fish in watersheds in each of eight core parks in the Western United States, including Alaska (Landers et al., 2008). Results from WACAP highlighted existing strong correlations between the concentration of several chemicals (namely DDT, polycyclic aromatic hydrocarbons–PAHs, chlordanes and endosulfans) in lichens and in conifer needles in areas located in proximity of intensive farming, indicating that in most cases high environmental concentrations are probably attributable to regional agricultural sources. On the other hand, the detection of banned contaminants at relevant concentrations in protected areas suggests that POPs re–volatilization is occurring from soil sources located in the neighbouring regions (Landers et al., 2008).

The Monitoring Network in the Alpine Region for Persistent and other Organic Chemicals (MonarPOP, http://www.monarpop.at/), was initiated in the Alps, operating from 2004 to 2006 and providing information on POPs' concentration along altitudinal profiles in air, needles and soil samples (Tremolada et al., 2009); biomonitoring was also carried out.

As the time series get longer and are more statistically robust, it is possible to discern how contaminant trends are affected by climate variability and possible changes in POPs' exposure pathways (WHO, 2003; Macdonald et al., 2005). Several studies aimed at improving our understanding of how global warming could undermine global efforts to reduce environmental and human exposure to POPs, demonstrating that the impacts of policy decisions on environmental levels will require continued monitoring of "legacy POPs" in both abiotic environments and in key biota. In the past few years a larger list of new chemicals detected in remote regions as the Arctic and mountain regions was observed. Measurement activity of new chemicals has a great relevance for the Stockholm Convention and for other national assessments of chemicals such as REACH, and for the assessments of potential health effects on Arctic ecosystems and humans (Wilson and Symon, 2009; Boethling et al., 2009; Muir and de Wit, 2010).

3. Implications of Environmental Change on POPs' Fate and Transport

As we introduced earlier in this paper, climate change involves changes in the mean value or changes in the variability of a climate property that persists for an extended time (Solomon et al., 2007). Starting from these alterations, climate change–oriented trends in POPs' environmental fate and transport may also be identified. Climate variability forecasted by climate change scenarios presented by the 4th IPCC assessment report (Solomon et al., 2007) shows that atmospheric temperature is increasing globally, this leading to the loss of ice–cover in forms of permafrost and ice–cap in Alaska, according to previous results (Macdonald et al., 2005). Furthermore, it was shown that the variability in sea–ice motion was correlated to the variations in the wind by the Arctic Oscillation (AO). During winter, wind anomalies associated with the positive AO index increases the advection of ice away from the North Pole (Eurasian and Alaskan coasts). Such advection enhances the production of thin ice, when the central pack ice moves away from the coastal ice creating a "flaw" in the ice surface (flaw leads) along the coast, and preconditions the sea–ice to be more prone to melt during the following spring and summer (Rigor and Wallace, 2004).

Since chemicals' environmental behaviour is driven by environmental variables, climate change may alter any aspect related to POPs' environmental fate and transport from POPs' emissions to the environment, to their transport and their degradation. The changes involved by climate fluctuations on POP's fate and transport are often investigated through the application of Eulerian multimedia fate and transport models both at global and regional scales (e.g. Gusev et al., 2005; Semeena et al., 2006; Lammel et al., 2009; Lamon et al., 2012), or through the application of Global Circulation Models (GCMs) (e.g. Stemmler and Lammel, 2011).

Climate fluctuations operate through climate variables such as temperature, wind speed, water and snow precipitation, solar radiation, atmospheric pressure, marine salinity, dry period, marine currents, ice cover, as well as interactions among these, like vegetation mass and type, organic carbon content, sediment resuspension and deposition, and population dynamics (Lamon et al., 2009a; Gusev et al., 2009). These variations drive temporally and spatially averaged exchanges of heat, momentum, and water vapour that ultimately alter re–volatilisation, transport and distribution patterns of POPs (Hung et al., 2005).

The Task Force on Hemispheric Transport of Air Pollution (HTAP) established by the UNECE for the planning and drafting the technical work necessary to estimate intercontinental transport of air pollution in the Northern Hemisphere, provides information to policy–makers and international organizations on issues related to long–range and intercontinental transport of air pollution and to serve as a basis for future cooperative research and policy action also assuming climate change effects on POPs' environmental fate and transport (TF-HTAP, 2010). The HTAP task force pursues these objectives through the application of global–scale multimedia mass balance models, that significantly improved scientific knowledge about how chemicals' emissions occurring in industrialized regions may affect contamination of pristine regions, such as the Arctic and mountainous regions (Wania, 2006; Hung et al., 2005; Gouin and Wania, 2007; TF-HTAP, 2010). These models are a key component in understanding the environmental dynamics of POPs; in fact they combine information about emissions, chemicals' properties and interactions with the environmental compartments to be merged in final results which can be evaluated against observations provided by monitoring studies.

Temperature is one of the main climate drivers in determining POPs' global distribution as it has a direct influence through affecting chemicals' half–life, partitioning, volatilisation and re–emissions, and indirectly, for instance through effects on hydroxyl radical formation process (Wania, 2005; Gouin and Wania, 2007; TF-HTAP, 2010). The potential of higher temperatures to enhance secondary emissions from previously contaminated environments through increased volatility, more rapid degradation and altered partitioning between phases, play an important role in global distribution of POPs (Lamon et al., 2009b). These secondary sources will dominate the total inventories of POPs that were largely used in the past, especially in remote regions due to their own nature of diffusive sources (Ma et al., 2004; Nizzetto et al., 2010; Lamon et al., 2012). In a secondary source–controlled world, physical–chemical processes such as burial and storage in terrestrial subsurface layers during organic carbon transformation, riverine export in dissolved and particulate form, and export of particle–bound chemicals by settling to deep sediments are

Table 2. *International agreements and POPs monitoring programs. The table shows a summary of existing monitoring programmes at regional and global scale, and international agreements and marine conventions; where existing, the relationship between conventions and the monitoring programs is also shown.*

		International Agreements	
Parties	**Name**	**Description**	
51 Parties from: European Union, North America, Eastern Europe, Caucasus and Central Asia	UNECE-LRTAP	United Nations Economic Commission for Europe (UN-ECE): agreement on a POPs protocol under the Geneva Convention on Long-Range Transboundary Air Pollution (LRTAP) from 1979. POPs Protocol (Aarhus Protocol) entered into force by the end of 2003	
177 Parties from United Nations	UNEP-Stockholm Convention	United Nations Environmental Program (UNEP): Stockholm Convention entered into force in May 2004	
Eight Arctic States: Canada, Denmark (including Greenland and the Faroe Islands), Finland, Iceland, Norway, Russia, Sweden and USA	Arctic Council	The Arctic Council is a high-level intergovernmental forum which addresses issues of sustainable development and environmental protection in the Arctic, formally established in 1996	
		Marine Conventions	
Parties	**Name**	**Description**	
AMAP, EMEP	OSPAR	Protection of the Marine Environment of the North-East Atlantic: The parties are Belgium, Denmark, Finland, France, Germany, Iceland, Ireland, Luxembourg, The Netherlands, Norway, Portugal, Spain, Sweden, Switzerland and United Kingdom since 1992	
AMAP, EMEP	HELCOM	The Helsinki Commission: Convention on the Protection of the Marine Environment of the Baltic Sea Area; intergovernmental co-operation between Denmark, Estonia, the European Community, Finland, Germany, Latvia, Lithuania, Poland, Russia and Sweden	
EMEP	MEDPOL	The Marine Pollution Assessment and Control Component of Mediterranean Action Plan; it is responsible for the follow up work related to the implementation of the LBS Protocol, the Protocol for the Protection of the Mediterranean Sea against Pollution	
		Global POPs monitoring programs	
Parties	**Name**	**Description**	**Type of sampling**
Canada, UNEP-Stockholm Convention, Arctic Council, NCP	GAPS	Global Atmospheric Passive Sampling study; started in 2004. More than 65 monitoring sites on 7 continents	Passive sampling
UNEP-Stockholm Convention, Arctic Council, NCP	AMAP	Artic Monitoring and Assessment Programme for: Canada, Denmark (including Greenland and the Faroe Islands), Finland, Iceland, Norway, Russia, Sweden and USA. Established in 1992.	High volume active air sampling, biota and humans sampling
OSPAR	CAMP	Comprehensive Atmospheric Monitoring Program.	

Table 2. *Continued*

Regional POPs monitoring programs			
Parties	**Name**	**Description**	**Type of sampling**
UNEP-Stockholm Convention, UNECE, Arctic Council	EMEP	European Monitoring and Evaluation Programme. Established in 1977.	High volume active air sampling and PUF foam, precipitation measurements
Canada, USA, UNEP-Stockholm Convention	IADN	Integrated Atmospheric Deposition Network around Great Lakes in North America. Started in 1990	Active air sampling, precipitation measurements
Canada, AMAP, Arctic Council	NCP	Canadian Northern Contaminants Program co-ordinates Canada's action on northern contaminants, including Persistant Organic Pollutants (POPs), both nationally and internationally (Canada and Russia). Started in 1991.	Active air sampling, biota and human sampling
UNEP-Stockholm Convention, GAPS, WHO; Coordinated by RECETOX	MONET-Africa	National Monitoring Network in Northern Africa; long-term monitoring of air quality at background locations: Canary Islands, Congo, DR Congo, Egypt, Ethiopia, Ghana, Kenya, Mali, Nigeria, Mauritius, Senegal, South Africa, Sudan, Togo, Zambia. Started in 2008	Passive sampling
UNEP-Stockholm Convention, GAPS, WHO; Coordinated by RECETOX	MONET-CEEC	National Monitoring Network in Central and Eastern European Region; Long-term monitoring of air quality at background locations: Czech Republic, Bosnia and Herzegovina, Estonia, Latvia, Lithuania, Romania, Serbia and Slovakia. Started in 2005.	Passive sampling
UNEP-Stockholm Convention, GAPS, WHO; Coordinated by RECETOX	MONET-PI	National Monitoring Network Pacific Islands (2006-2007); Long-term monitoring of air quality at background locations: Fiji Islands	Passive sampling
United Nations University; Institute for Sustainability and Peace (UNU-ISP)	EMGEAH	Environmental Monitoring and Governance in the East Asian Hydrosphere (EMGEAH) - Monitoring of POPs. Coastal waters of nine East Asian countries, namely China, India, Pakistan, Indonesia, Japan, the Republic of Korea, Malaysia, Singapore, Thailand, Vietnam and Philippines	Biomonitoring and water monitoring
US- EPA	WACAP	Western Airborne Contaminants Assessment Project (2002 to 2007); the contaminant situation at 20 national parks from the Arctic to the Mexican border	Passive sampling
UNEP-Stockholm Convention, UNECE, EMEP, EU	MonarPOP	Five countries (Austria, Germany, Italy, Slovenia, and Switzerland) established a network to assess the POP load of the Alps. From 2004 to 2006	Active and passive air sampling, biomonitoring, soil, water
Canada	Western Canadian Mountains	22 sites along three transects (Revelstoke, Yoho, and Observation, 6–8 sites for each transect) in the mountains of Western Canada	Passive air sampling, soil and needles
UK, UNECE-LRTAP	TOMPs	The Toxic Organic Micro Pollutants Network operates since 1991, currently collects samples at six sites across England and Scotland.	Andersen GPS-1 samples
GEF-UNEP	GEF-UNEP	Global Environmental Facility (GEF): Capacity Strengthening and Technical Assistance for the Implementation of Stockholm Convention National Implementation Plans (NIPs) in African Least Developed Countries (LDCs). Supporting the Implementation of the Global Monitoring Plan. Enabling activities for the Stockholm Convention on Persistent Organic Pollutants (POPs): Development of a National Implementation Plan in Developing Countries. Started in 2006.	Passive and active sampling

expected to represent the major active stores and sources of pollutants. Nizzetto et al. (2010) proposed that a better understanding of the evolution of anthropogenic and natural factors will enable the definition of future re–emissions of pollutants in the environment (Breivik et al., 2007).

Sea surface temperature (SST) and wind speed influence volatilisation rates, that will vary for different temperature and wind speed regimes, hence in different climatic zones and by climate change (Stemmler and Lammel, 2011). Wind speeds also play an important role in influencing POPs' global fate and transport, as the enhanced intercontinental transport of POPs (e.g. PCBs) is also a result of increased wind speeds, as it was investigated in several studies (MacLeod et al., 2005; Lamon et al., 2012). Seasonal temperature cycles that induce the chemical to undergo one or more transport and deposition cycles (hops) are related to atmospheric pressure patterns. For instance, enhanced transport of some POPs (e.g. PAHs) to the Arctic was observed in winter, in conjunction with the high atmospheric pressure characterising the Siberian winter, originating the Arctic haze event together with higher emissions in Eurasia during this period. This has provided important basis for the qualitative prediction, control, and management of regional air pollution problems TF-HTAP, 2010; Gusev et al., 2011).

Water and snow precipitation events involve washout, wet deposition of POPs from the atmosphere to surfaces (water and soil compartments). Changes in duration and intensity in seasonal precipitation may lead to changes in the spatial and temporal distribution of POPs wet deposition and their degradation products (Daly and Wania, 2005; Tremolada et al., 2008). An increase in heavy precipitation events and in floods has been recently observed, while the total amount of precipitation may be considered unchanged, or decreasing (Trenberth et al., 2007; Lenderink and Meijgaard, 2008; Lamon et al., 2009b).

The fate of POPs in the ocean depends on key processes related to transport by ocean currents, air–sea exchange (volatilisation and phase partitioning), degradation in sea water and partitioning and deposition with suspended particulate matter (Lamon et al., 2012).

Changes in light conditions and radiation intensity, that influence photochemical degradation, may change transport patterns of POPs (UNECE, 2010) also to the Arctic (Kallenborn et al., 2007), affecting POPs distribution, transformation and degradation processes (e.g., photochemistry) (Lamon et al., 2009a).

3.1. Main findings

Several studies addressed climate change influence on POPs' environmental fate and transport in order to elucidate the effects on their long–range transport. Mountain regions have been proposed as ideal sensitive places (Daly and Wania, 2005) besides Arctic region (Kallenborn et al., 2007; UNEP/AMAP, 2011), as long as these are not directly impacted by human activities, and are characterised by peculiar meteorological patterns, e.g., diurnal surface temperature change, precipitation rate and diurnal winds. Notwithstanding their remoteness from sources of pollution they still are contaminated by POPs. Alpine lakes also have been referenced to clarify the contribution of atmospheric long–range transport to the POPs' contamination–specifically PBDEs, HBCD, PCNs PCBs and DDT– (Bogdal et al., 2008; Schmid et al., 2011). First, Schmid et al. (2011) compared historical trends of some POPs and found that those banned, after showing a declining trend after their ban, exhibit an unexpected increase in concentrations. Such increase was accompanied with accelerated adjacent glaciers' melting. In contrast, lowlands lakes presented steadily decreasing levels since the 1970's (Bogdal et al., 2008). These studies supported the hypothesis that a closer link exists between the glaciers' melting and the released pollutants in sediments in proglacial lakes, making the ice–melting an important secondary source of POPs. Furthermore, a relationship between the amount of ice formed in a given period and the concentration of each pollutant: a relative small amount of ice formed at a period when environmental pollution was high can represent an important reservoir of contaminants, and results in an important release with ice melting. Other relevant secondary sources acting as POPs reservoirs are soil, vegetation, and sediments (Nizzetto et al., 2010).

Several studies addressed the global POPs' distribution, reporting temperature as one of the most relevant variables influencing both emission rates from primary and secondary sources, particle–phase partitioning, reaction rates (biodegradation, oxidation), air–surface exchange (volatilisation), air–soil exchange (deposition) and hydroxyl radical formation (Gusev et al., 2005; Ma et al., 2004; Jurado et al., 2007; Lamon et al., 2009b; Lamon et al., 2012). Wind speed is another environmental variable that is associated with certain atmospheric POPs' fluxes. Lamon, et al. (2009b) investigated the processes related to climate patterns and interannual variations of POPs' concentrations. Their results show that climate change may affect emissions of POPs by enhancing volatilisation and re–volatilisation (Hung et al., 2005), and lead to increased atmospheric emissions especially in remote areas.

Other parameters related to POPs' environmental distribution are quantity, quality and spatial variation of precipitations. These climate parameters associated with various climate patterns would also affect scavenging by absorption to snow and thence transport and deposition of organic pollutants (Daly and Wania, 2005; Gabrieli et al., 2010; UNEP/AMAP, 2011).

In recent studies, it was possible to detect evidence of POPs' revolatilisation induced by regional warming (Ma et al., 2011). Increasing trends of HCB and PCBs that have been observed in Arctic air could be attributed to their tendency of volatilisation from water to air.

3.2. Possible next steps in regulating climate change effects on POPs' environmental behaviour

In the previous paragraphs we reported several studies where climate variables influence Predicted Environmental Concentrations (PECs) for POPs.

The UNEP Stockholm convention addresses how to classify POPs by considering substances' physical chemical characteristics. Recently, the UNEP/AMAP expert group reported on climate change implications on the management of POPs and found that climate change may have a potential direct impact on present and future management of POPs, and is likely to enhance the exposure to POPs in some regions (UNEP/AMAP, 2011). However, the way to go for obtaining an integration of climate change effects on POPs' environmental behaviour in chemicals' regulation is still missing. A thorough integration between the different international initiatives on the management of POPs would help to define a reference framework for POPs' and other chemicals' assessment also taking in consideration climate change.

As described earlier in this paper, REACH is the reference guidance in Europe for the definition of procedures in the assessment of exposure to PBTs and vPvBs (ECHA, 2011), and thus providing assessment and analysis on chemicals' health or environmental effects (ECHA, 2011).

Chemicals' legislation may take into consideration climate change by giving more emphasis to the variability of environmental parameters that should be selected on the basis of sound studies on climate change effects on POPs' environmental behaviour. Also, more emphasis should be given to uncertainty analysis in the definition of PECs.

When it is necessary to take into account climate change in estimating the environmental behaviour of chemicals, uncertainty plays a key role, and this could be one key aspect to be better addressed by regulation. Climate change scenarios are characterized by different typologies of uncertainties (IPCC, 2005), and these should be integrated with the uncertainties related to chemicals' modelling. A dialogue between scientists and regulators could be oriented towards the definition of probability distributions of input climate parameters in chemicals' multimedia models, and/or the definition of a sensitivity analysis where special attention is dedicated to climate parameters. This is still a challenging issue, as no guidance on how to estimate uncertainty in exposure modelling exists, and it is actually difficult even to compare results and uncertainty estimates from similar models. For instance, the REACH regulation addresses how to deal with uncertainty estimation in the evaluation of the risk factor, but does not provide a detailed insight on the uncertainty estimation on PECs in exposure modelling. This may be the next step towards taking into consideration environmental variability and change in chemicals' regulation.

4. Conclusions

In the previous sections we have considered how POPs' environmental distribution is influenced by emissions' spatial distribution, chemicals' physical–chemical properties, and global and regional atmospheric and oceanic circulation patterns (i.e. wind speeds, precipitation, interactions with the Earth's surface, ocean currents, organic carbon content, and biological pathways) (AMAP, 1998). Such variability is affected by climate change, that in turn influences POPs' environmental fate and transport.

From the case studies reported in this review paper, we can infer that variables with a pronounced effect on POPs' environmental fate and transport are temperature, affecting also revolatilisation from POPs' reservoirs, wind speeds and spatial distribution of primary and secondary pollution sources in each environmental compartment. Since primary and secondary emissions play a key role in describing POPs' mass balance it is necessary to collect information on POPs' emissions and to build updated emissions' inventories taking into account historical emissions and performing simulations in order to define the location and the amount of future sources of pollution. This objective is to be considered partially achieved, thanks to the activities organised under the Task Force on Emissions Inventories and Projection (TFEIP), consisting of the active involvement of public authorities and of the communication between policy makers and science experts.

Snow and ice melting effects on POPs deserve special consideration, as several studies report that mountain environmental quality is impacted by POPs' transport in such remote environments that act as a sink similarly to the Polar regions. For this reason, mountain regions can be identified as the most sensitive and important ''early warning'' sites to assess climate change effects on POPs' fate and transport.

Continuous monitoring activities are important as they could push the identification of new and unsuspecting primary or secondary sources. Moreover, continued monitoring supports fate and transport modelling activities aimed at identifying and characterising long–range transport from industrial and agricultural emissions (Kallenborn et al., 2007).

Considering that several modelling exercises demonstrate that long–range transport potential increases under a climate change perspective, policy actions should be more restrictive in managing production and use of POPs and POPs–like chemicals.

Acknowledgements

This paper was supported by the GEMINA project, funded by the Italian Ministry for the Environment, Land and Sea (MATTM), and the EC FP7 (2007–2013) Marie Curie EPSEI project (grant agreement no. 269327).

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