Climate change and environmental concentrations of POPs: A review

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ABSTRACT

In recent years, the climate change impact on the concentrations of persistent organic pollutants (POPs) has become a topic of notable concern. Changes in environmental conditions such as the increase of the average temperature, or the UV-B radiation, are likely to influence the fate and behavior of POPs, ultimately affecting human exposure. The state of the art of the impact of climate change on environmental concentrations of POPs, as well as on human health risks, is here reviewed. Research gaps are also identified, while future studies are suggested. Climate change and POPs are a hot issue, for which wide attention should be paid not only by scientists, but also and mainly by policy makers. Most studies reported in the scientific literature are focused on legacy POPs, mainly polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), polychlorinated biphenyls (PCBs) and pesticides. However, the number of investigations aimed at estimating the impact of climate change on the environmental levels of polycyclic aromatic hydrocarbons (PAHs) is scarce, despite of the fact that exposure to PAHs and photodegradation byproducts may result in adverse health effects. Furthermore, no data on emerging POPs are currently available in the scientific literature. In consequence, an intensification of studies to identify and mitigate the indirect effects of the climate change on POPs fate is needed to minimize the human health impact. Furthermore, being this a global problem, interactions between climate change and POPs must be addressed from an international perspective.

Keywords:

Climate change

Persistent organic pollutants (POPs)

Environmental fate and transport

Polycyclic aromatic hydrocarbons

Legacy POPs

Scientific literature

1. Introduction

The reconstruction of the Earth's historical climate trends has demonstrated that climate is constantly changing, showing peculiar oscillations at different time scales. However, in recent years a particular climate acceleration has been observed over the last decades. The assessments of the Intergovernmental Panel on Climate Change (IPCC) have evidenced that, due to increasing greenhouse gases, the Earth's climate is substantially changing (IPCC 2013). A number of studies have confirmed that the global mean temperature increased by 0.6±0.2ºC during the 20th century (IPCC, 2001). In addition, the air temperature is projected to increase 1.8-4.0ºC by the end of the 21st century, under a range of probable greenhouse gas emission scenarios, being high latitudes those more severely affected (Noyes et al. 2009). The Mediterranean region is a vulnerable zone, where a number of climate-related events, such as higher sea levels, increased frequency of extreme climatic events including intense storms, heavy rainfall events and droughts (Kusangaya et al. ; McBean and Ajibade 2009) are probably going to occur. Moreover, the increase in the surface UV-B radiation induced by ozone depletion has received wide attention as an environmental issue of great concern (Watanabe et al. 2011). Hence, climate change is an increasingly urgent problem, with wide consequences for the environment and life on Earth (Kim et al., 2014). The global change effects will impact not only animal species and ecosystem processes (Moe et al., 2013), but they will also alter the degree of human exposure to pollutants, changing the risks in the future (Balbus et al., 2013). A number of studies have identified the expected degree of these impacts in various regions. Instrumental observations suggest that the Arctic has been changing faster than any other region on the Northern hemisphere over the past decades (Serreze and Barry 2011; van der Bilt et al., 2015), being evidenced by melting of ice caps and glaciers, as well as the rise of sea level and temperature (Jayawardena, 2014). Moreover, the Mediterranean basin is considered one of the most vulnerable regions of the world to climate change. According to (IPCC 2013), air temperature is expected to increase up to 4ºC (Sánchez-Canales et al. 2012; Schröter et al. 2005; Bangash et al. 2012; Marquès et al. 2013; Terrado et al. 2014). This region is lying in a transition zone between the arid climate of North Africa and the temperate and rainy climate of central Europe, being therefore affected by interactions between mid-latitude and tropical processes (Giorgi and Lionello 2008).

Persistent organic pollutants (POPs) have become chemicals of concern during the last decades due to: a) their considerable resistance to degradation, b) their ability to be transported over long distances from sources by air and ocean currents, in a process known as Long Range Atmospheric Transport (LRAT), c) their potential to be bioaccumulated through terrestrial and aquatic food webs, to levels that may result in adverse health effects for animals and humans (Hung et al. 2013), and d) their potential toxic effects such as immunotoxicity, neurotoxicity, developmental toxicity, carcinogenicity, mutagenicity, and endocrine disruption (Chao et al., 2014; Domingo, 2012a; Gascón et al., 2013; Gasull et al., 2013; Kim et al., 2013(WHO 2011). In 2001, the Stockholm Convention on Persistent Organic Pollutants elaborated a first list of POPs whose emissions and/or production must be eliminated, or at least notably reduced. That list included a variety of POP candidates: organochlorine pesticides (DDT, aldrin, chlordane, dieldrin, endrin, heptachlor, hexachlorobenzene (HCB), mirex, and toxaphene), as well as polychlorinated biphenyls (PCBs) and polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) (commonly known as 'dioxins'). However, this is a dynamic list, and therefore, more compounds with similar properties have been added throughout time. In May 2009, a new set of chemicals, including other pesticides (chlordecone, α-hexachlorocyclohexane, β-hexachlorocyclohexane, lindane, pentachlorobenzene and pentachlorobenzene), polybrominated biphenyls (PBBs) (hexabromobiphenyl, hexabromodiphenyl ether and heptabromodiphenyl ether, tetrabromodiphenyl ether and pentabromodiphenyl ether), as well as perfluoroalkyl substances (perfluorooctanesulfonic acid (PFOS) and its salts, and perfluorooctanesulfonyl fluoride (PFOSF)), were also included in the list. Furthermore, endosulfan and hexabromocyclododecane were added in the Fifth and Sixth Conference of Parties, held in 2011 and 2013, respectively (Jennings and Li, 2015).

Parallel initiatives have highlighted the PBT properties of other chemical pollutants. For instance, the Executive Body of the United Nations Economic Commission for Europe (UNECE) included four polycyclic aromatic hydrocarbons (PAHs) (benzo[a]pyrene, benzo[b]flouranthene, benzo[k]fluoranthene, and indeno[1,2,3-cd]pyrene) in the Protocol on POPs, signed in 1998 in Aarhus (Denmark). PAHs were also included in the original list of POPs by the UNECE's Convention on Long-Range Transboundary Air Pollution (CLRTAP). The above 4 PAHs are persistent, bioaccumulative, toxic, and can be transported to long distances through the air (LRAT). Consequently, they can be also considered as POPs. Thus, they were included

in a list of 16 priority substances by the 1998 Aarhus Protocol on POPs together with eleven pesticides, two industrial chemicals, as well as two other by-products/contaminants (UNECE, 2014). The Protocol banned the production of some substances (aldrin, chlordane, chlordecone, dieldrin, endrin, hexabromobiphenyl, mirex and toxaphene), while others (DDT, heptachlor, hexaclorobenzene, PCBs) were scheduled for elimination at a later stage. Moreover, the use of some products such as DDT, hexachlorocyclohexanes (HCHs) and PCBs, is severely restricted and provisions for dealing with the wastes of substances that will be banned are included. Finally, the Protocol obliges Parties to reduce their emission of PCDD/Fs, PAHs and HCB below their levels in 1990.

The fate and behavior of POPs have attracted considerable political and scientific interest, particularly when local releases have resulted in dispersed contamination far from source regions (Paul et al. 2012). The transport distance and the number of air-surface exchange episodes depend on the surface characteristics (e.g. soil, water, vegetation, etc.), as well as the physical-chemical properties of the compound. Thus, persistent chemicals, with a lower vapor pressure, will be preferably deposited in areas closer to the emission source, while those with a higher vapor pressure are more easily transported far away. In addition, there are other mechanisms and factors which influence the distribution of POPs in the atmosphere. These are the capacity of the environmental compartments to accumulate or degrade POPs, the general atmospheric patterns, and the kinetic of the air-surface exchange, among others.

One of the consequences of climate change that has recently attracted some interest is its potential to alter the environmental distribution and biological effects of chemical toxicants (Noyes et al. 2009). Environmental variables such as temperature, wind speed, precipitation, and solar radiation, have influence, either directly or indirectly, on the environmental fate and transport of POPs (Gusev et al., 2012). As climate change will obviously alter most of those factors to varying degrees, it is generally accepted that climate change can influence every step along the fate, transport and distribution pathways of semi-volatile organic chemicals, including PAHs and POPs (Cai et al. 2014; Kallenborn et al. 2012; Schiedek et al. 2007; Teran et al. 2012). Temperature is one of the key meteorological parameters that is able to impact more severely the global distribution of POPs in the environment (Dalla Valle et al. 2007). According to the results of multicompartment chemistry-transport models, the degradation rates of POPs in the environment are estimated to increase by a factor of two to three for every 10ºC

increment (Lammel, 2001; Macdonald et al., 2005). Therefore, global warming is probably influencing the environmental behavior of POPs. It enhances the volatilization from primary and secondary sources, and influencing their partitioning between soil, sediment, water and atmosphere, including air-surface exchange and wet/dry deposition (Noyes et al. 2009; Teran et al. 2012; Armitage et al. 2011). An increase of rainfall can cause a raise of POP deposition onto soil. The more frequent storm surges may enhance the mobilization of chemicals stored in the soil compartment, which can be transported by land runoffs, making them available to the aquatic organisms (Dalla Valle et al. 2007). In turn, cold temperatures can induce their deposition and accumulation in Arctic environmental media, resulting in the so-called cold-trapping effect (Rahn and Heidam 1981; Perrie et al. 2012). It has been also observed in other recent studies that the fate of POPs depends on the meteorological conditions, and therefore climate change will modify their concentrations and trends in the Arctic (Perrie et al. 2012), ultimately affecting the rest of the world. In addition to the Arctic, which is already experiencing substantial changes (Armitage and Wania, 2013), other world areas such as the Alps or the Mediterranean, are also sensitive to POP deposition. For instance, the central and eastern Mediterranean is a receptor area for POPs emitted in western, central and Eastern Europe, particularly during summer (Mulder et al., 2015). Furthermore, the role of climate change and eutrophication on POP dynamics is a topic that needs further consideration by scientists (Wania and Mackay, 1999).

Despite of the lack of knowledge of climate change impacts on the POP occurrence, it has been suggested that the temperature increases should cause a faster degradation of these chemicals in the aquatic ecosystem, resulting in a reduction of the dietary exposure to POPs (McKone et al. 1996; Macdonald et al. 2005; Ma et al. 2004; Bard 1999)). However, these pollutants could be also transported to higher latitude areas where wet deposition would lead to a potentially elevated POP dietary intake among exposed northern and indigenous communities (Sonne et al., 2014). Health impacts of POPs are not immediate. They have usually resulted from chronic, cumulative and long-term exposure to one or more substances, being the major exposure route non-atmospheric. Hence, POPs do not usually cause respiratory health effects. In contrast, ingestion and bioaccumulation are routes of concern. Dietary exposure seems to be the most contributive exposure pathway for POPs and other semi-volatile chemical contaminants (Domingo et al., 2008, 2012b,c; Martí-Cid et al., 2010; Martorell et al., 2012; Perelló et al., 2015). Moreover, although toxic effects of POPs are elucidated at

multiple endpoints, they are not well defined when evaluating mixtures of POPs (Hung et al. 2013).

In the present paper, we have reviewed the state of the art regarding the influence of climate change on the environmental concentrations of POPs. This paper was aimed at gathering publicly available information on this topic. It should help to identify research gaps in the design of future environmental and health studies, considering plausible changes in human health risks, which are associated to global warming.

2. Climate change impact on POPs

The scientific literature on the potential effect of climate change on POPs was reviewed by using the Scopus database (www.scopus.com). A first selection of papers was performed by using the terms: "climate change and POPs" or "global warming and POPs" in their title, abstract, or keywords. Afterwards, a specific choice was carried out by using, as keywords, "climate change" or "global warming", as well as the name of each individual chemical. POPs were selected considering the current list addressed in the Stockholm Convention.

Finizio et al. (1998) reported for the very first time the potential impact of climate change on POPs, and more specifically on some organochlorine pesticides (DDT, HCHs, chlordane, toxaphene, aldrin). Other halogenated chemicals such as PCDD/Fs and PCBs, were also mentioned. The authors remarked that the long-range transport of POPs in the environment was largely dependent on the environmental conditions, particularly air temperature, particulate air matter and wind direction/speed. Consequently, a change of conditions might mean notable consequences for POP distribution. It was concluded that a change in the global atmospheric conditions, and therefore the average condition for a region, can influence the biogeochemical cycle of POPs and then their presence in a specific ecosystem. According to Finizio et al. (1998), there were no studies reporting these interactions at that moment. However, they suggested that some scenarios could be simulated with the use of "global chemodynamic models" as those previously developed by Wania and Mackay (1995).

2.1. Pesticides

Pesticides form the larger group of POPs included in the Stockholm Convention. Consequently, a considerable number of data from different perspectives are currently available. Ma and Cao (2010) aimed at quantifying the perturbations of POPs as result of the climate change. A perturbed air-surface coupled model was developed to simulate and predict perturbations of POP concentrations in various environmental media under several climate change scenarios. The selected chemicals were α- and γ-HCHs, as well as HCB and PCB-153 congener. All POPs exhibited a strong response to specified climate change scenarios, as shown by their high concentrations perturbations in air. In the air-soil system, the model predicted 4-50% increases in the air concentrations of these chemicals associated to a potential increase of 0.05-0.1ºK/yr in the air temperature. The authors estimated that a 20% increase/decrease in precipitation may result in a 53% and 4% decrease/increase, in perturbed air concentration of γ-HCH and α-HCH, respectively (Ma and Cao, 2010).

Wöhrnschimmel et al. (2013) applied a global-scale multimedia fate model to analyze and quantify the impact of climate change on emissions and fate of POPs, and their transport to the Arctic. Two climate scenarios (base-scale and IPCC-based SRES-A2) were used to characterize the evolution over time of two well-characterized POPs (α-HCH and PCB) after an air temperature increase of 2.0-5.4 ºC. Regarding to climate change, four different spatially and temporally resolved generic emission scenarios were defined, covering the period 2020−2050. The model was run from the first year of emission (around 1950s) until 2100. The temporal evolution followed a plausible pattern for substances that were thought to be introduced in 2020, and successfully merchandised up to the saturation of the market in 2035, and then phased out over the following 15 years. This phase-out would be linked to concerns on their hazard for the human health and/or the environment, or to their replacement in the market. These scenarios were used after considering both current climate conditions and with the climate change parameterization of *BETR Research model* described by MacLeod et al. (2011). Beyond temperature, other parameters of the model were air, land, and ocean surface temperatures, precipitation, atmospheric and oceanic circulation, land-ice and sea-ice cover, and organic carbon content in soils. Reported results from the simulation showed that, in the atmosphere, according to the simulations, the maximum α-HCH value was 10^3 pg/m^3 in 1980, decreasing to 10^{-4} pg/m^3 in 2100. Reported α-HCH concentrations in ocean waters had a similar behavior, showing their maximum level

around 1990, with a reported concentration of 1 ng/L. In turn, the minimum value (10^{-5} ng/L) was estimated to occur in 2010.

Sun et al. (2005) attributed the increase of HCHs in coastal sediments to the glaciers meltwater derived from the regional warming from the early 1970s. Accumulation flux profiles and temporal trends of HCHs were determined through the analysis of two lake cores (NR and AX) collected from Niudu Lake in King George Island (West Antarctica). NR was a core under the influence of glacier meltwater, while AX was the control core. With respect to DDT, NR core showed an abnormal peak around 1980s in addition to the expected one in 1960s. On the other hand, the accumulation flux of DDT in AX core showed a gradual decline after the 1960s peak. This difference was most probably caused by regional climate warming and the resulted discharge of the DDT stored in the Antarctic ice cap, into the lakes in the Antarctic glacier frontier.

The revolatilization of α-HCH, DTT and cis-chlordane deposited in water and ice sinks was investigated by Ma et al. (2011). The records of their concentrations in Arctic air since the early 1990s were analyzed, and further compared with results from modelled simulations of the climate change effect. A correlation analysis was used to detect any evidence of POP revolatilization in the Arctic linked to regional warming. Strong correlations between POP air concentrations and mean surface air temperatures/sea-ice extent indicated the potential volatilization from secondary emission sources/reservoirs in water, snow, ice and land across the Arctic. POPs had been remobilized into the Arctic atmosphere over the past two decades as a result of climate change, confirming that the Arctic warming could undermine global efforts to reduce environmental and human exposure to these toxic chemicals.

To assess time trends and recycling of DDTs and HCHs, Cheng et al. (2014) analyzed the levels of these pollutants in lake sediment cores from five critical regions in the Tibetian Plateau. A recent increase of both chemicals was found, likely due to the retreat of glaciers in response to climate warming. In the past 30 years, and because of the climate warming, glaciers have shrunk more than 6606 km^2 on the entire Tibetan Plateau, with the greatest retreat since the mid-1980s. Hence, glacier and snow melt due to climate warming is able to release stored contaminants accumulated during years of higher transport of such pollutants to this region. Consequently, a warmer climate is expected to enhance the amount of glacial water discharge into lakes and depositing more DDTs into lake sediments. In fact, these closed-basin lakes fed mainly by glacier

meltwater in particular became a more sensitive monitor of global warming, in terms of temporal trends of organochlorine pesticides (OCPs).

Bogdal et al. (2009) also reported a decrease in the usage and emissions of the OCPs in Switzerland in recent decades. The main reason was the ban of DDT in that country in 1972, and that of HCB and γ-HCH, dieldrin and heptachlor in 1986. Results obtained by means of sediment cores from Lake Oberaar (Switzerland) indicated low fluxes of pesticides in the deepest sediment sample, dated from the early 1950s. From late 1990s onwards, the fluxes of pesticides, as well as those of other compounds (PCDD/Fs, PCBs, as well as polychlorinated naphthalenes) clearly increased. When assessing the supposed accelerated release of DDT in lake Oberaar due to ice and snow melting, the same authors found similar results to other POPs, such as PCDD/Fs (Bogdal et al. (2010).

Similar findings were also reported in different mountainous watersheds across a broad latitudinal, longitudinal and altitudinal range in the Canadian Cordillera in British Columbia and the Yukon, western Canada (Elliott et al., 2012). This bioaccumulation study investigated the temporal trends of DDT concentration in osprey eggs from those areas, which were in agreement with some modeled predictions of release from melting glaciers due to climate change. Predictions from the modeled dynamics of POPs released by a Swiss glacier were coincident with the apparent temporal trends observed in ospreys (Bogdal et al. 2010), indicating that previous glacial melting might have been a factor influencing contaminant trends in western Canada.

Studies on the effects of the climate change on POP dynamics in the marine environment also exist. Borgå et al. (2010) applied a bioaccumulation model to calculate the effect of two different climate change scenarios on the pelagic marine food web of the Arctic. The authors introduced a factor of change (i.e., the ratio of pollutant levels in a future climate scenario vs. current levels) to describe bioaccumulation and contaminant concentration changes in a projected future climate for each food web organism, and for 3 different pollutants (γ-HCH, PCB-52, and PCB-153). Two different scenarios were defined: 1) temperature in water and air increased by 2.0ºC, and 2) raise of 4.0ºC in both parameters. It was found that γ-HCH did not biomagnify. In turn, PCB-52 showed a higher degree of biomagnification. In addition, the modeled and measured biomagnification values of PCB-153 were even higher than those corresponding to PCB-52, with values increasing with the trophic position. γ-HCH showed the lowest and least spread in magnitude of the individual process rates and parameters. The effect

of increased temperature on the octanol-water (K_{ow}) and octanol-air (K_{oa}) partitioning coefficients, as well as the decreased lipid content, were the two most influential parameters resulting in reduced bioaccumulation on a wet weight basis. It was concluded that increased temperature would reduce the overall bioaccumulation of PCBs in Arctic marine food web.

Hallanger et al. (2011) assessed the differences between Arctic and Atlantic fjord systems on the bioaccumulation of POPs in zooplankton. Samples of zooplankton and seawater were collected from Liefdefjorden and Kongsfjorden, Svalbard, Norway. In zooplankton, the predatory species tended to have higher values of POPs than other species, while the lowest concentrations of POPs were found in the herbivorous *Calanus* species. ΣPCBs and Σpesticides were higher in Kongsfjorden than in Liefdefjorden. Differences in POP concentrations were assumed to be due to fjord specific characteristics, such as ice cover and timing of snow/glacier melt. Hence, it was difficult to conclude where there were Artic vs. Atlantic specific differences, and to extrapolate these results to possible climate change effects on accumulation of POPs in zooplankton.

McKinney et al. (2015) reviewed the ecological impacts of global climate change on the pathways of POPs and mercury, as well as their exposure in Arctic marine ecosystems. Most of the reviewed studies reported changes in POP concentrations in biological tissues linked to climate change-induced changes in species trophic interactions, particularly in relation to sea ice changes. However, the influence of changing trophic interactions on POP levels and trends varied widely in both magnitude and direction.

Recently, O'Driscoll et al. (2014) simulated, by means of HAMSOM and FANTOM models, the fate and cycling of γ-HCH and PCBs in the North Sea in the 21st century. Sediment concentrations of γ-HCH were estimated to be reduced in 2015, with respect to 2005 values as a consequence of the lower dry gas deposition. Although these authors concluded that the influence of climate change on those two POPs was small, the increased number and magnitude of storms in the 21st century will give place to POP resuspension and revolatilization processes. On the other hand, Morselli et al. (2014) created a combination of a dynamic fate model, and a hydrological module, capable of estimating water discharge and snow/ice melt contributions on an hourly basis. The resulting model, which was applied to the case study of the Frodolfo glacier-fed stream (Italian Alps), was fed with levels of PCBs and p,p'-DDE in stream water, being

366 available four macroinvertebrate groups. The model showed to be appropriate to
367 estimate pollutant concentrations under diverse climate change scenarios.
368 According to climate change predictions, it is well established that the probabilities
369 of flooding will increase (Wu et al., 2015). Flood events will make easier the sediments
370 resuspension. It means that contaminants contained in polluted sediments will be more
371 easily transferred to the surrounding water. Smit et al. (2009) carried out a laboratory
372 experiment where flood events were simulated in a reactor to estimate the desorption of
373 dieldrin from field aged sediments. The authors concluded that the concentration
374 gradient plays a major role in desorption, while mass transfer was kinetically hindered
375 within the sediment particles.
376 Riou et al. (2012) assessed the influence of the increase of salinity in an estuary, as a
377 consequence of the severe droughts induced by climate change in presence of waterbone
378 DTT in a Tilapia species. The experiment was conducted with young adults of
379 *Sarotherodon melanotheron*, hatched and grown in constantly aerated freshwater at
380 29ºC. *S. melantheron* was reported to be very resistant to waterborne DDT
381 contamination. However, it was brought clear evidence that this pesticide affects the gill
382 multi-functionality at different salinities. Although it seemed very resistant to short-term
383 waterborne DDT contamination, the resulting alterations of the gill issue, cells and
384 enzymes, might affect longer term respiration, toxicant depuration, and/or
385 osmoregulation in highly fluctuating salinities.
386 In a recent investigation, Komprda et al. (2013) assessed the influence of climate
387 and land use change on the potential re-emission of organochlorine pesticides (HCB and
388 DDE, as the prevalent DDT metabolite) from background and agricultural soils of the
389 Czech Republic. The studied region presented a relatively large portion of the land
390 covered by forest (32.6%). Regarding the influence of temperature change on POP
391 emissions, an increase of air temperature by 1ºC resulted in an increase of the total
392 yearly volatilization flux by approximately 7.8% and 8.5%, for HCB and DDE,
393 respectively, in all land use types. Data about the influence of land use change on POP
394 emissions showed that the arable-to-grassland scenario had a strong influence on
395 volatilization fluxes, resulting in a decline of secondary emission of -7.5%, for both
396 POPs, at all altitudes. These results showed that the potential increase of emissions
397 associated with increased temperature under climate change can be completely
398 neutralized by projected changes in land use. The study also indicated that an increase
399 of 1ºC in air temperature would produce an increase of 8% in the averaged total

volatilization flux. However, this effect could be neutralized by a change of land use of 10% of the arable lands to grassland or forest (Komprda et al. (2013).

Land use aspects are highlighted as an important issue to be considered in future assessments of climate change impacts on POP fate and distribution. The first outcomes of EU ArcRisk project on human health impacts in the Arctic owing to climate-induced changes in contaminant cycling were reported by Pacyna et al. (2015). They highlighted the need to characterize better the primary and secondary sources of POPs, as well as to quantify current and future releases of POPs from these sources, for a better prediction of the environmental exposure to these contaminants. Furthermore, not only direct effects of climate change (e.g., changes in temperature, ice and snow cover, precipitation, wind speed and ocean currents) on contaminants fate and behavior but also indirect effects (e.g., alterations in carbon cycling, catchment hydrology, land use, vegetation cover, etc.) should be considered. It has been stated that the climate change has the potential to impact on the usage patterns of chemicals (e.g. pesticides) through land-use change (Paul et al. 2012). For example, agricultural land use may be forced to migrate due to alterations in temperature, precipitation or sea level, which may indirectly change the amount and dose of chemicals applied in the field.

2.2. Polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs)

Dalla Valle et al. (2007) applied a dynamic multimedia model to selected POP congeners to simulate the effects of different climate change scenarios on their distribution and fluxes over the next 50 years in the Venice Lagoon (Italy). A level IV dynamic model (Mackay 2001), considering five compartments (air, soil, sediment, water, and suspended particulate matter), was developed and applied to this area. 2,3,7,8-tetrachlorodibenzofuran (2,3,7,8-TCDF) and 1,2,3,4,7,8-hexachlorodibenzofuran (1,2,3,4,7,8-HxCDF) were the selected PCDD/F congeners. Three different climate change scenarios (A, B, C) were tested, in accordance to the climate change scenarios envisaged by the 2001 IPCC climate change assessment report. Potential differences of PCDD/F concentrations in sediments and suspended particulate matter were observed among the three scenarios, showing a factor of around two between final levels in the two extreme scenarios. In turn, water concentrations of PCDD/Fs decreased in the same way, independently of the climatic conditions. Modelling results suggested that although global warming may have the potential of

reducing the environmental levels of these chemicals, probably it enhances their mobility, and hence, their potential for long-range atmospheric transport.

Focused on the water compartment, Carere et al. (2011) predicted the effects of climate change on the chemical quality in lakes species. The results showed an enhanced capacity of bioaccumulation of PCDD/Fs and dioxin-like PCBs (dl-PCBs) in an expected warming world. Thus, concentrations of POPs in fishes showed significant temperature correlations. An important aspect of POP cycling in these environments concerned the extent to which these pollutants may be remitted to atmosphere in the snow/ice pack, or percolated to soils and water bodies, particularly during snow melt. Temperature was found to be an important driver to the global cycling of POPs, through its influence on emissions from primary and secondary sources, gas-particle distributions, reaction rates, air-surface exchange, and global transport.

Regarding freshwater, Bogdal et al. (2009) hypothesized about a possible release of legacy pollutants from melting Alpine glaciers and their relevance. Two sediment cores retrieved in 2006 from Lake Oberaar (Switzerland) were extracted and further analyzed for PCDD/Fs, PCBs, DDT and their transformation products. The results revealed a consistent trend for all persistent organochlorinated compounds, showing a peak in 70s and an increase since 2000. In a subsequent investigation performed by the same research group, the release of POPs from Alpine glaciers was assessed in Lake Oberaar (Switzerland) by using a dynamic multimedia mass balance model (Bogdal et al. (2010). It was concluded that the effects of the climate warming will accelerate the release of previously deposited POPs.

The results of one of the most meaningful studies on climate change effects over the fate and transport of PCDD/Fs were recently reported (Chi et al. (2013). Specifically, extreme weather events (winter monsoon, southeast biomass burning, and tropical cyclone -typhoon-) were explored in Taiwan. During the winter monsoon period, the quantity of PCDD/Fs absorbed onto air total suspended particles was found to increase. Therefore, the monsoon was not only found to bring cold air, but also to transport air pollutants and dust over long distances, from mainland China to Taiwan. The authors demonstrated the effect of typhoon events on the long-term remobilization of PCDD/Fs, as well as supported the hypothesis that such events would have the potential to remobilize previously deposited pollutants. Consequently, climate change will alter the primary and secondary release of PCDD/Fs, mainly because of higher wind speeds. Stronger air circulation will increase the airborne transport to downwind locations (e.g.,

Taiwan) from the main emission areas of the Asian continent. The possibility of an enhanced frequency and intensity of extreme weather events will also lead to increased release and a higher risk of remobilization of PCDD/Fs from soils, sediments, and other reservoirs of PCDD/Fs.

2.3. Polychlorinated biphenyls (PCBs)

In addition to PCDD/Fs, Dalla Valle et al. (2007) also reported the reduction of the environmental concentrations of PCB-118 and PCB-180 congeners in a moderate climate change scenario. Concentration changes of POPs in sediments, water and suspended particulate matter of the Venice Lagoon (Italy), between 2000 and 2050, were simulated and compared to a baseline situation. The authors noted that environmental concentrations might differ by a factor of two in a moderate climate change scenario, compared to a situation with stable climate from 2000 to 2050. However, these results also suggested that global warming might have the potential of reducing the environmental levels of these chemicals, enhancing their mobility and their potential for long-range atmospheric transport.

Wöhrnschimmel et al. (2013) applied a global-scale multimedia fate model to analyze and quantify the impact of climate change on the emissions and fate of POPs, and their transport to the Arctic, being PCB-153 the selected pollutant. Based on the model, there will be an atmospheric reduction from 5 to 10^{-3} pg/m^3 between 1980 and 2100. According to the simulations, in ocean waters, PCB-153 concentration will be also decreased in 2100 with respect to data of 1980. Recently, Cabrerizo et al. (2013) assessed how changes in soil biogeochemistry driven by climate perturbations may increase to a larger degree the soil fugacity capacity of POPs. The potential perturbations of climate change on the remobilization and reservoirs of PCBs in the Antarctica, were explored. A climate change related increase of 1ºC in air temperature was estimated to increase the Antarctic atmospheric burdens of PCBs by 21-45%. In addition, a concurrent increase of 0.5% of solid organic matter will counteract the influence of warming by reducing the POP fugacity in soil. A 1ºC increase in Antarctic temperatures will induce an increase of soil-vegetation organic carbon and associated POP pools by 25%, becoming a net sink of POPs. Therefore, up to 70 times more POPs than the amount remobilized to the atmosphere, will be trapped.

Lamon et al. (2012) developed and applied a Level III fugacity model to estimate the current mass balance of PCBs in the Adriatic Sea, and to examine the effects of climate change on the distribution of these pollutants. The model, which differentiated 3 bulk compartments (sediment, coastal water, and atmosphere), assessed the influence in the variation of up to 8 environmental parameters on the environmental fate and transport of PCBs 52, 138 and 153. Two scenarios were considered: 20CE (1990 as 20th Century Scenario) and A1B (2100, as forecast under conditions described in the Special Report on Emission Scenarios by IPCC). Modeled fugacities of PCBs in air, water and sediments of the Adriatic Sea were in good agreement with experimental observations. Under the A1B climate scenario, modeled fugacities resulted to be higher because higher temperatures reduce the fugacity capacity of chemicals in air, water and sediments, and because diffusive sources to air are stronger.

Ma and Cao (2010) also quantified the perturbations of PCBs (28 and 153), in addition to HCHs and HCB, as result of the precipitation decrease related to climate change. PCB-28 exhibited a trend similar to that of α-HCH, showing a concentration increase of 2%.

Carrie et al. (2010) assessed the influence of climate change on the concentrations of PCBs in Mackenzie River (Canada) burbot (*Lota lota*), in which PCB levels were analyzed. Lipid-corrected concentrations over the period 1988-2008 for Σhexa-PCB and Σhepta-PCB showed a progressive increase with a minimum in early 2000, and a maximum in 2008. A strong temporal correlation between increasing primary productivity and PCBs in burbot was found, suggesting that warming temperatures and reduced ice cover might lead to an increased exposure to these contaminants in high trophic level Arctic freshwater biota.

Bogdal et al. (2009) also measured PCBs, which started to be globally produced in the 1930s and dramatically increased in the 1960s. PCBs were banned in Switzerland for open applications in 1972, while in 1986 a complete ban for all applications was established. However, PCB fluxes clearly increased again from 1990s to 2005. The same research group also considered PCBs in the forecast of POP melting release in an expected warming world (Bogdal et al. (2010). The amount of PCBs incorporated into the glacier between 1930-2006 was 335 g, following an increasing temporal trend.

To assess the bioaccumulation of PCBs, Borgå et al. (2010) applied a bioaccumulation model to calculate the effect of two different climate change scenarios on the pelagic marine food web of the Artic. Based on the results of this group of

pollutants, PCB-52 showed intermediate changes in the bioaccumulation compared to γ-HCH and PCB-153. Due to the temperature effect on K_{ow} and K_{oa}, there was a higher reduction in the bioaccumulation of PCB-52 than that of PCB-153. This was a consequence of the temperature effect on growth rate, which was less influential for PCB-52 than for PCB-153. The former congener showed the largest reduction in bioaccumulation, and a large spread in direction and magnitude of influential parameters and processes.

Ng and Gray (2011) coupled bioenergetic and bioaccumulation models to investigate the biological and chemical effects of climate change into three Great Lakes (US and Canada) fish species (round gobies, mottled sculpin, and lake trout). The accumulation of PCBs was calculated under four climate scenarios for Lake Erie and Lake Superior scenarios. Round goby and lake trouts showed the highest PCB concentration in Lake Superior under a 100-year projection (temperature increase of 5ºC). In turn, mottled sculpin did not show an important distinction in concentration terms between scenarios. In turn, Elliott et al. (2012) also reported temporal trends for PCB concentration in osprey eggs in different mountainous watersheds in western Canada, being coincident with some modeled predictions of release from melting glaciers. ΣPCB concentrations in eggs and plasma were up to 1420 and 28.2 ng/g, respectively. Similarly to other POPs, it was concluded that there would be lower levels of PCBs in relatively small lakes draining areas of large watersheds in the future.

Recently, Hansen et al. (2015) applied The Danish Eulerian Hemispheric Model (DEHM) to investigate how projected climate changes will affect the atmospheric transport of 13 POPs -10 PCB congeners and 3 HCHs- to the Arctic and their environmental fate within that Ocean. Under the applied climate and emission scenarios, the total mass of all compounds was predicted to be up to 55% lower across the Northern Hemisphere, at the end of the 2090s, than in the 1990s. The mass of HCHs within the Arctic was predicted to be up to 38% higher, while the change in mass of the PCBs was predicted to range from 38% lower to 17% higher, depending on the congener and the applied initial environmental concentrations.

MacLeod et al. (2005) developed the Berkeley-Trent Global model (BETR-Global) and evaluated its performance in describing atmospheric concentrations of individual PCB congeners and the dependence of these concentrations on large-scale climate variability. The role of the variability in global-scale climate conditions was proved to be very important. More specifically, they estimated that the maximum variability in

atmospheric PCB concentrations attributable to the North Atlantic Oscillation variability is approximately a factor of 2. In contrast, Kong et al. (2013) reported that the uncertainty in chemical properties (e.g., degradation half-life), and not the uncertainty associated to the climate change, dominates the variance of modelled absolute fate of POPs. These findings were reported when evaluating the influence of input data to multimedia models, on the modelled fate of 6 PCB congeners under various climate and emission scenarios. Long-term average environmental concentrations of PCBs were forecasted to change in a factor of 2, when comparing present conditions with those estimated for the period 2080-2099 (Kong et al., 2013). In another investigation (Kong et al., 2014), the same group of researchers calculated the steady-state concentrations of hypothetical perfectly persistent chemicals in the Baltic Sea water column under two alternative climate change scenarios (IPCC A2 and B2), being compared to results for a baseline climate scenario. The application of the POPCYCLING-Baltic multimedia chemical fate model highlighted temperature as the most influential individual climate parameter, being more relevant than precipitation, wind speed and particulate organic carbon.

In a modelling study of the concentrations of PCB-153 in the North Sea during the 21st century, O'Driscoll et al. (2014) found that the total mass of PCB-153 in sediments will decrease because of degradation, erosion and subsequent volatilization during storms. In contrast to γ-HCH, which was identified as a net depositional compound at the North Sea surface, PCB-153 was noted to be "volatilizational". However, the influence of the climate change on PCB-153 was suggested to be small, while trends in emissions from primary and secondary sources will remain as the key driver. In turn, the net export of PCB-153 out of the Artic was suggested to increase under future climate conditions, according to the estimations of Octaviani et al. (2015) when studying the long-term atmospheric cycling and fate of POPs. These data contrasted with those found for DDT, for which a trend of decreasing net Arctic import would reverse to an increasing trend, 100 years after peak emission (Octaviani et al., 2015). Surface exchange (water/air and soil/air) is much more important for the cycling of PCB-153 than that of PCB-28 (because of short atmospheric lifetimes) and of DDT (because of very low volatility).

2.4. Polycyclic aromatic hydrocarbons (PAHs)

603 The number of investigations focused on the effect of climate change on PAHs is
604 very scarce, with a certainly limited number of approaches. The only two areas where
605 the climate change impact on the fate of PAHs has been studied, are South Korea and
606 the Arctic. Cai et al. (2014) quantitatively assessed the predicted impacts of the climate
607 change on the transport and fate of PAHs within and across environmental media in
608 South Korea. Simulations were conducted for the period from 2000 to 2049 under the
609 A1B scenario, being compared with a non-climate change scenario. Similarly to recent
610 investigations on other POPs (Gouin et al., 2013; Kong et al., 2013), changes within a
611 factor of 2 for the average concentration of PAHs in air, soil and water, were estimated.
612 Degradation rate would play a leading role in the change of PAH levels in soils, while
613 in water, runoff and degradation would be the key processes.
614 On the other hand, the effects of PAH emissions for the period 2000–2050 and the
615 climate change on the atmospheric transport of three PAHs (phenanthrene, pyrene, and
616 benzo[a]pyrene) were investigated by Friedman et al. (2014). The GEOS-Chem model,
617 coupled to meteorology from a general circulation model, was used. The study was
618 focused on impacts to Northern hemisphere midlatitudes and the Arctic. A small 2050
619 "climate penalty" for volatile PAHs, and "climate benefit" for particle-bound PAHs,
620 was estimated. Deposition and surface-to-air fluxes of the 3 analyzed PAHs were
621 suggested to be the critical factors for the increase or decrease of environmental PAHs
622 in air.
623 In another study not entirely focused on the links between climate change and
624 PAHs, Brinkmann et al. (2010) assessed how flood events will affect rainbow trout as a
625 consequence of biomarker cascade, after exposure to PAH-contaminated sediment
626 suspensions. The main motivation was the fact that temperatures of German rivers
627 frequently exceed 25ºC during summer, because of the recent changes in climate.
628 Effects of re-suspension of sediments on biota under elevated temperature regimes are
629 likely to differ from those under lower temperature regimes. On the other hand, Nadal et
630 al. (2006) assessed the joint impact of UV-B radiation and temperature on the
631 photodegradation of PAHs. This approach was experimentally performed by means of
632 comparison of two different environments: Atlantic (Lancaster, UK) and Mediterranean
633 (Tarragona, Catalonia, Spain) climatic conditions. A significant faster photodegradation
634 rates were detected, specially for light PAHs, suggesting some kind of synergistic effect
635 when both temperature and UV-B dose increased. This synergism might have a great

implication on the long-range transport of environmental organic pollutants, taking into account that low-latitude areas are the hottest and most irradiated of the planet.

3. Conclusions

Legacy POPs (pesticides, PCBs and PCDD/Fs) are the compounds to which researchers have paid more attention, when studying the influence of climate change on their environmental behavior. In general terms, it is estimated that the global change will alter the environmental concentrations of POPs within a factor of 2-3. In turn, information on PAHs is particularly scarce, even being likely to be significantly affected by the climate change. Moreover, few investigations have focused on PAHs and metabolites, whose incidence may be even higher than parent compounds. Furthermore, data on the effect of climate change on the environmental fate of emerging contaminants, such as polybrominated diphenyl ethers (PBDEs) and perfluoroalkyl substances (PFASs), are not currently available. Consequently, more information is clearly necessary (Pacyna et al., 2015).

Nonetheless, global modeling studies do not agree on whether climate change acts to reduce or increase environmental concentrations of POPs in the Arctic (Hansen et al., 2015), the area where the linking between the global change and the occurrence of POPs has been more extensively studied in recent years. Furthermore, as modeling uncertainty plays a key role, interpretation or speculation from data coming from the application of multimedia environmental fate models should be treated with caution (Gouin et al., 2013). Although very preliminary, a number of studies have remarked the effect of melting glaciers, as well as some extreme events, such as floods and droughts, on the remobilization and bioaccumulation of POPs. The influence of temperature increase and precipitation decrease, on the fate and transport of POPs, has been also investigated. However, studies considering the expected increase of UV-B radiation as a consequence of ozone layer depletion have not been found. Sensitive areas, such as the Arctic, the Alps or the Mediterranean, should be particularly considered, as they are more exposed to variations resulting of the global warming. Furthermore, the generation and toxicity of byproducts, as well as their potential interaction, deserves further attention. Finally, the lack of studies focused on assessing potential changes of health risks associated to the exposure to POPs, as a consequence of the climate change, needs also to be addressed.

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