

Climate change and global cycling of persistent organic pollutants: A critical review

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Abstract Climate warming, one of the main features of global change, has exerted indelible impacts on the environment, among which the impact on the transport and fate of pollutants has aroused widespread concern. Persistent organic pollutants (POPs) are a class of pollutants that are transported worldwide. Determining the impact of climate warming on the global cycling of POPs is important for understanding POP cycling processes and formulating relevant environmental policies. In this review, the main research findings in this field over the past ten years are summarized and the effects of climate warming on emissions, transport, storage, degradation and toxicity of POPs are reviewed. This review also summarizes the primary POP fate models and their application. Additionally, research gaps and future research directions are identified and suggested. Under the influence of climate change, global cycling of POPs mainly shows the following responses. (1) Global warming directly promotes the secondary emission of POPs; for example, temperature rise will cause POPs to be re-released from soils and oceans, and melting glaciers and permafrost can re-release POPs into freshwater ecosystems. (2) Global extreme weather events, such as droughts and floods, result in the redistribution of POPs through intense soil erosion. (3) The changes in atmospheric circulation and ocean currents have significantly influenced the global transport of POPs. (4) Climate warming has altered marine biological productivity, which has changed the POP storage capacity of the ocean. (5) Aquatic and terrestrial food-chain structures have undergone significant changes, which could lead to amplification of POP toxicity in ecosystems. (6) Overall, warming accelerates the POP volatilization process and increases the amount of POPs in the environment, although global warming facilitates their degradation at the same time. (7) Various models have predicted the future environmental behaviors of POPs. These models are used to assist governments in comprehensively considering the impact of global warming on the environmental fate of POPs and therefore controlling POPs effectively. Future studies should focus on the synergistic effects of global changes on the cycling of POPs. Additionally, the interactions among global carbon cycling, water cycling and POP cycling will be a new research direction for better understanding the adaptation of ecosystems to climate change.

Keywords Global and regional warming, POPs, Primary and secondary emission, Global cycling

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1. Introduction

The basic characteristics of regional and global climate can

be described as cold, warm, dry and wet. The temperature over the past 100 years has gradually increased, which has been proved by large numbers of observations (Meinshausen et al., 2009; IPCC, 2013, 2014), and lake (Wang et al., 2007b) and ice-core records (Thompson et al., 1997;

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Petit et al., 1999; Thompson et al., 2003). The fifth Intergovernmental Panel on Climate Change (IPCC) assessment report addressed the fact that global warming is undoubted and unprecedented (Sheng et al., 2013). The temperature during 1983–2012 was the highest since 1400 years ago, particularly in the Northern Hemisphere (IPCC, 2013, 2014). Global warming is therefore the main characteristic of climate change at present. Scientists have reached a consensus that global warming has mainly been caused by greenhouse gas emissions since the industrial revolution.

Global warming has already triggered a series of environmental problems. For example, the increase in glacier melting rates driven by climate warming has resulted in the rise of sea levels, which has initiated changes in global water cycling (Meehl et al., 2005; Vermeer and Rahmstorf, 2009). Another example is the large amount of carbon released into the environment from fossil fuel combustion, which directly causes global warming, has broken the original carbon balance in ecosystems, leading to changes in the global carbon cycle (Cox et al., 2000). Therefore, the relationships between climate change and water and carbon cycling, and their effects and response mechanisms are research hotspots in the field of global environment change (Cao and Woodward, 1998), and large numbers of related review papers have been published (Cao and Woodward, 1998; Huntington, 2006; Luo, 2007). In addition, climate warming also has complex and significant effects on the biosphere, especially in phenology (Hughes, 2000; Parmesan, 2007), food-chain structure (Petchey et al., 1999), biological diversity (Botkin et al., 2007) and the agriculture and animal husbandry industries (Mendelsohn et al., 1994).

There are also close relationships between climate change and pollutant emissions, distribution and toxicity (Bridgman, 1991; Noyes et al., 2009; Seinfeld and Pandis,

2012). Both the IPCC report and the United Nations Environment Programme (UNEP) annual report emphasized that we should pay attention to the problem of environmental pollution, especially under the influence of global warming (UNEP, 2010; IPCC, 2013). So far, persistent organic pollutants (POPs) have drawn special attention. Considering the persistence, volatility, toxicity, bioavailability and long-distance atmospheric transport of POPs, more than 150 countries worldwide signed the Stockholm Convention on POPs to jointly protect the environment (Zheng, 2013). Similar to the influence of climate warming on the carbon and water cycles, the distribution, behavior and transport of POPs are also global-scale problems (Figure 1) and all these global environmental problems are interrelated. POP cycling is closely associated with biological material cycling and energy flow (Figure 2). In fact, emission, transport, sources and sinks, bioavailability, degradation, food-chain transfer and toxicity of POPs are all closely related to climate change. But how does climate change affect the global cycling of POPs? In this paper, the research progress regarding this scientific problem is summarized and reviewed, and existing problems and future research direction are highlighted.

2. Changes in POP emission sources

POPs are a series of pollutants that are persistent, toxic, bioavailable and semi-volatile in the environment (O'Sullivan and Megson, 2014). The Stockholm Convention lists several precedent-controlled POPs, including organochlorine pesticides (OCPs, representative compounds include dichlorodiphenyltrichloroethane (DDT)), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers

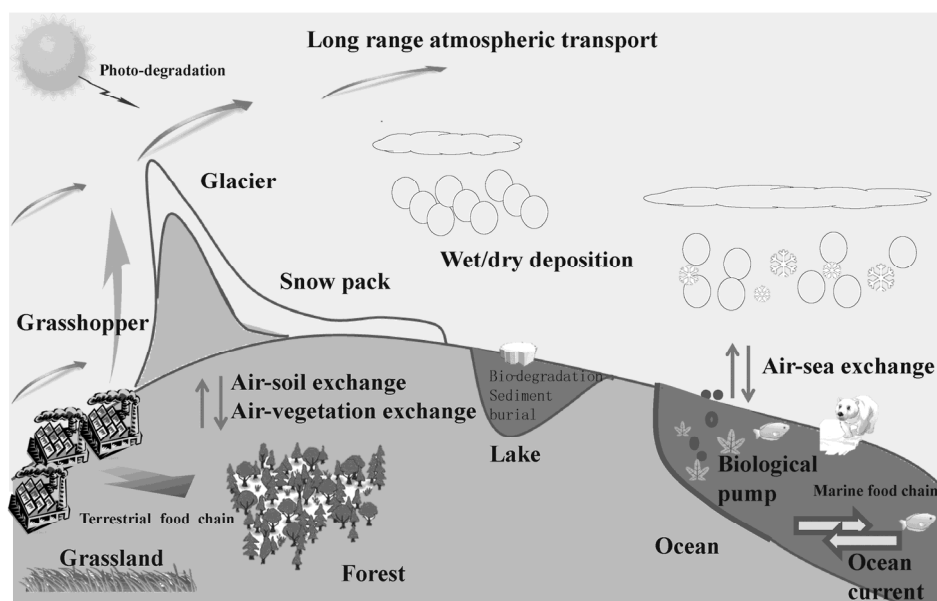


Figure 1 Transport processes for persistent organic pollutants.

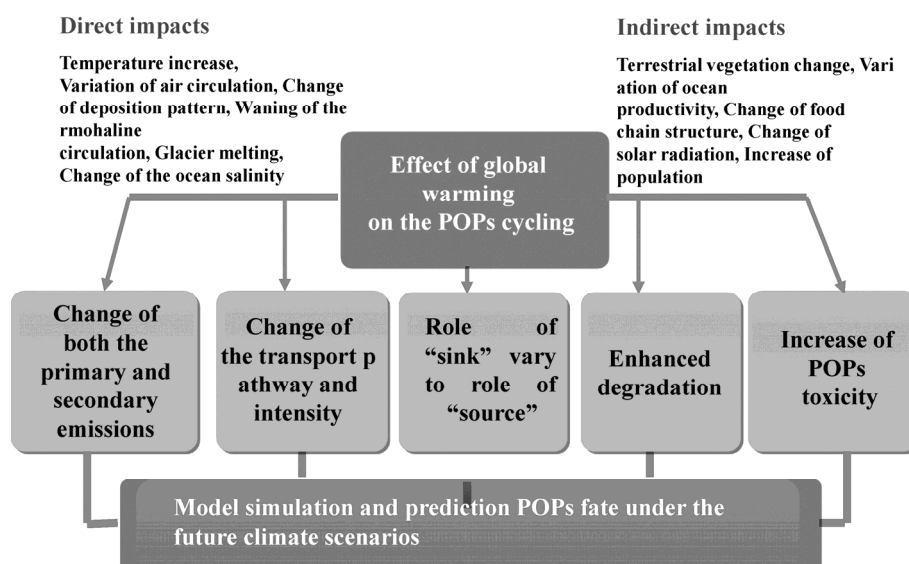


Figure 2 Direct and indirect impacts of climate change on the behavior and fate of persistent organic pollutants.

(PBDEs) and perfluorinated substances (Wang et al., 2009; Wang et al., 2010b). These pollutants are intentionally made synthetic chemicals, and industrial and agricultural activities are their main emission sources. In addition, some unintentional discharged byproducts (i.e. polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) generated by metal smelting and waste incineration) and polycyclic aromatic hydrocarbons (PAHs) are also included in the Stockholm Convention control list and the long-range atmospheric transport draft (Wania, 2003; Liu and Zheng, 2013; Zheng, 2013). Whether intentional or unintentional, the direct emission of POPs through human activities is regarded as the primary emission. In addition, owing to their semi-volatility, elevated environmental temperatures result in the re-evaporation of POPs from surface media, which is called the secondary emission. Obviously, global warming will affect industrial and agricultural production and the secondary emission of chemicals, leading to changes in POP emission sources (Figure 2).

Global warming has triggered droughts and rising sea levels, which has resulted in the reduction of global arable land (Parry and Ruttan, 1991). The fourth IPCC report indicated that under the influence of global warming, the northern boundary for crops will continue to move to higher latitudes (IPCC, 2013; Wöhrnschimmel et al., 2013). Overall, the influence of global warming on crop production is complicated and shows strong regional differences. In general, the impact of climate warming on agriculture is more negative than positive. One of the main negative influences is the agricultural pest problem. The distribution, growth and reproduction of crop pests are closely related to temperature. Low temperatures prohibit pest growth and reproduction. With the increase in temperature, the distribution area of crop pests is expected to expand (Porter et al., 1991;

Rosenzweig et al., 2001). Both changes in agricultural planting areas and the ravage of pests will require changes to the pesticide type and dosage. Therefore, from the perspective of global warming, this would likely result in massive emission of pesticidal POPs, including some that have been listed in the Stockholm Convention, such as endosulfan and lindane, as well as some new pesticides.

The IPCC's fifth report states that it is very likely that the frequency of heatwaves and rainstorms will increase with global warming. This means that global warming prompts extreme climatic events such as droughts, floods and hurricanes (Shen and Wang, 2013) and it is generally acknowledged that malaria is widespread after flooding (Easterling et al., 2000; Rosenzweig et al., 2001). Furthermore, with the expansion of climate warming, the malaria-affected area might also extend from the tropics to the Polar Regions (Githeko et al., 2000). Studies predict that the number of malaria patients will double in the tropical regions by 2100 and increase tenfold in temperate zones. In the next century, 45–60% of the global population may live in the potential transport area of malaria (Martens et al., 1995; Lindsay and Birley, 1996). DDT, one kind of POP, is a powerful chemical used to control the spread of malaria and has been widely used to fight the malaria parasite *Plasmodium* in many countries (Roberts et al., 2000; Stapleton, 2004). Owing to its toxicity, DDT has been banned since the 1970s. However, due to its low cost and high efficiency in terms of anti-malarial activity, in 2006, the World Health Organization (WHO) suggested the re-use of indoor spraying of DDT to block malaria transport (Sadasivaiah et al., 2007). Malaria-vulnerable countries, such as India and South Africa, also applied to the Stockholm Convention for exemption of DDT (Sharma et al., 2005; Channa et al., 2012). Consequently, global use of DDT is expected to in-

crease due to the impact of climate warming.

Owing to their semi-volatility, POPs widely exist in all kinds of media in the world. They continuously undergo dynamic distribution between the atmosphere and the land surface, e.g. soil, water, vegetation and so on. (Bidleman, 1999). The physicochemical parameters of POPs, such as saturated vapor pressure, Henry's law constant, gas-water partition constant and octanol-air partition coefficient, are all temperature-dependent constants (Paasivirta et al., 1999). Therefore, the constantly rising temperatures will significantly affect the distribution of POPs between different phases, and promote POPs to migrate from the land surface to the atmosphere. Consequently, there are more active POPs in atmosphere (Hung et al., 2010; Ma and Cao, 2010). This process is called the secondary emission of POPs. Studies have shown that most European soils are a secondary source of low-molecular-weight PCBs (Meijer et al., 2003; Růžičková et al., 2007), and Indian soils are a secondary source of DDT (Sharma et al., 2014). The elevated PCB concentrations in the Arctic atmosphere have been mainly attributed to the gradual rise in environmental temperature driven by global warming (Becker et al., 2006). Although the use of hexachlorocyclohexane (HCH) had been banned for more than 30 years, the current HCH concentration in the Arctic atmosphere is not decreasing, but slightly increasing. Ma et al. (2011) reported that secondary release of HCH caused by climate warming has buffered the effect of HCH prohibition.

3. Changes in the long-range transport of POPs

Atmospheric circulation and ocean currents are two major ways in which POPs are globally transported. POPs can exist in the atmosphere in both gas and particulate phases. Hence, with the help of atmospheric circulation, gaseous and particulate POPs in atmosphere can be globally spread (Jones and De Voogt, 1999). During the atmospheric transport process, POPs are likely to be deposited on the land surface when the temperature decreases, while they will evaporate into the atmosphere to migrate again when the temperature rises. This process continually occurs, so POPs can be transported and deposited to remote areas. This is the so-called grasshopper effect (Gouin et al., 2004). In addition, some POPs with relatively higher solubility, such as HCH (Kucklick et al., 1991) and perfluorooctane sulfonates (PFOS) (Giesy and Kannan, 2002), can also enter into surface waters, and then feed into ocean currents and undergo global ocean transport (Yamashita et al., 2008). Climate warming leads to changes in climatic factors, such as temperature, wind speed, wind direction and precipitation. The variation of these factors will certainly alter both the air and ocean transport intensity and pathways of POPs.

The global temperature gradient decreases with increasing latitude. The emission sources of POPs are mostly dis-

tributed in low-latitude areas with relatively warm temperatures. Accordingly, chemicals with high volatility will undergo multi-hopping, and will be easily transported to the high-latitude Polar Regions, while less-volatile compounds undergo less or single hopping. This leads to global chemical fractionation. This effect is termed the global distillation effect (Fernández and Grimalt, 2003). The global distillation effect of POPs is an important hypothesis to explain the global transport mechanism of POPs, which has been verified by numerous studies (Wania and Su, 2004; Blais, 2005; Yang et al., 2008). In this model, the ambient temperature is a decisive factor driving POP transport. If the ambient temperature increases by 1°C, the volatility of POPs increases by 10–15% (Lamon et al., 2009b; Wöhrnschimmel et al., 2013).

The global atmospheric circulation shows obvious fluctuations with climate change. The major climate phenomena are the El Niño-Southern Oscillation (ENSO) in the South-east Pacific and Indian oceans, the Northern Atlantic Oscillation (NAO) and the Pacific North American (PNA) teleconnection. Based on a 20-yearlong dataset of atmospheric POP observations in the Great Lakes region, Ma et al. (2003, 2004), Ma and Li (2006) assessed the relationship between POP levels and climate signals, and found that there was a significant correlation between variations in the hexachlorobenzene (HCB) concentration and ENSO fluctuations, and between variations in the HCH concentration and NAO fluctuations in winter and spring, respectively. Although various studies have shown that the primary emission of POPs has gradually decreased, HCB and HCH were found in elevated concentrations, which has been attributed to the increased temperature associated with the ENSO and NAO in the Great Lakes region. Temperature is a driving force for secondary emission of pollutants from soil and water, which is the reason for the increase in atmospheric concentration of HCB and HCH. Climate change will also affect the atmospheric transport of POPs in China. China was the main user of α -HCH and soils may have been contaminated with higher levels of residual chemicals. Tian et al. (2012) found that the output flux of α -HCH through the atmospheric circulation from Northeast China significantly increased under climate warming.

In addition, higher wind speeds will lead to the large-scale and more-effective intercontinental transport of POPs. During the positive phase of the NAO, strong westerly winds blow over the North Atlantic, which enhances the transport of used POPs in the Canadian prairies to the Great Lakes region (Gao et al., 2010; Christoudias et al., 2012). When the PNA pattern is enhanced, southwesterly winds from Canada are increased, which leads to POP transport to the north polar area (Hung et al., 2005). As another kind of climate system, monsoons can change the direction of the atmospheric circulation and precipitation intensity on the seasonal scale. The pathways and intensity of atmospheric transport of POPs is thus shifted seasonally. Previous stud-

ies have found that the monsoon enhances the vertical input of atmospheric POPs to the Arabian Sea (Dachs et al., 1999). Moreover, atmospheric transport driven by the monsoon continuously transports POPs of South Asia to the Tibetan Plateau (Sheng et al., 2013). Climate change influences not only the intensity of the monsoon, but also the direction of the wind at large spatial scales, both of which affect the global transport of POPs.

Atmospheric deposition is an important process in the global cycling of POPs. It plays an important role in scavenging. Temperature is the main factor that affects the form and the efficiency of atmospheric wet deposition (Lei and Wania, 2004). When the temperature reaches 0°C, rainfall has a greater removal efficiency of low-molecular-weight compounds existing in the gas phase, while snow has a higher removal efficiency of high-molecular-weight compounds and non-polar compounds (Lei and Wania, 2004). When the temperature is below 0°C, snow can more effectively remove compounds from the atmosphere (including both gaseous and particulate POPs); thus, this process can block POP transport to the atmosphere. There is a great difference in terms of atmospheric wet deposition for different compounds due to their different physical and chemical properties (Lei and Wania, 2004). From a global perspective, high POP deposition flux values in the intertropical convergence zone and high-latitude areas are expected due to high precipitation-induced wet deposition and low temperature-induced cold trapping, respectively (Jurado et al., 2005). In fact, the intertropical convergence zone will continue to move to northward under the influence of global warming (Frierson and Hwang, 2012), and the global precipitation patterns will be more varied (Philander et al., 1996; Held and Soden, 2006), which could possibly further change the global transport and distribution pattern of POPs.

Compared with the atmospheric transport (unit of speed: m s^{-1}) of POPs, ocean current transport (unit of speed: cm s^{-1}) is a relatively slow process. POPs enter into the ocean currents by both atmospheric deposition and surface runoff (Zingde, 2005; Zhang et al., 2011). A series of observation data from offshore has demonstrated that seawater of the European Mediterranean (Gómez-Gutiérrez et al., 2007; Castro-Jiménez et al., 2012), Baltic Sea (Theobald et al., 2011), North Sea (Ilyina et al., 2006), Bohai Sea (Chen et al., 2011), Yellow Sea (Lin et al., 2012; Zhong et al., 2014; Yang et al., 2003), South China Sea (Lin et al., 2012), Indian Ocean (Xie et al., 2011; Huang et al., 2013) and the Bay of Bengal (Sarkar et al., 2008) contain higher levels of POPs. These pollutants include compounds banned in the Stockholm Convention as well as some emerging POPs (Kallenborn et al., 2012). The sea itself is a huge circulation system: warm streams transfer tropical heat poleward, while ocean currents cool and sink in the Polar Regions, transferring matter and energy on the global scale (thermohaline circulation). Lohmann et al. (2006) calculated the sinking

flux of PCBs on the basis of the sink rate of seawater. The results showed that the sinking flux of PCBs in the Norwegian Sea was larger than that in the Labrador Sea, the Ross Sea and Weddell Sea in the Southern Ocean. They also pointed out that PCBs can be used as tracers to track the movement of deep ocean currents. Currently, with global warming continuing, the temperature gradient that drives the thermohaline circulation is waning (Rühlemann et al., 1999). Thereby, the global ocean transport of POPs would be affected and needs to be investigated in future.

4. Effect of climate change on the fate of POPs

After transport, POPs eventually sink into the ocean, cryosphere, forest and soil (Figure 1). The ocean accounts for more than 70% of the global surface and is the largest sink of most pollutants (Sobek and Gustafsson, 2014). Seawater, zooplankton and particles contain measurable amounts of POPs, but most POPs are stored in ocean sediments (Nizzetto et al., 2010). Moreover, the forest canopy and soil organic matter can effectively accumulate atmospheric POPs. Therefore, the terrestrial ecosystem also stores a large amount of POPs (Ockenden et al., 2003; Nizzetto et al., 2010). As well as marine and terrestrial ecosystems, the cryosphere is another important destination for POPs. With wet deposition, atmospheric POPs can be scavenged by rain and snow and further stored in glaciers (Hermanson et al., 2005; Meyer and Wania, 2008). The cryosphere is extremely sensitive to climate change, so POPs stored in the cryosphere are prone to re-release under global or regional warming, and become secondary sources of POPs. Under the influence of global warming, the role (sink or source) of marine and terrestrial ecosystems and the cryosphere in POP global cycling will certainly change.

Owing to the hydrophobicity of POPs, they exist in the ocean in the particulate phase. Dissolved organic carbon, particulate matter, phytoplankton and so on all act as carriers of hydrophobic POPs in water. Maldonado et al. (1999) studied the vertical profile of PAHs in the Black Sea, and the results indicated that the highest value of PAHs appeared at a depth of 30 m, which was mainly attributed to the high biomass of phytoplankton at that depth. Phytoplankton plays an important role in the transport of POPs from water to sediment. On the one hand, phytoplankton absorb POPs, which decreases the POP concentration in water (Dachs et al., 2000). On the other hand, POPs associated with phytoplankton sink down the water column and finally accumulate in sediment. These two processes drive the flow of POPs from the atmosphere to the ocean, which is known as the biological pump (Dachs et al., 2002). Obviously, POPs in the atmosphere continuously enter into the deep-sea sediments through this effect. However, seawater temperature is rising as a consequence of global warming, which has reduced ocean primary productivity by 6% since

the 1980s (Hoegh-Guldberg and Bruno, 2010; Galbán-Malagón et al., 2012). Therefore, climate warming indirectly affects the ocean's ability to absorb POPs by changing ocean primary productivity.

Soil with a high organic matter content has a higher POP storage capacity. Owing to the different distributions of soil temperature and organic matter content at different scales, the POP storage ability of soil varies. The POP storage ability in the Sahara Desert with a low soil organic matter content is relatively weak, while the POP storage ability in Siberia, Scandinavia, Canada and Alaska, which are characterized by a higher organic matter content, is much stronger (Dalla Valle et al., 2005).

Forest accounts for about 31% of the land area worldwide (Guan, 2003). The forest canopy provides an extensive organic surface for the partitioning of POPs in the atmosphere, and increases the net atmospheric deposition of POPs (Horstmann and McLachlan, 1998; McLachlan and Horstmann, 1998; Wania and McLachlan, 2001). Owing to vegetation renewal and litter fall, forest increases the atmospheric deposition of POPs. As a consequence, terrestrial forest ecosystems are important sinks for environmental POPs (Horstmann and McLachlan, 1998; McLachlan and Horstmann, 1998). It is well established that boreal forest can store 2–20% of global PCB emissions (Moeckel et al., 2009). From a global perspective, land cover has been transforming at a relatively high speed. Deforestation, agricultural reclamation and urbanization destroy the soil layer, so POPs that have accumulated in soil are re-released. These processes also reduce the POP retention capacity of soil (Komprda et al., 2013). In contrast, climate warming may also exert positive effects on the storage of POPs; for example, climate warming can extend the vegetation growing season and increase the content of soil organic carbon (Komprda et al., 2013), which may consequently increase the accumulation of POPs in terrestrial ecosystems.

Precipitation plays an important role in the deposition of POPs. POPs scavenged by atmospheric wet deposition can be stored in glaciers, snow pack and sea ice (Yao et al., 2002; Hermanson et al., 2005; Wang et al., 2007a; Meyer et al., 2008a, 2008b; Meyer and Wania, 2008; Wang et al., 2008, 2014; Guglielmo et al., 2012). Antarctica has the largest glacial area worldwide. Peterle et al. (1969) estimated the storage of DDT in Antarctica (2.4×10^6 kg) based on concentrations of DDT and Antarctic snow-ice reserves (24×10^7 – 30×10^7 m³) measured in 1966. Hofmann et al. (2012) calculated the storage of HCH and DDT in environmental medium and identified that approximately 8.4% of atmospheric γ -HCH and 10.4% of atmospheric DDT were deposited to snow and glaciers.

It is possible to evaluate the atmospheric concentration level, accumulation flux and historical accumulation trend of POPs based on dated ice cores (Hong et al., 2009). Currently, research regarding accumulation of POPs in ice cores is mainly focused on Arctic Svalbard (Hermanson et

al., 2010), the Italian Alps (Villa et al., 2003, 2006; Maggi et al., 2006), China's Tibet (Wang et al., 2008, 2010a, 2014) and the Canadian Ellesmere Island (Veillette et al., 2012; Zhang et al., 2013). Ice cores are regarded as natural archives of pollutants as well as past climate. Global warming is having a remarkable impact on the cryosphere (Parry, 2007). The IPCC report states that during the past 30 years, global glaciers have continued to retreat in length, area and volume (Stocker, 2014). In addition, snow cover in the Northern Hemisphere has decreased, and the thickness and area of permafrost in Russia and northern Europe evidently reduced between 1975 and 2005 (IPCC, 2013). Melting of Arctic glaciers has released massive amounts of POPs back into atmosphere, leading to a slight increase in the concentration of a variety of POPs in the Arctic atmosphere during the past 20 years (Ma et al., 2011). Furthermore, the concentration of DDT in penguins from Antarctic has not decreased, which might be associated with the release of POPs from melting glaciers (Geisz et al., 2008). Melting glaciers may also increase the concentration of POPs in fresh water (Ma et al., 2011). Blais et al. (2001) found that 50–90% of POPs in Glacier Lake were sourced from glacial melt water. Under global warming, POPs can also migrate into the lakes through the water cycle and even accumulate in lake sediments (Bettinetti et al., 2008; Bogdal et al., 2009).

In summary, climate warming will alter the role (source or sink) of the land surface in the global cycling of POPs. Originally, glaciers, oceans and soil were sinks of POPs. However, the re-release of accumulated POPs under the influence of global or regional warming has resulted in glaciers, oceans and soil becoming sources of POPs (Bettinetti et al., 2008; Cabrerizo et al., 2011). As the primary emission of POPs has been regulated, secondary emission is now the main source of POPs in the atmospheric environment.

5. Effect of climate change on degradation, bioavailability and toxicity of POPs

The degradation of POPs in the environment is mainly divided into two types: photodegradation and biodegradation. The degradation rate of POPs is influenced by hydroxyl radical ($\cdot\text{OH}$) and ozone (O_3) contents in the atmosphere (Klöpffer, 1992; Sinkkonen and Paasivirta, 2000; Wania and Daly, 2002). The microbial degradation of POPs primarily occurs in soils and sediments (Kawamoto and Urano, 1990; Hirano et al., 2007). The pH value of soil and sediment, organic carbon content and the amount and type of microorganisms affect the biodegradation rate of POPs. A higher organic carbon content can provide microorganisms with the energy to degrade POPs, thus speeding up the degradation process of pollutants (Hirano et al., 2007).

Global warming has led to an increased ozone concentration in the troposphere (Racherla and Adams, 2006; Steven-

son et al., 2006 ; Cheng et al., 2007). In addition, model simulations have shown that the concentration of ozone will continue to rise from now to 2050 (Hogrefe et al., 2004). From this point of view, the scientific community generally believes that photodegradation of POPs will become more severe under the effect of global warming (Brubaker and Hites, 1998; Macdonald et al., 2003; Sweetman et al., 2005; Ma et al., 2004; Meyer and Wania, 2008).

Similarly, Dalla Valle et al. (2007) concluded that increasing temperatures will lead to the prosperity of the microbial community in Italy's Venice Lake, which should enhance the biodegradation of PCBs and chlorinated furan. The decomposition of organic carbon in terrestrial and aquatic ecosystems is an important biological process that affects the bioavailability of POPs. Research in Ontario revealed that drought and dissolved organic carbon decomposition associated with global warming increased the effective content of POPs, which meant the bioavailability of POPs increased (Magnuson et al., 1997; Schindler et al., 1997).

Numerous studies have concluded that the bioavailability and toxicity of POPs in wild organisms increases with the increase of temperature and salinity (Wang et al., 2001; Waring and Moore, 2004; Capkin et al., 2006; Jenssen, 2006; Schiedek et al., 2007). Possible mechanisms for this are as follows. (1) The dynamic toxicity effect of compounds increases with the increasing temperature (Buchwalter et al., 2003; Maruya et al., 2005). (2) Warming gives rise to a weak immune ability of wildlife to POPs (Broomhall, 2002, 2004; Patra et al., 2007), which results in increasing toxicity of POPs. (3) Studies in Seattle Bay identified that increasing water salinity led to acute toxicity of salmon as anticholinesterase metabolites increased (Schlenk and Lavado, 2011). (4) Compared with the above three mechanisms, structural changes in ecosystem food chains derived from global warming is the leading factor that increases the toxicity of POPs to whole ecosystems (Macdonald et al., 2005; Jenssen, 2006; Burek et al., 2008; Noyes et al., 2009; Borgå et al., 2010; Bustnes et al., 2011).

6. Model simulation and prediction of the environmental behavior of POPs

So far, plenty of models have been applied to simulate and predict the environmental behavior and transport trends of POPs under the influence of global warming (Huang et al., 2007; Shindell et al., 2008; Guglielmo et al., 2009; Lamon et al., 2009a, 2009b; Stemmler and Lammel, 2009, 2012; Lammel and Stemmler, 2012; Gouin et al., 2013; Kong et al., 2013, 2014). These models can be grouped into two general categories. One is multimedia models based on fugacity theory, which is simple and has been widely used (Mackay, 2001). The other is general circulation models (GCMs), which divides the world into different areas and focuses on the transfer processes of mass, energy and momentum of chemicals among the atmosphere, land surface and ocean (Ilyina et al., 2006). These models can not only predict the environmental behaviors of POPs, but also determine the key environmental factors that affect the global transport of POPs. The main characteristics of these models are summarized below and in Table 1.

The Berkeley-Trent Global Model (BETR-Global) is a global mass-balance model based on steady state (level III) or unsteady state (level IV) fugacity scenarios. It synthetically considers the influence of temperature, atmospheric stability, atmospheric boundary layer and photochemical reaction processes on the transport of POPs under different environmental conditions (MacLeod et al., 2005).

Globo-POP is an integrated model with a prediction function based on the level IV fugacity model. Compared with the differences between this model and other fugacity models, Globo-POP considers non-temporal resolved input parameters, including organic carbon, aerosol and suspended particulate matter (Wania and Mackay, 1995; Meyer and Wania, 2007). Globo-POP resolves inputs consisting of temperature, ice cover and hydroxyl radical concentration. In the improvement of the Globo-POP model by Stocker et al. (2007), the impact of glacier melting on POPs was also taken into consideration.

Table 1 The main multimedia fate and transport models cited in this paper

Model	Description	Features
BETR-Global	Level III-IV fugacity model. The world is divided latitudinally and longitudinally into 288 regions. Seven environmental compartments are considered (two atmospheric layers, soil, vegetation, coastal water, freshwater and sediments).	This model can be flexibly applied on different scales (local) or global simulation.
Globo-POP	Level IV fugacity model. Latitudinal resolution up to 10 regions. It takes into account 9 compartments (freshwater and sediment, four vertical atmospheric layers, upper ocean layer, cultivated and uncultivated soils).	This model can be used for assessing the POPs fate and under the influence of ice melting.
G-CIEMS	Level IV fugacity model. GIS-based models. Six environmental compartments are assumed in this model (atmosphere, freshwater - rivers and lakes, coastal water, sediments, soil).	It could be used for calculating the fate of persistent pollutants in a climate change perspective especially at high spatial resolution for the dry land areas, where this model allows to easily distinguish different environments.

Another possibility for POP fate modeling is the spatially resolved and geo-referenced dynamic multimedia environmental fate model, G-CIEMS (Grid-Catchment Integrated Environmental Modelling System). This model integrates a basic fugacity formulation following a level IV calculation, on a geographic information system (GIS) (Suzuki et al., 2004). Six compartments, namely air, freshwater (rivers and lakes), sediments, forest, seawater, soil, and advective transport are considered.

Given that POP fate models can predict the transport pathways, intensity and concentrations of POPs for future climate scenarios, the model results provide policy makers with scientific information for an early response to these changes. Recently, on the basis of the moderate intensity greenhouse gas emission scenario, Octaviani et al. (2015) used a GCM to study meridional transport of POPs under the present-day (1970–1999) and future (2070–2099) climate. POP transport channels into the Arctic were identified as (1) the Alaska-North America channel, (2) the Greenland channel, (3) the Norwegian Sea-West Russia channel (Europe) and (4) the Urals-Siberian channel (Asia). Moreover, ignoring the primary emission of POPs, the model also predicts that secondary volatilization of DDT caused by global warming will still be continuously transported to the north polar area. However, in contrast, the results also predicted that the net export of PCB 153 out of the Arctic will increase under future climate conditions (2070–2099, Octaviani et al., 2015).

In general, model application provides a view for evalu-

ating the process and degree of the rise, cycling and fading away of POPs on the larger spatial and longer temporal scales. The current laws and regulations regarding environmental pollution do not consider the influence of climate change on the concentration of POPs in the environment. In fact, several studies have demonstrated that global warming could increase the long-range transport ability of POPs. As a result, it is necessary to consider the impact of climate warming on environmental loading of POPs in the formulation of relevant policies in future.

7. Research prospects

From the statistical results of the literature, studies related to climate change and the global transport of POPs were rarely reported until 2004. However, this research direction began to flourish from 2004 onwards and has made considerable progress during the past ten years. Both field observations and model simulations have proved that warming directly and indirectly affects the transport, distribution and final fate of POPs. Table 2 summarizes the response of POPs to global change for publicity and policy-making departments to understand and judge the factors.

Relevant research work in the Arctic is impressive. Scientists are conducting systematic studies in the following areas: (1) the effect of melting snow and ice on the secondary release of POPs; (2) the effect of secondary release of POPs on the temporal and spatial distribution of POPs in

Table 2 Environmental behavior of persistent organic pollutant in response to global change^{a)}

Scenario	Environmental consequence	Response of POPs	Effect on POPs level	Reference
Glaciers melting	Fresh water was injected into the environment; chemicals stored in the glacier were released.	POPs were rereleased into the environment e.g. atmosphere and lakes	+	UNEP, 2010; Grannas et al., 2013
Permafrost degradation	Exacerbated surface erosion	Increased the second emission of POPs	+	Evans et al., 2005; Grannas et al., 2013
Sea level rise	Increased erosion	Increased the second emission of POPs	+	Kwok et al., 2009
Salinity of sea water changes	Two situation: salinity is decreased due to injection of glacier melting water; seawater salinity increase because of local drought	Change of the marine food chain structure, variation of metabolism of marine biota	±	Olsen et al., 2011
Flood	Severe erosion	Increased the second emission of POPs	+	Holoubek et al., 2007
Forest fire	Surface soil temperature rise up the non-intentional POPs	Increased both the primary and secondary discharge of POPs	+	Kim et al., 2003; Kallenborn et al., 2011
Changes of atmospheric circulation	Changed POPs transport intensity and direction of	Changed the environment distribution of POPs	±	Ma et al., 2004, 2011; Ma and Li, 2006
Ocean current change	Changed POPs transport intensity and direction of	Changed the environment distribution of POPs	±	Lohmann et al., 2006
Population increase	Increased use of pesticides, malaria outbreak, and worsened environmental pollution	Increased both the primary and secondary discharge of POPs	+	Noyes et al., 2009
Soil desertification	The reconstruction of the vegetation zone around the world	Changed the fate of POPs	+	Noyes et al., 2009
Variation of biodiversity	The components of regional biosphere changed	Changes on the biological enrichment of POPs at regional scale	±	Brander, 2007

a) +, positive; -, negative

multimedia of the Arctic; (3) changes in the accumulation characteristics of POPs in Arctic ecosystems (marine food chain and tundra food chain) under the background of climate warming; (4) long-distance transport potential of different POPs to enter into the Arctic under the condition of climate warming; (5) how POPs will be imported to and exported from the Arctic area in a warming future. However, there are still various research gaps:

(1) The POP emission inventory is relatively clear, but under the gradual decrease of primary source POPs and the increasing importance of secondary emission, caused by climate warming, from the soil and sea, how can the contribution of secondary emission of POPs be quantified?

(2) It is necessary to study the effect of extreme weather, such as floods and storms, and soil sustainable desertification on migration of POPs on the global scale.

(3) It is necessary to carry out research on the relationship between emerging POPs and climate change.

(4) It is necessary to comprehensively assess the environmental and ecological impacts of POPs released from glaciers melting on global main mountain areas, e.g. the Alps, the Himalayas, and the Rocky Mountain.

(5) How can the relative contribution of secondary emission and degradation of POPs prompted by climate warming be measured?

In conclusion, the impacts of climate change on the global cycling of POPs are linked to the production and use of chemicals, temperature increases, variations in ocean and atmospheric circulation, change of land cover, biological absorption and metabolism, multiple photochemical reactions, multiple interface processes and other complex processes. The study of these impacts requires long time series observational data and large-scale research projects with international cooperation (Hung et al., 2010; Kallenborn et al., 2015; Pacyna et al., 2015), as well as collaborations of scientists from various disciplines (Nadal et al., 2015).

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