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SOILS, SEC 3 • REMEDIATION AND MANAGEMENT OF CONTAMINATED OR DEGRADED LANDS • REVIEW ARTICLE

How do persistent organic pollutants be coupled with biogeochemical cycles of carbon and nutrients in terrestrial ecosystems under global climate change?

Ying Teng · Zhihong Xu · Yongming Luo · Frédérique Reverchon

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Abstract
Purpose Global climate change (GCC), especially global warming, has affected the material cycling (e.g., carbon, nutrients, and organic chemicals) and the energy flows of terrestrial ecosystems. Persistent organic pollutants (POPs) were regarded as anthropogenic organic carbon (OC) source, and be coupled with the natural carbon (C) and nutrient biogeochemical cycling in ecosystems. The objective of this work was to review the current literature and explore potential coupling processes and mechanisms between POPs and biogeochemical cycles of C and nutrients in terrestrial ecosystems induced by global warming.

Results and discussion Global warming has caused many physical, chemical, and biological changes in terrestrial ecosystems. POPs environmental fate in these ecosystems is controlled mainly by temperature and biogeochemical processes. Global warming may accelerate the re-emissions and redistribution of POPs among environmental compartments via soil–air exchange. Soil–air exchange is a key process controlling the fate and transportation of POPs and terrestrial ecosystem C at regional and global scales. Soil respiration is one of the largest terrestrial C flux induced by microbe and plant metabolism, which can affect POPs biotransformation in terrestrial ecosystems. Carbon flow through food web structure also may have important consequences for the biomagnification of POPs in the ecosystems and further lead to biodiversity loss induced by climate change and POPs pollution stress. Moreover, the integrated techniques and biological adaptation strategy help to fully explore the coupling mechanisms, functioning and trends of POPs and C and nutrient biogeochemical cycling processes in terrestrial ecosystems.

Conclusions and perspectives There is increasing evidence that the environmental fate of POPs has been linked with biogeochemical cycles of C and nutrients in terrestrial ecosystems under GCC. However, the relationships between POPs and the biogeochemical cycles of C and nutrients are still not well understood. Further study is needed to explore the coupling mechanisms of POP environmental fate and C biogeochemical cycle by using the integrated techniques under GCC scenario and develop biological and ecological management strategies to mitigate GCC and environmental stressors.

Keywords Biogeochemical cycles of carbon · Global climate change · Persistent organic pollutants · Terrestrial ecosystems

1 Introduction

Global climate change (GCC), especially global warming, is unequivocal. The linear warming trend over the 50 years from 1956 to 2005 (0.13°C per decade) is nearly twice that for the 100 years from 1906 to 2005 and atmospheric carbon dioxide (CO_2) annual emissions have grown between 1970 and 2004 by about 80% (IPCC 2007). Global warming has caused many physical, chemical, and biological changes in terrestrial ecosystems (e.g., flooding, drought, wildfires,

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Y. Teng · Y. Luo (✉)
Key Laboratory of Soil Environment and Pollution Remediation,
Institute of Soil Science, Chinese Academy of Sciences,
Nanjing 210008, People's Republic of China
e-mail: ymluo@issas.ac.cn

Y. Teng · Z. Xu (✉) · F. Reverchon
Environmental Futures Centre and School of Biomolecular and
Physical Sciences, Griffith University,
Nathan, QLD 4111, Australia
e-mail: zhihong.xu@griffith.edu.au

insects, ice cap melting, sea level rise, species extinction) (Blum 2005; Running 2006; Kerr 2007; Turner et al. 2009; Xu et al. 2009; Schenker et al. 2010; Hoffmann and Sgrò 2011; Schmid et al. 2011). Like GCC, the environmental pollution by persistent organic pollutants (POPs) is an issue of global concern (Lamon et al. 2009). POPs are chemical substances that persist in the environment, bioaccumulate through the food web, and pose a risk of causing adverse effects to human health and ecosystems (UNEP 2005). Each POP has a long-range transportation potential in air and/or water and can bioaccumulate in lipid-rich tissues of the biota and biomagnify through terrestrial and aquatic food chains (Ma et al. 2004). Hence, POPs have a potential adverse impact on higher trophic animals in terrestrial and coastal systems (Olsson et al. 2000; Christensen et al. 2005; Kucklick et al. 2011) and on human health (Arnot et al. 2011). Moreover, with the intensification of global warming, an increase in POPs levels has been found in environmental compartments because of the release from environmental reservoirs such as soil, water, and ice (Nizzetto et al. 2010; Ma et al. 2011). These higher emissions induced by GCC would increase the vulnerability of exposed organisms including humans through the food chain and lead to greater adverse effects on human health and terrestrial ecosystems (Cousins et al. 2010). POPs pollution induced by GCC has recently attracted political concern and significant attention. Last year, the United Nations Environment Programme (UNEP) Stockholm Convention has announced a major international study on the influence of GCC and POPs on human health and the environmental ecosystem.

Terrestrial ecosystems can control and steer the Earth system and respond strongly to GCC (Heimann and Reichstein 2008; Arnone et al. 2008; Piao et al. 2009; Singh et al. 2010). The net exchange of C between the terrestrial biosphere and the atmosphere is the difference between C uptake by plant photosynthesis and releases by plant or ecosystem respiration, soil respiration, and disturbance processes such as fire, land management, and land-use change (IPCC 2007). The C biogeochemical cycling is a key coupling point between terrestrial ecosystems and the climate system (Cao and Woodward 1998a, b; Falkowski et al. 2000; Xu and Chen 2006; Chen and Xu 2010). Therefore, GCC can have significant impacts on the structure and function of terrestrial ecosystems (Root et al. 2003; Williams et al. 2004). Some studies have shown that the C biogeochemical cycling under high CO_2 concentrations is further drived and/or limited by nutrient availability and hydrological cycle (Schimel et al. 1997, 2001; Körner et al. 2005; Oki and Kanae 2006; Chen and Xu 2006, 2008; Wentz et al. 2007; Jung et al. 2010). Thus, the question is how POPs, as anthropogenic organic C sources from fossil-fuel emissions and chemicals, are coupled with the natural C and nutrient biogeochemical cycles in terrestrial ecosystems. Here, we advance some hypotheses on the potential coupling process and mechanisms between POPs and biogeochemical cycles of C and nutrients (Fig. 1), review the current literature in this area, and make some suggestions for further study.

2 POPs distribution and biogeochemical cycles of C and nutrients

Some studies have indicated that GCC would likely increase the exposure of the environment and ecosystem to POPs (MacLeod et al. 2005; Lamon et al. 2009; Ma and Cao 2010). POPs are widely distributed among the environmental compartments (i.e., air, soils, vegetation, water bodies, sediments, ice) of terrestrial ecosystems. Under the direct influence of GCC, POPs environmental fate has been undergoing significant changes and is controlled mainly by temperature and biogeochemical processes (Valle et al. 2007; Ma and Cao 2010; Nizzetto et al. 2010). Increasing temperature enhances volatilization and therefore leads to increased emissions into air (Lamon et al. 2009; Gioia et al. 2011). Global warming also increases the frequency of extreme events such as melting ice, storms, floods, and forest fires (IPCC 2007). Extreme weather events have a distinct impact on the remobilization and subsequent bioavability of POPs (Schenker et al. 2010; Schmid et al. 2011). Flooding events occur frequently in some regions and may significantly contribute to re-emissions and redistribution of POPs formerly stored in the sediments and agricultural soils (Holoubek et al. 2007; Pulkrabova et al. 2008; Noyes et al. 2009; Bogdal and Scheringer 2010). Biomass burning is an important linkage point between POPs and biogeochemical cycles of C and nutrients in terrestrial ecosystems affected by GCC. It not only can convert plant and soil organic matter to CO_2 (Kasischke et al. 1995; Westerling et al. 2006; Marlon et al. 2009; Turetsky et al. 2011; Zhang et al. 2011) but also can cause the emission of particulate matter and other gaseous pollutants such as CO, SOx, NOx, volatile organic compounds, and POPs (Chi et al. 2010; Grandesso et al. 2011).

The coupling of the OC and organic contaminant fluxes and budget in the global environment was recently addressed as one key scientific issue (Nizzetto et al. 2010). The biogeochemical cycles of POPs and OC may be linked in various ways (e.g., soil or sediment particles, soil–air exchange, plant accumulation, soil respiration, etc.) (Ver et al. 1999; Wegmann et al. 2004; Moeckel et al. 2008, 2009). Organic C pools in terrestrial subsurface layers represent the major active stores and sources of POPs. Some studies have shown that dissolved OC strongly affects the sorption and mobility of organic chemicals (Totsche et al. 1997; Flores-Cspedes et al. 2002), causing POPs in dissolved and particulate form to migrate by leaching to water bodies and deep sediments (Moeckel et al. 2008).

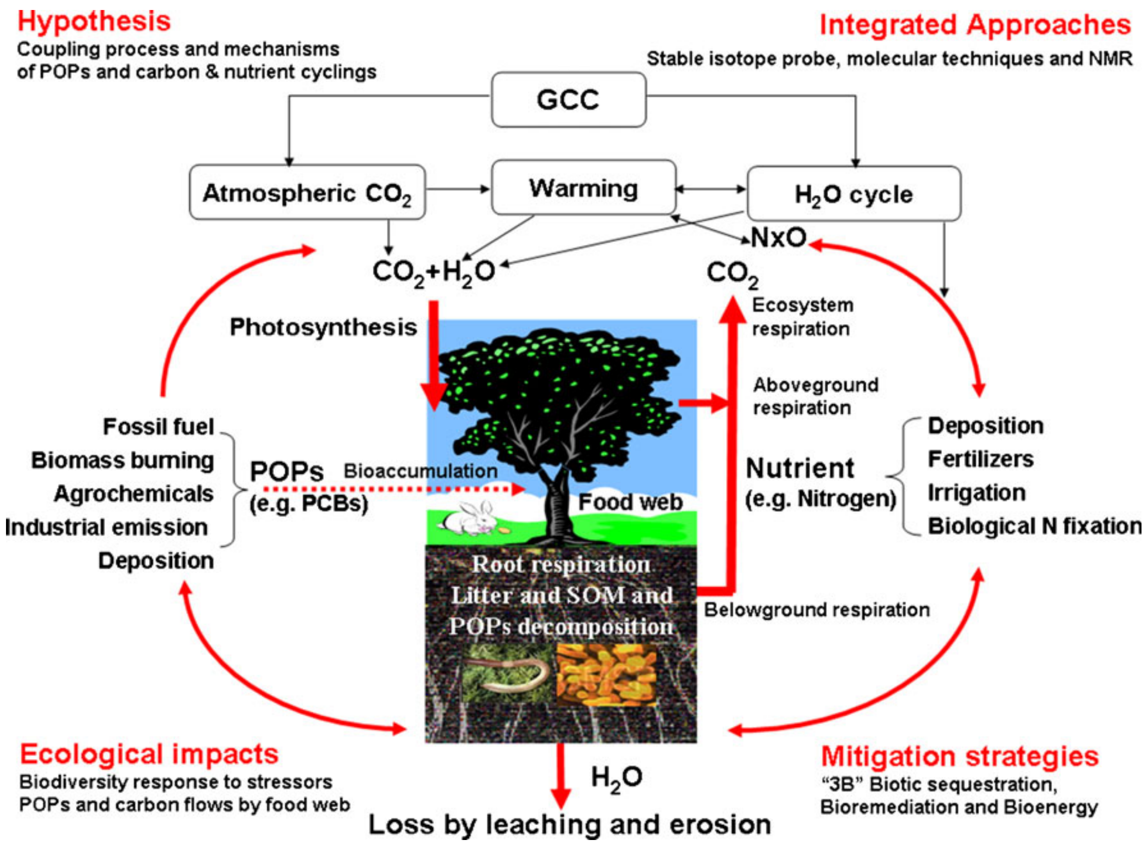

Fig. 1 The coupling between persistent organic pollutants (POPs) and biogeochemical cycles of carbon and nutrients in terrestrial ecosystems under global climate change (GCC)

Global warming may accelerate the release of POPs previously deposited in environmental media and enhance air–soil exchange of POPs (Bogdal et al. 2010; Hung et al. 2010). Soil–air exchange and partitioning are key processes controlling the fate and transport of POPs and terrestrial ecosystem C at regional and global scales (Cabrerizo et al. 2009, 2011). Terrestrial plants not only fix CO_2 as organic compounds through photosynthesis (Beer et al. 2010), but also sequester POPs from the atmosphere (McLachlan and Horstmann 1998), and transport them into forest soil C pools, which leads to the forest filter effect (FFE) (Nizzetto et al. 2008). Some recent studies investigated the vertical concentration profile of several global contaminants in the litters and soils, and showed that the litters represent a significant compartment for POPs mass balance, and moreover, the mass of POPs was associated with the active C pool over time (e.g., through a plant growing season) (Moeckel et al. 2008, 2009). However, little is known on the coupling relationship between plant POPs accumulation and C fixation in terrestrial ecosystems.

Soil respiration is the second largest terrestrial C flux induced by microbe and plant metabolism from the soil surface to the atmosphere (Bond-Lamberty and Thomson 2010). However, POPs partition and transformation are usually affected by soil respiration in terrestrial ecosystems. To date, only few studies have focused on POPs transferring and partitioning processes in soil organic and inorganic matrix at the microscale (Doick et al. 2005). It is worth noting that there are major uncertainties in the reaction of soil respiration to temperature and soil humidity and how this will affect the capacity of soils for biodegradation of POPs and OC turnover (Semple et al. 2007). Therefore, more insights are needed on the dynamics of POPs and C biogeochemical cycle to better understand their global and regional fluxes under global warming.

3 GCC affects both POPs and biogeochemical cycles of C and nutrients through food webs in terrestrial ecosystems

The food web is essential for maintaining life in the ecosystem, but environmental change affects food-web structure and ecosystem function (Petchey et al. 1999; Harmon et al. 2009). Ecologists and environmental scientists are now actively seeking ways to detect the movement of energy, nutrients, and contaminants through food webs (Elser et al. 2000; Sharpe and Mackay 2000; Kelly and Gobas 2003). Terrestrial organisms in the food web structure usually act as "bioreservoirs", "biovectors", and "biotransformers" of C, nutrients, and POPs (Nizzetto et al. 2010). Especially, the fungus-driven food web has important implications for the fate of soil organic C in temperate ecosystems under warmer scenarios (Briones et al. 2009). Changes in the food web structure may have important consequences for the biomagnification of POPs in the food webs. In terrestrial ecosystems, organisms at the top of the food web can adapt to habitat change and largely alter their exposure to POPs (Macdonald et al. 2005). Blankenship et al. (2005) found that the differential accumulation of PCB congeners in the terrestrial food web can be explained by congener-specific differences in bioavailability from the soil, exposure pathways, and metabolic potential of each of the food web components. Moreover, biotransportation (i.e., biovectors) of POPs often occurs and likely being global distributed because of changes in habitat at various scales (Bustnes et al. 2006; Evenset et al. 2007).

During bioaccumulation and biotransportation of POPs through the food web, POPs may be biotransformed by enzymatic systems into a different chemical species (Borga et al. 2004; Dang et al. 2010) with enhanced toxicity (Kay et al. 2005; Noyes et al. 2009), especially under higher temperature. Buckman et al. (2007) observed that rising temperature enhanced biotransformation of PCBs into toxicologically active hydroxylated PCB metabolites. Thus, a warmer climate will affect the toxicokinetics of POPs within organisms in the food web and the biomass turnover rate of each trophic level and will ultimately disturb the biogeochemical cycles of C and nutrients in the ecosystems. Unfortunately, due to a lack of information about ecosystems exposure to POPs under GCC, it is currently difficult to estimate accurately how GCC may impact wildlife exposure to POPs. Morrissey et al. (2010) used stable isotope and contaminant analyses to reveal differences in nutrient sources and contaminant pathways in two species of dipper in western Canada and western Britain. Di Paolo et al. (2010) highlighted the importance of including black C (BC) as an adsorbing phase to study the dynamics of biotransformation and bioformation of polybrominated diphenyl ethers (PBDEs) in an estuarine food web. There are large uncertainties concerning how GCC has affected ecosystems food web structures. Therefore, we will further focus on how GCC affect the trophic structure and POPs fluxes in the ecosystems.

4 The vulnerability and adaptation of biodiversity in terrestrial ecosystems to POPs pollution and GCC stressors

Biodiversity in terrestrial ecosystems plays a key role in the maintenance of C and nutrient cycles (Müller et al. 2010; Nielsen et al. 2011). However, there is a significant increase in the rate of biodiversity loss induced by GCC and other environmental stressors such as POPs pollution (Butchart et al. 2010; Curran et al. 2011; Dawson et al. 2011). Thus, assessing the vulnerability and possible adaptation of biodiversity to environmental changes is necessary to further understand the biogeochemical cycles of C and nutrients and their influence on POPs dynamics and to mitigate GCC effects and POPs pollution. A result by IPCC (2007) showed that approximately 20% to 30% of plant and animal species assessed so far are likely to be endangered if increases in global average temperature exceed 1.5°C to 2.5°C. In some tropical regions characterized by high biodiversity, many species may be susceptible to the exposure of multiple stressors (e.g., GCC, habitat loss and fragmentation and pesticide pollution). More appropriate conservation actions will result from taking into account all these aspects of vulnerability. On the other hand, the adaptation of species takes place through adjustments to reduce vulnerability or enhance resilience in response to GCC and associated environmental stressor (IPCC 2007). Adaptation is selective and can take advantage of positive impacts and reduce negative ones (Goklany 2005), especially biological adaptation is more effective. Therefore, biological adaptation strategy should be adopted to develop some environmentally friendly management techniques (e.g., biological C sequestration, bioenergy, bioremediation, namely the "3B" technique) for enhancing biodiversity and mitigating GCC and environmental stressor.

Biological C sequestration (BCS) refers to the assimilation and storage of atmospheric CO_2 in the vegetation, soils, woody products, and aquatic environments (Lal 2004; Gitz et al. 2006), which has the potential to offset the global fossil fuel emissions. For example, C uptake by forests contributed to a "residual" 2.6 Pg C year^{-1} terrestrial C sink in the 1990s, about 33% of anthropogenic C emissions from fossil fuel and land-use change (Bonan 2008). Soil C sink capacity of managed ecosystems approximately equals the cumulative historic C loss estimated at 55 to 78 Gt (Lal 2004). Furthermore, Jackson et al. (2005) highlight that C sequestration strategies through tree plantations should be considering their full environmental consequences. The national assessment for biological C sequestration in the USA will be conducted in the course of the next 3–4 years. At the same time, biofuels are being promoted as an important part of the global energy needed to reduce fossil fuels use and to decrease anthropogenic greenhouse gas fluxes (Vuichard et al. 2009; Barton et al. 2010). Many studies evaluated the C mitigation potentials of biofuels through cultivation technologies and ecological vulnerability (e.g., land use change and biodiversity loss, etc.) (Searchinger et al. 2008; West et al. 2010; Mullins et al. 2011) and compared them to the generated "carbon debt" when clearing natural ecosystems for cultivating biofuels (Fargione et al. 2008). Tilman et al. (2006) found that biofuels derived from the low-input high-diversity (LIHD) mixtures of native grassland perennials can provide more usable energy, greater greenhouse gas reductions, and less agrochemical pollution per hectare than can corn grain ethanol or soybean biodiesel. Moreover, biofuels can be produced on agriculturally degraded lands and thus both protect the habitat of biodiversity and environment. In addition, the energy crops used for phytoremediation are attractive (Witters et al. 2011), which could turn phytoremediation into a profit-making operation. However, information about phytoremediation applications based on biological energy is rather limited and we still seldom assess its biological C sequestration potential in mitigating GCC.

5 Integrated approaches to reveal the coupling mechanisms between POPs and biogeochemical cycles of C and nutrients

The development of integrated techniques (e.g., stable isotope, biomarker, modeling, etc.) in terrestrial ecosystem research is necessary to fully understand the mechanisms, functioning, and trends of POPs and biogeochemical cycles of C and nutrients. In the last two decades, stable isotope techniques have been widely used to study the biogeochemical processes such as C cycle and primary productivity, nutrient cycling, microbial community functioning, transportation and biodegradation of pollutions, the hydrological cycle, and terrestrial and aquatic food chain (Näsholm et al. 1998; Pace et al. 2004; Dickhut et al. 2004; Govindarajulu et al. 2005; Morin et al. 2008). Recent advances in the application of molecular genetic approaches have provided another powerful tool to analyze potentially huge microbial diversity in natural environments (Anderson and Cairney 2004; He et al. 2009; Zhang et al. 2009); however, this gives no direct information about the biogeochemical processes in which microorganisms are active. A combination of stable isotope probing (SIP) and biomarker-based fingerprinting can be a powerful approach to directly link the C (^{13}C) biogeochemical process with active specific microorganisms groups in natural environments (Boschker et al. 1998; Lu and Conrad 2005; Xu et al. 2009; Sun et al. 2010). With the ^{13}C-labeled tracers available, nuclear magnetic resonance (NMR) spectroscopy technique has been increasingly used in ecology, geochemistry, and environmental science (Mathers et al. 2000; Hedges et al. 2001; Käcker et al. 2002; Chen et al. 2004; Johnson et al. 2005). These approaches provide more structural information for biogeochemical cycles of C and organic pollutants in terrestrial ecosystems, particularly when combined with stable isotope and bio-molecular techniques. On the other hand, modeling is very useful to integrally understand and predict the C and POPs biogeochemical cycling processes at a global and regional scales under GCC (Cao and Woodward 1998a; Thomas et al. 2005; MacLeod et al. 2005; Lamon et al. 2009), including Dynamic Global Vegetation Models (DGVMs), General Circulation Model (GCM) and Berkeley-Trent Global Multimedia Mass Balance Model (BETR Global). The environmental fate and transportation of POPs are to some extent controlled by the C biogeochemical cycle in terrestrial ecosystems, which should be considered in global C cycle models in terrestrial ecosystems.

6 Conclusions

There is increasing evidence that GCC has significantly affected the C and nutrient cycling processes in terrestrial ecosystems. However, GCC impacts on the environmental fate and biological effects of POPs are not well understood, particularly in terms of the relationship between POPs and biogeochemical cycles of C and nutrients. Therefore, further research questions should focus on the dynamics of POP environmental fate and C biogeochemical cycle under different GCC scenarios. These questions would answer how GCC affects the trophic structure of ecosystems and food web magnification of POPs, and would address the effects of multiple stressors on the vulnerability and adaptation of biodiversity, and the development of integrated techniques (e.g., stable isotope, biomarker, NMR, modeling, etc.) could help unravelling the links between POPs and the C cycle. It is worth pointing out that the hydrological cycle is a key ecosystem process which drives the C and nutrient cycles and POPs dynamics under global warming, and therefore needs to be considered when we study the above mentioned issues. In addition, developing environmentally friendly management strategies (“3B” technique) is urgently required for biodiversity conservation and mitigating GCC and environmental stressors.

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