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Effects of input uncertainty and variability on the modelled environmental fate of organic pollutants under global climate change scenarios

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HIGHLIGHTS

- The effects of uncertainty and variability on modelled fate of PCBs are assessed.
- Uncertainty in degradation dominates variance of projected absolute fate of PCBs.
- Climate parameters dominate variance of climate change/baseline scenario ratios.
- Model predictions change by up to a factor of 2 due to climate change.

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ABSTRACT

Global climate change (GCC) is expected to influence the fate, exposure and risks of organic pollutants to wildlife and humans. Multimedia chemical fate models have been previously applied to estimate how GCC affects pollutant concentrations in the environment and biota, but previous studies have not addressed how uncertainty and variability of model inputs affect model predictions. Here, we assess the influence of climate variability and chemical property uncertainty on future projections of environmental fate of six polychlorinated biphenyl congeners under different GCC scenarios using a spreadsheet version of the ChemCAN model and the Crystal Ball® software. Regardless of emission mode, results demonstrate: (i) uncertainty in degradation half-lives dominates the variance of modelled absolute levels of PCB congeners under GCC scenarios; (ii) when the ratios of predictions under GCC to predictions under present day climate are modelled, climate variability dominates the variance of modelled ratios; and (iii) the ratios also indicate a maximum of about a factor of 2 change in the long-term average environmental concentrations due to GCC that is forecasted between present conditions and the period between 2080 and 2099. We conclude that chemical property uncertainty does not preclude assessing relative changes in a GCC scenario compared to a present-day scenario if variance in model outputs due to chemical properties and degradation half-lives can be assumed to cancel out in the two scenarios.

1. Introduction

Global climate change (GCC) is expected to impact humans and the environment that they depend on (IPCC, 2007). One concern is GCC-induced impacts on the fate of pollutants, and the corresponding impacts on the direct exposure of both human and wildlife to altered environmental concentrations (Macdonald et al., 2005; Noyes et al., 2009; UNEP/AMAP, 2011). Macdonald et al. (2005) reviewed climate change-induced impacts on pollutant pathways with a particular focus on the Arctic region, and more recently a UNEP/AMAP expert group released a review of how GCC may impact persistent organic pollutants from primary emission to degradation in both physical and biological media (UNEP/AMAP, 2011) on the global scale.

Multimedia chemical fate models that reflect our understanding of how organic chemicals behave in the natural environment have been developed and applied since the 1990s (Diamond et al., 1992; Wania and Mackay, 1999; MacLeod et al., 2010). These models have been developed to simulate the fate and behaviour of organic chemicals based on historical emissions and also to forecast future time trends (Dalla Valle et al., 2007; Armitage et al., 2009). Modelling assessments incorporating future GCC projections have provided insights into potential GCC-induced impacts on chemical fate and exposure, both in the physical and biological environment. Gouin et al. (2013) provided an extensive review of the relevant modelling studies.

Multimedia fate models rely on two major groups of input parameters to calculate the environmental fate of organic chemi-

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cals: (i) chemical-specific properties and (ii) environmental parameters (Mackay, 2001). Large datasets of both chemical and environmental parameters have become available in recent years. For example, future projections of climate variables such as temperature, wind speed, and precipitation have been made publically available by the Intergovernmental Panel on Climate Change (IPCC, 2000). Furthermore, numerous efforts have been made to improve the quality of data for chemical-specific properties aiming to facilitate chemical fate and risk assessment (Beyer et al., 2002; Li et al., 2003; Schenker et al., 2005; MacLeod et al., 2007). However, it is well known that there are large uncertainties associated with the available datasets for these parameters, where variability is particularly relevant for climate variables, and uncertainty for chemical properties. It is thus important to confront uncertainty and variability when assessing the environmental fate and levels of organic pollutants using models (McKone and Bogen, 1991; MacLeod et al., 2002; Hollander et al., 2009).

Previous studies using multimedia fate models have generally found that chemical properties and degradation half-lives are much more important in determining modelled concentrations than environmental conditions (McKone et al., 1996; Hertwich et al., 1999; MacLeod et al., 2002; Hollander et al., 2009; Sommerfreund et al., 2010; Kim et al., 2012). This result raises the question of whether the effects of GCC on pollutants are likely to significantly perturb forecasts of future concentrations. The specific goals of this work are to determine: (i) how the two input parameter groups, climate and chemical property, influence the modelled outcomes under GCC scenarios and (ii) to what extent the variability associated with climate and uncertainty of chemical properties contribute to the variance of projected GCC-induced changes in model outcomes. This work builds upon previously published modelling studies that forecasted the influence of GCC on the fate of organic pollutants by including an analysis of uncertainty and variability and identifying cases where GCC scenarios lead to significantly different model outcomes compared to a base-case climate scenario.

2. Material and methods

2.1. Model and parameterisation

We selected the spreadsheet ChemCAN model for this study as a representative Level III multimedia model, and applied it to the Japan region. ChemCAN has been applied in many studies for various purposes, and proved to be capable of producing useful information for chemical fate assessments (Mackay et al., 1991; MacLeod and Mackay, 1999; Webster et al., 2004). In particular, Kawamoto et al. (2001) parameterised ChemCAN for Japan to investigate the fate of 68 chemicals with emissions reported to the Japanese National Pollutant Release Inventory. They observed general agreement between the modelled and observed chemical concentrations in the region. Their study provides an added degree of confidence in the model parameterisation for Japan with respect to landscape properties. We therefore adopted the same parameterisation (Table A.1) and applied ChemCAN to the same region (Fig. A.1).

In order to comprehensively evaluate the influence of climate variability and uncertainty in chemical properties and degradation half-lives, several adaptions have been made to take into account missing climate-dependent processes in the ChemCAN model. Wind speed-dependent mass transfer at the diffusive boundary between air and water is integrated into ChemCAN using the nonlinear algorithm recommended by Mackay and Yeun (1983). The residence time of air is changed to vary with wind speed (Eqs. (A.4)-(A.6)). As this study does not aim to compare modelled and observed chemical concentrations, an illustrative emission rate of 1000 kg year^{-1} is assumed in all model calculations for the baseline climate scenarios. The effect of temperature on chemical properties, degradation half-lives and emissions in the GCC scenario is taken into account by using van't Hoff-type equations:

$$\ln X = \ln X_r + \Delta E/R \times (1/T_r - 1/T) \tag{1}$$

where X_r and X represent any property or emission rate at the reference temperature (of 25 °C) and an actual environmental temperature, respectively, R is the gas constant (8.314 J K^{-1} mol^{-1}), and ΔE (J mol^{-1}) represents the various energy required for temperature correction (Table 1).

We focus on 13 model outcomes to assess the influence of climate variability and chemical property uncertainty. The four media-specific bulk concentrations (C_a, C_w, C_s, C_{sed}; mol m^{-3}, where subscripts a, w, s and sed are air, water, soil and sediment, respectively) are of interest because they can determine directly the exposure levels of wildlife and humans. The four media-specific percentage distributions ($\%_a$, $\%_w$, $\%_s$, $\%_{sed}$) indicate where the majority of emitted chemical is retained. The overall (*Ove*), reactive (*Rea*), and advective (*Adv*) persistence (days) have been widely used as measures of the time scale of a chemical leaving an environment as a result of degradation and advective transport (Eqs. (A.1)-(A.3)). The two long-range transport potential (LRTP) indicators (characteristic travel distance (*CTD*, km) and transfer efficiency (*TE*, %)) are of interest because they indicate a generic travel distance and how much of a chemical can reach a certain distant target region, respectively (Wegmann et al., 2009; Eqs. (A.7) and (A.8)).

We further adapted ChemCAN to derive the ratios of 13 model outcomes under the GCC scenario compared to model outcomes under the present climate conditions.

$$y/Y \tag{2}$$

where the small y and capital Y represent any model outcome obtained under the GCC scenario and the present day climate, respectively (see Section 2.3). Such ratios have previously been used to indicate GCC-induced changes in modelled environmental fate of persistent organic contaminants (Lamon et al., 2009; Wöhrnschimmel et al., 2013).

2.2. Uncertainty in chemical properties and climate variability

We selected six polychlorinated biphenyl (PCB) congeners for assessment. PCBs are prototypical persistent organic pollutants (POPs), and the effect of GCC on POPs is an area of current research and regulatory interest (UNEP/AMAP, 2011). The key chemical properties chosen for this study are the temperature-dependent solubility in water (*SW*), vapour pressure (*VP*), octanol–water partition coefficient (K_{OW}) and four environmental media-specific degradation half-lives (*HL*; Table 1). The climate variables chosen include temperature (T), precipitation (P) and wind speed (u). They are defined as $T = T' + \Delta T$, $P = P' + \Delta P$, and $u = u' + \Delta u$, where T', P', and u' represent long-term observational arithmetic means, and ΔT, ΔP, and Δu represent projected future anomalies of the long-term means (Tables A.2 and A.3). The long-term means are considered to represent the present day climate, and the projected future anomalies represent the shift due to GCC. Model outcomes based on the sum of the long-term means and future anomalies of climate variables are considered to represent future forecasts of the environmental fate of studied PCB congeners.

We use confidence factors (*Cf*) to characterise log-normal distributions and to quantify the climate variability, and uncertainty in chemical properties and degradation half-lives:

$$Cf = exp\left\{2 \times \left[\ln((\sigma/\mu)^2 + 1)\right]^{1/2}\right\} \tag{3}$$