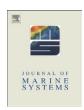
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The distribution of persistent organic pollutants in a trophically complex Antarctic ecosystem model



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ABSTRACT

Despite Antarctica's isolation from human population centres, persistent organic pollutants (POPs) are transported there via long range atmospheric transport and subsequently cold-trapped. The challenging nature of working in the Antarctic environment greatly limits our ability to monitor POP concentrations and understand the processes that govern the distribution of POPs in Antarctic ecosystems. Here we couple a dynamic, trophically complex biological model with a fugacity model to investigate the distribution of hexachlorobenzene (HCB) in a near-shore Antarctic ecosystem. Using this model we examine the steadystate, and annual cycle of HCB concentration in the atmosphere, ocean, sediment, detritus, and 21 classes of biota that span from primary producers to apex predators. The scope and trophic resolution of our model allows us to examine POP pathways through the ecosystem. In our model the main pathway of HCB to upper trophic species is via pelagic communities, with relatively little via benthic communities. Using a dynamic ecosystem model also allows us to examine the seasonal and potential climate change induced changes in POP distribution. We show that there is a large annual cycle in concentration in the planktonic communities, which may have implications for biomagnification factors calculated from observations. We also examine the direct effects of increasing temperature on the redistribution of HCB in a changing climate and find that it is likely minor compared to other indirect effects, such as changes in atmospheric circulation, sea ice dynamics, and changes to the ecosystem itself.

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1. Introduction

Antarctica is one of the most isolated and apparently pristine regions on earth. However, toxic anthropogenic pollutants, such as persistent organic pollutants (POPs), are present (Bengtson Nash, 2011). The semi-volatile nature of most POPs, combined with their resistance to degradation, mean that they are capable of long range environmental transport via atmospheric circulation (Wania and Mackay, 1996; Lohmann et al., 2007). POPs emitted at temperate and tropical latitudes undergo global "distillation" along latitudinal temperature gradients, depositing out of the atmosphere according to their volatility. Ultimately, fractions that reach the poles experience "cold trapping" and settle in water and sediments, and may subsequently be taken up by biota (Wania and Mackay, 1996).

Polar regions have long been considered an environmental sink for POPs (Dachs, 2011). Restrictions of emissions, in concert with increasing temperatures, mean that POPs that were once cold trapped in the Arctic are possibly being remobilised (Ma et al., 2011), with some evidence that similar processes are occurring in the Antarctic (Cabrerizo et al., 2013). The interplay between various factors affecting the distribution of POPs in a changing polar climate is complex, however, ecological changes (e.g. primary productivity) are likely very important (Armitage et al., 2011).

POPs distribute between different environmental phases, including biological phases where their lipophilicity causes them to bioconcentrate and biomagnify, with potential associated toxic effects. The dependence of polar species on lipid-rich energy sources makes them particularly vulnerable to these effects (Goerke et al., 2004; Borgå et al., 2004). However, the significant seasonal variations in light and temperature that are characteristic of polar regions means that there may also be, as yet ill-defined, seasonal variation in the POP exposure of polar marine biota (Cropp et al., 2011).

The difficulties associated with conducting Antarctic field studies means that there is a great paucity of observations of POPs in both space and time. Furthermore, the little data that is available is

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difficult to compare due to differences in methodologies (Corsolini, 2009). In particular, the extreme difficulty of collecting samples in Antarctica during the austral winter means that there is little information about the seasonal variability of POP concentrations in Antarctic systems.

Mathematical models can be used to understand the mechanisms influencing the distribution of POPs in natural systems and to help fill knowledge gaps in places like the Antarctic, where taking observations is extremely difficult. Multimedia models of POPs are common (Wania and Mackay, 1999), and have been used extensively to study the POP burden of biota for at least the last 25 years (Thomann, 1989; Thomann et al., 1992; Gobas, 1993; Campfens and Mackay, 1997), and more generally the environmental burden for at least 35 years (Mackay and Paterson, 1981; Mackay, 2001). Whilst, to the best of our knowledge, this is the first comprehensive study of POPs in a marine Antarctic ecosystem, there are a number of modelling studies that have examined POPs in the Arctic. Borgå and Di Guardo (2005) used the steady state model of Campfens and Mackay (1997), adapted to the Barents Sea, to examine the bioaccumulation of polychlorinated biphenyls (PCBs). They used the inability of the model to reproduce observed concentrations to help identify that more studies were required to more accurately determine the PCB concentration in polar waters, de Laender et al. (2010) used the model of Hendriks et al. (2001) to examine the seasonal variability of PCBs in the Barents Sea by running their model under various conditions typical of different seasons and locations. Borgå et al. (2010) used a model to examine the effect of climate change on an Arctic ecosystem that comprised seven biological groups (phytoplankton, two types of copepod, krill, an amphipod, a fish, and a piscivorous seabird) finding that the effects of increased temperature and primary production were different for different chemicals. In their model there was very little change in δ -hexachlorocyclohexane burdens whilst PCB-153 dropped to approximately half of present-day concentrations in the piscivorous seabird.

As with the aforementioned studies we couple an ecosystem model to a fugacity model, however, our approach differs in two major ways. Firstly, our ecosystem model covers the full trophic spectrum from primary producers to apex predators, a method known as end-to-end modelling (Fulton, 2010). Secondly, our model ecosystem solves for the abundance of functional groups (measured by the population's total organismal mass of nitrogen), which in turn informs the growth and loss terms in the model equations (along with, for autotrophs, the availability of light and inorganic nutrient), whereas all of the aforementioned studies essentially have a static food web in which feeding, excretion, volume dilution, etc. are specified as model parameters. Our methodology facilitates the investigation of transient states, which is an intrinsic part of the boom-and-bust annual cycle of polar ecosystems, as well as for climate change.

Our approach of using a dynamic, population-scale ecosystem model has had limited use to date by the POP modelling community (Guglielmo et al., 2009; Cropp et al., 2011; Lammel and Stemmler, 2012), however, it is widely used in fisheries models (e.g. Fulton, 2010, and references therein) and models of biogeochemical cycling (e.g. Denman, 2003; Hood et al., 2006; Hashioka et al., 2013). A strong motivating factor for us to use a population approach, rather than an individual approach, is the inclusion of plankton which, due to their sheer numbers (in plankton blooms in the Southern Ocean diatoms, for example, commonly reach 6.5×10^5 individuals per litre of seawater; Kopczynska et al., 1986) makes modelling of individuals impractical. Previous studies that have included plankton in their food web models have built their models around an average individual (e.g. Thomann, 1989; Borgå et al., 2010), however, such an approach is incompatible with having a dynamic food-web as information about the (time varying) abundance of individuals is also

necessary with our approach and is ultimately dependent on forcing (i.e. environmental conditions such as temperature and light).

Since our ecosystem model solves for the population scale, rather than the dynamics of individuals in that population, intra-population fluxes of POPs (e.g. birthing and lactation) are not explicitly dealt with as they do not represent a net gain or loss of POP for a given population in the fugacity model. In this sense, we are modelling each population (more properly, from the perspective of the fugacity model, the volume of lipid of each population) as a single environmental phase. Due to the dynamic nature of our model the volume of each lipid (or biological) phase changes dynamically, depending on the abundance of a functional group. Therefore, in our model volume dilution is not specified explicitly but instead is taken into account (along with concentration amplification) in the volume correction term in our model equations (Bates et al., 2016a).

In order for us to be able to consider the population as a whole, we must ensure that the model domain is of sufficient size such that the population contained within is large enough that it is not necessary to consider individuals (Bates et al., 2015). This is analogous to the continuum versus molecular descriptions of a fluid, where, once enough molecules are present it is no longer necessary to consider the behaviour of individual molecules but instead their collective behaviour can be described as a continuum.

In this study we used a numerical model to better understand the distribution of hexachlorobenzene (HCB) in a near-shore Antarctic marine environment. Specifically, the aims of this study were to:

- 1. Examine the distribution, bioconcentration, biomagnification of HCB in a model Antarctic ecosystem,
- 2. Identify the major pathways of HCB through the model food web
- 3. Investigate the seasonal variability of HCB concentrations, and
- 4. Explore how the distribution of HCB may change under the influence of a changing climate.

HCB is an organochlorine that was historically used as a fungicide, especially to control wheat bunt, and was one of the initial 12 POPs listed under the Stockholm Convention (UNEP, 2001). We modelled HCB because it has repeatedly been found to be the dominant POP compound in ambient Antarctic air (Kallenborn et al., 2013; Wild et al., in preparation) and accumulating in Antarctic biota (Bengtson Nash et al., 2008, 2013; Waugh et al., 2014). Using HCB further allows us to build on the work of Cropp et al. (2011) who used a simple plankton model to look at the distribution of HCB in different phases of an Antarctic marine environment.

2. Model description

For this study we used the trophically complex, near-shore Antarctic ecosystem model developed by Bates et al. (2015) which consists of 21 functional groups (i.e. groups of biota that perform similar ecological functions). We coupled this biological model to a fugacity model, where each functional group in the biological model is a chemical compartment. In addition, there are a number of physical compartments, namely the atmosphere, ocean, and sediment. Table 1 lists all of the populations and physical compartments along with the abbreviations we use for each group.

The model uses a limiting nutrient (nitrogen) as the model currency. Thus, all populations are measured in terms of the population's total organismal mass of nitrogen, rather than biomass or number of individuals. This approach ensures that the total nutrient mass in our model is conserved. For further details the reader is referred to Bates et al. (2015, 2016b) as well as Cropp and Norbury (2009, 2012, 2013). Table 1 lists the model's steady state (i.e. no seasonal cycle) distribution of organismal nitrogen mass in the model ecosystem.

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