Marine Pollution Bulletin 85 (2014) 824–830

Contents lists available at ScienceDirect

Marine Pollution Bulletin

journal homepage: www.elsevier.com/locate/marpolbul

# Modelling exposure of oceanic higher trophic-level consumers to polychlorinated biphenyls: Pollution 'hotspots' in relation to mass mortality events of marine mammals

Itsuki C. Handoh [a,*], Toru Kawai [b]

[a] Center for Research Development, Research Institute for Humanity and Nature, 457-4 Motoyama, Kamigamo, Kita-ku, Kyoto 603-8047, Japan
[b] Center for Environmental Risk Research, National Institute for Environmental Studies, 16-2 Onogawa, Tsukuba 305-8506, Japan

ARTICLE INFO

*Article history:*
Available online 9 July 2014

*Keywords:*
Chemical pollution
Polychlorinated biphenyls
Multi-media model
Mortality events of marine mammals
Critical exposure level
Planetary Boundaries

ABSTRACT

Marine mammals in the past mass mortality events may have been susceptible to infection because their immune systems were suppressed through the bioaccumulation of environmental pollutants such as polychlorinated biphenyls (PCBs). We compiled mortality event data sets of 33 marine mammal species, and employed a Finely-Advanced Transboundary Environmental model (FATE) to model the exposure of the global fish community to PCB congeners, in order to define critical exposure levels (CELs) of PCBs above which mass mortality events are likely to occur. Our modelling approach enabled us to describe the mass mortality events in the context of exposure of higher-trophic consumers to PCBs and to identify marine pollution 'hotspots' such as the Mediterranean Sea and north-western European coasts. We demonstrated that the CELs can be applied to quantify a chemical pollution Planetary Boundary, under which a safe operating space for marine mammals and humanity can exist.



## 1. Introduction

Chemical pollution has long been a serious threat to ecosystems and to human health and wellbeing and has been included as one of nine items listed in the Planetary Boundaries framework for global sustainability (Rockström et al., 2009). There are ample scientific data on individual chemicals, but an aggregate, global-level analysis is lacking, hence the Planetary Boundary for chemical pollution has not been quantified (Persson et al., 2013). Potential variables and/or endpoints include the amounts of persistent organic pollutants (POPs), plastics, endocrine disrupters, heavy metals and nuclear waste emitted to, and their concentrations in, the global environment, and the resulting effects on ecosystems and the functioning of Earth systems. One of the most readily quantifiable variables among these may be the response of an ecosystem to exposure to persistent toxic substances that have been listed in the Stockholm Conventions on POPs.

Quantification of POPs exposures in environmental media is relatively straightforward owing to rapid progress in chemical pollution studies in recent years. These have led to, for example, global emission inventories (Breivik et al., 2007), International Pellet Watch (Ogata et al., 2009; Heskett et al., 2012), and global and regional multi-media modelling of POPs (Hollander et al., 2008; Ilyina et al., 2008; Task Force on Hemispheric Transport of Air Pollution. Global and regional modelling of POPs, 2010; Kawai et al., 2014; O'Driscoll et al., 2013). In contrast, the response of ecosystems to chemical exposures is poorly understood. Nevertheless, attempts have been made to examine the relationships between toxic chemical concentrations and obvious occurrences of ecological damage such as epizootic and mass mortality events of marine mammals, most notably of harbour seals (*Phoca vitulina*) in Europe in 1988 and 2002 (Härkönen et al., 2006) and of the Mediterranean striped dolphins (*Stenella coeruleoalba*) in 1990–1992 and 2006–2008 (Rubio-Guerri et al., 2013) in which the phocine distemper virus and dolphin morbillivirus played a major role. Mortalities of marine mammals have been attributed to other causes, such as shellfish poisoning (e.g., by brevietoxin), induced by harmful algal blooms (González et al., 2002; Flewelling et al., 2005; Fire et al., 2011), parasites, and some pathologies associated with nutritional status, and to a lesser extent, to anthropogenic causes such as ship collisions, interactions with fisheries activities, and military maneuvers using sonar (Conn and Silber, 2013; Arbelo et al., 2013; Goldbogen et al., 2013).

The working hypothesis causally linking chemical pollution and mass mortality is that higher trophic-level marine mammals may have been particularly susceptible to infection because their immune systems were suppressed through the bioaccumulation

* Corresponding author. Tel.: +81 75 707 2496; fax: +81 75 707 2513.
*E-mail address:* itsuki@chikyu.ac.jp (I.C. Handoh).

http://dx.doi.org/10.1016/j.marpolbul.2014.06.031
0025-326X/

of environmental contaminants such as polychlorinated biphenyls (PCBs) (Van Loveren et al., 2000; Jepson et al., 2005; Kajiwara et al., 2008). Several case studies have provided robust evidence to support this hypothesis (Lie et al., 2004; Hall et al., 2006; Raga et al., 2008). However, the spatial and temporal distribution of mortality events has not been compared to those of the exposure to PCBs of higher trophic-level consumers despite the availability of global multi-media models.

In this paper, largely using literature data for mortality events and a state-of-the-art multi-media model for PCBs, we demonstrate model-predicted exposure of higher trophic-level consumers to PCBs in the oceans, and identify pollution 'hotspots' in relation to mass mortalities of marine mammals.

## 2. Materials and methods

### 2.1. Mass mortality events data

There is no precise definition of a 'mass mortality event' of a marine mammal. The criteria for 'mass' vary with the species of interest because each has a different background population. The International Union for Conservation of Nature Red List of Threatened Species (IUCN Red List) categorises a number of marine mammals as Vulnerable (VU), Endangered (EN), or Critically Endangered (CR). Mediterranean monk seals (*Monachus monachus*), southern sea otters (*Enhydra lutris kenyoni*), sperm whales (*Physeter macrocephalus*), and Florida manatees (*Trichechus manatus latirostris*) are such threatened species. For simplicity, here we define a mass mortality event as the death of more than 100 adults per year for the species of interest.

We conducted a systematic literature review to compile past mortality events of marine mammals using the Thomson Reuters Web of Science system and e-resources. The keywords employed here were generic terms such as 'mortality', 'stranding', 'marine mammals', 'seals', 'whales', and 'dolphins'. There were more than 70 peer-reviewed papers and technical reports available for our purpose, although this could have been subject to a sampling bias such that some mass mortality events are more likely to be found than others. The location, the onset and cessation, the species, and the number of dead bodies for each event were extracted. The number of stranded animals was not considered unless it was identical to that of dead and/or diseased animals. There were 33 species suffering from mortality events, of which the majority were Odontoceti (13 species) and Pinnipedia (13 species). Of the 33 species, 9, 3, and 17 are currently classified as threatened (VU, EN, and CR), near threatened (NT), and of least concern (LC), respectively, in the IUCN Red List. There were no extinct species. Any evidence of virus or parasite involvement and the trophic level (TL) of the species (Pauly et al., 1998; Kelly et al., 2007; Pauly and Watson, 2005) was also noted.

We also compiled the reported PCBs concentration data sets from the open literature (Borrell et al., 2007; Hansen et al., 2004; Holsbeek et al., 1999; Kajiwara et al., 2001; Noël et al., 2009; Reich et al., 1999; Smyth et al., 2000; Ylitalo et al., 2001,2005), so as to extract the mean and standard deviation of PCB congener concentrations in the blubber of marine mammals at each available site, where the longitude, latitude, and year of sampling were known. There were 8 species over 25 sampling sites for adult males, and these were California sea lions (*Zalophus californianus*), Mediterranean monk seals (*M. monachus*), bottlenose dolphins (*Tursipos truncatus*), common dolphins (*Delphinus delphis*), harbour porpoises (*Phocoena phocoena*), killer whales (*Orcinus orca*), sperm whales (*P. macrocephalus*), and striped dolphins (*S. coeruleoalba*). Noting that PCB-bioaccumulation processes could differ significantly between males and females (Hickie et al., 2013), we excluded adult female and juvenile data sets.

### 2.2. Modelling PCBs exposure

We employed the Finely Advanced Transboundary Environmental model (FATE; Kawai et al., 2014) to quantify PCBs concentrations in oceanic Particulate Organic Matter (POM), and the exposure to PCBs of marine organisms from zooplankton communities (TL = 2) to top-predators such as killer whales (*O. orca*) (TL = 4.5) and polar bears (*Ursus maritimus*) (TL = 5.1). FATE is the most recently developed 3D dynamic multi-media model and is capable of predicting the global fate of PCBs in and across the atmosphere, oceans, soil, vegetation, and cryosphere. The modelled atmosphere and ocean compartments reproduce dynamic processes (advection and diffusion) with spatial resolutions of $2.5^\circ \times 2.5^\circ \times 20$ layers (1–0.01 in the sigma vertical coordinate system throughout the depth of troposphere) and $1.0^\circ \times 1.0^\circ \times 50$ layers (0–5500 m), respectively. The model also reproduces bioconcentrations in lower trophic-level organisms (phytoplankton and zooplankton) and other important biogeochemical processes in the ocean interior, dry and wet depositions, inter-media diffusive exchange of gaseous substances, degradations, phase partitions, and 1D transport across the vegetation–soil boundary (defoliation, infiltration, and diffusion).

Four PCBs congeners, PCBs#28, 105, 118, and 153 were selected for this study. The FATE model was forced by various input data sets including emission inventories, climate data, land cover and marine phytoplankton data, to facilitate evaluation of global PCB dynamics for the period 1931–2010. Data sets included the PCBs emission data of Breivik et al. (2007) and extensive climate data sets (see Kawai et al. (2014) for specific details of FATE). Sea-viewing Wide Field-of-view Sensor (SeaWiFS) data were used to estimate particulate organic carbon biomass (Stramska, 2009), the vertical carbon cycle in the oceans (Dunne et al., 2007), and the spatial distribution of the mean TL of the marine fish community including teleost fish and elasmobranchs (Jennings et al., 2008). Net primary production used in models of Dunne et al. (2007) and Jennings et al. (2008) was estimated by Carbon-based Production Model (CbPM; Westberry et al., 2008). An empirical biomagnification model (Kelly et al., 2007) was then applied to evaluate exposure to PCBs of the trophic-level animals of interest. For convenience, the trophic magnification factor (TMF) and the trophic level of POM were set at 3.55 (Toyoshima et al., 2009) and 1.5 (Miller et al., 2012), respectively. The model results were interpolated onto a $2.5^\circ \times 2.5^\circ$ grid.

We applied Bayesian uncertainty analysis (Kennedy et al., 2008) to FATE predictions for exposure to PCBs in order to quantify uncertainties in the model predictions which arise from uncertainties in the 15 model parameters such as those of plankton functional types and degradation constants for PCBs (Seto and Handoh, 2009). For each PCB congener 50 model runs were performed. FATE predictions for exposure to PCBs of the trophic-level animals of interest were then compared to the reported PCBs concentrations in the blubber of male marine mammals.

### 2.3. Critical exposure Level of PCBs

Here we introduce a critical exposure level (CEL) as a critical threshold or tipping point at which the state of marine mammals is qualitatively altered from normal to a mass mortality state. The CEL of PCB#$x$ concentration in $y$ ($C_y$), CEL(PCB#$x$, $C_y$), can be defined as:

$$CEL(PCB\#x, C_y) = \frac{\sum_{i=1}^{n} M_i C_{y,i}}{\sum_{i=1}^{n} M_i}, \quad (1)$$

where the suffix $y$ is either POM (TL = 1.5) and the fish community (TL = 3.5–3.8) in which PCBs are present and/or bioaccumulated. It