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Global deposition of airborne dioxin

Shawn Booth [a,*], Joe Hui [a], Zoraida Alojado [a], Vicky Lam [a], William Cheung [a], Dirk Zeller [a], Douw Steyn [b], Daniel Pauly [a]

[a] Sea Around Us Project, Fisheries Centre, University of British Columbia, 2202 Main Mall, Vancouver BC V6T-1Z4, Canada
[b] UBC Department of Earth and Ocean Sciences, 6339 Stores Road, Vancouver BC V6Y-1Z4, Canada

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ABSTRACT

We present a global dioxin model that simulates one year of atmospheric emissions, transport processes, and depositions to the earth's terrestrial and marine habitats. We map starting emission levels for each land area, and we also map the resulting deposits to terrestrial and marine environments. This model confirms that 'hot spots' of deposition are likely to be in northern Europe, eastern North America, and in parts of Asia with the highest marine dioxin depositions being the northeast and northwest Atlantic, western Pacific, northern Indian Ocean and the Mediterranean. It also reveals that approximately 40% of airborne dioxin emissions are deposited to marine environments and that many countries in Africa receive more dioxin than they produce, which results in these countries being disproportionately impacted. Since human exposure to dioxin is largely through diet, this work highlights food producing areas that receive higher atmospheric deposits of dioxin than others.

1. Introduction

Dioxins and furans consist of 210 structurally similar chemicals that are unintentional byproducts of combustion processes. Of these 210 chemicals, 17 have toxicological properties of concern to human and ecosystem health. The most potent form, 2,3,7,8-tetrachlorodibenzo-*p*-dioxin, is a known human carcinogen (International Agency for Research on Cancer, 1997) and the main exposure route to humans and other organisms is through diet (World Health Organization, 2007). Dioxins are one of the chemicals listed in the Stockholm Convention on Persistent Organic Pollutants. They are listed under Annex C, and, as such, parties must take measures to reduce the unintentional releases with the goal of continuing minimization and, where feasible, ultimate elimination (Olsen, 2003).

When dioxins are emitted to the atmosphere or land, they can subsequently undergo long range atmospheric transport until they are deposited to regions distant from the source (AMAP, 2004). This is due to the 'grasshopper effect' (Gouin et al., 2004) whereby dioxin previously deposited to land surfaces can undergo further transport through successive evaporation cycles. Dioxin has endocrine-disrupting effects and interacts with the components of ecosystems – it is lipophilic and concentrates in lipid tissues of living organisms causing health concerns for ecosystem components and people.

For toxicological purposes, dioxins can be expressed as toxic equivalents (TEQs) as the 17 forms of dioxins and furans of concern have different toxicological properties. TEQs express the toxicity of dioxins relative to the most toxic form – 2,3,7,8-tetrachlorodibenzo-*p*-dioxin. For human health purposes, dioxins have a tolerable daily intake (TDI) of 1–4 picograms-TEQ per kilogram body weight and day, but the United Nations has recommended that the intake of dioxin should be reduced to the lowest possible level because subtle effects can occur at levels of 2–6 picograms-TEQ per kilogram body weight and day (van Leeuwen et al., 2000).

Despite its known toxicity and adverse human health effects, there is little data concerning the global impacts of dioxin. Global emission inventories of dioxin production are incomplete and most inventories are available only for the year 1999 (United Nations, 1999). The importance of this study is that it (1) identifies on a global basis where the most heavily impacted terrestrial and marine ecosystems are likely to be, (2) helps to identify key regions where data are missing that could be of concern, and (3) highlights food production areas that receive higher inputs of dioxin than others. The purpose is to lay the foundation for further research into the effects of dioxin in marine waters and the food webs in marine ecosystems.

2. Methods

The cycling of dioxin in our global model involves the production of dioxin over land, the dispersion of dioxin through

* Corresponding author. Tel.: +1 604 707 0957; fax: +1 604 822 8934.
E-mail addresses: shawnrgbooth@gmail.com (S. Booth), joe_pw_hui@telus.net (J. Hui), z.alojado@fisheries.ubc.ca (Z. Alojado), v.lam@fisheries.ubc.ca (V. Lam), w.cheung@fisheries.ubc.ca (W. Cheung), d.zeller@fisheries.ubc.ca (D. Zeller), dsteyn@eos.ubc.ca (D. Steyn), d.pauly@fisheries.ubc.ca (D. Pauly).

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the atmosphere, the deposition of dioxin onto land and water, and the transportation of dioxin from land in water basins to coastal waters. For methodological details, see the Supplementary material. We use weekly averaged data for dioxin production and atmospheric dispersion. We simulate one year of production, dispersion, deposition and transport of dioxin (Fig. 1) and display the results for land and marine areas separately on a global map with a resolution of 259,200 ½° by ½° cells (79,296 terrestrial, and 179,904 marine). Since some countries are more heavily impacted by dioxin than others, we compare between countries by using a deposition to emission ratio. For each country, the amount of dioxin deposited to land and ocean cells over the one year simulation was compared to the annual amount of emissions.

A previous global model of dioxin noted that depositions were greater than emissions likely due to the photochemical transformation of pentachlorophenol, a common wood preservative, to dioxin (Baker and Hites, 2000). Thus, global emissions were approximately 12.5 times lower than the estimated total deposits of 13,100 kg, and only 5% of emissions were assumed to be deposited to oceans (Brzuzy and Hites, 1996). However, in a preliminary run of our model (that used monthly averaged wind fields), ocean depositions were shown to be significantly larger than previously thought with estimated dioxin deposits to oceans being approximately 38% of the total annual emissions. Therefore, we increased the global total of dioxin production to 17,226 kg. We refined the initial monthly model by using weekly averaged wind fields, and we assumed 1/52 of the annual global dioxin production was emitted each week from land to the atmosphere and used a factor of 60 to convert the production of dioxin to toxic equivalents (TEQs) of the most toxic congener 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (Thomas and Spiro, 1995).

The production of dioxin over land used a global spatialized data set of gross domestic product (GDP; Dilley et al., 2005) as the estimate for countries' dioxin production based on an environmental Kuznets curve (Fig. 2). Within each country, the production of dioxin was made proportional to the GDP of each country's land cells. The atmospheric dispersal of dioxin was modeled using a Geographic Information Systems (GIS)-based model incorporating diffusion of $4.86 \times 10^{-6}\ m^2\ s^{-1}$ (Chiao et al., 1994), wind speed and direction. Wind speed and direction were computed as weekly means from ten years of data (1991–2000) from the European Centre for Medium-Range Weather Forecasts (Anon., 2006) and were interpolated to ½° by ½° resolution from the original 2.5° by 2.5° format. Deposition of dioxin was simulated using a temperature-dependent characteristic travel distance (CTD) approach for 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (Bennett et al., 1998; Klasmeier et al., 2004) that incorporates the grasshopper effect (Gouin et al., 2004).

Once atmospheric dioxin was deposited to land, the movement of dioxin through water basins to marine coastal areas was also incorporated. All land areas were made part of a global network of water basins (n = 6,031) and dioxin was transported from land within water basins to coastal marine waters unless basins were identified as being landlocked and had no ultimate drainage to oceans (n = 166). Spatial data concerning water basins were made available by Prof. C.J. Vörösmarty of the Water Systems Analysis Group (<www.wsag.unh.edu>) at the University of New Hampshire. We based the amount of dioxin transported through water basins on a relationship between water run-off and the fraction of dioxin transported from soil in water basins (Kanematsu et al., 2009; Vasquez et al., 2004).

3. Results

The approach used for the initial distribution of dioxin emissions suggested several areas of likely high local production of dioxin due to higher levels of economic activity (Fig. 3). These were dominated by eastern North America, Europe, South Asia (particularly the Indian subcontinent), and East Asia (China, Japan and South Korea). Countries belonging to the G20 account for over 80% of the estimated annual emissions with Japan, the U.S., and China accounting for 30% of the annual global emissions. However, it is smaller states such as Singapore and Malta that have the highest emissions on a *per* area basis.

The annual global dioxin production of 17,226 kg is equivalent to approximately 287 kg-TEQ. After we ran the model to simulate one year's production, dispersion, deposition and transport of dioxin, approximately 9 kg-TEQ (3%) of the annual dioxin production remained in the atmosphere. The model predicted that most of the annual production of dioxin, 163 kg-TEQ (57%), was deposited to land areas, while ocean waters received approximately 115 kg-TEQ (40%). Large parts of North America, most of central, northern and Eastern Europe, as well as much of the Indian sub-continent

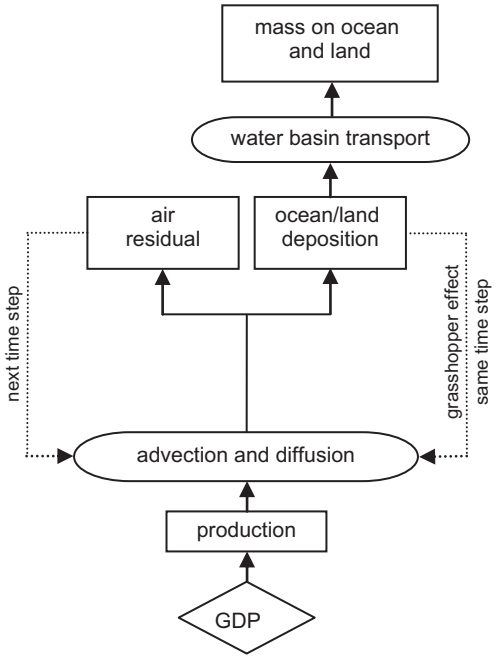

Fig. 1. Flow diagram representing the model simulation of production, atmospheric dispersion, water basin transport and deposition of airborne dioxin to the earth's surface.

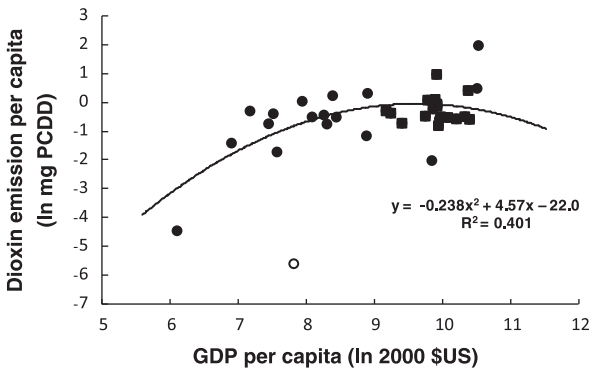

Fig. 2. The environmental Kuznets curve used to estimate countries' per capita dioxin emissions based on GDP per capita. Original data used in Baker and Hites (2000) are shown in square symbols (■), new data are shown in circles (●) with China's data (○) omitted from analysis.