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Changes in persistent organic pollutant levels from adolescence to young adulthood

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ABSTRACT

Elimination rates and their corresponding half-lives are conceptually important and intuitively accessible pharmacokinetic measures of toxicant elimination, but regression-based estimates are biased proportional to the degree of continuing (background) exposure. We propose an alternative estimator, the censored normal regression model, which uses all observations, but treats individuals whose initial level failed to exceed their follow-up level as censored observations to weight the regression estimates from those that declined between blood draws. In this manner, we derive the intrinsic elimination rate, the elimination rate free from ongoing exposure, as a parameter in a regression with an unobserved, latent dependent variable. We utilize sequential measurements of persistent organic pollutants (POPs) levels from adolescence to adulthood, a period of intense change in size and body composition, to quantify individual-level change within a community exposed to significant quantities of contaminants over an extended period of time. Although much research has been conducted on effects of POPs, far less attention has been given to vectors of intake and changes in toxicant levels during the life course. We apply exploratory factor analysis (EFA) to types and timing of consumption, along with physical behavioral characteristics, to identify a structure of seven underlying factors. Although several variables show factorial complexity, the latent constructs included an age/maturation and period-related factor, a nutritional composite, consumption prior to pregnancy, fish and fowl consumed during pregnancy, factors distinguishing body mass and weight from height, and bottom-feeding fish consumption. Unadjusted and adjusted half-lives using the censored normal regression estimator, as well as estimated half-lives from conventional log concentration regressions, are reported for PCB groupings, specific congeners, *p,p′*-DDE, and HCB.

1. Introduction

Persistent organic pollutants (POPs) are resistant to degradation, become widely distributed geographically, biomagnify in food chains, and bioaccumulate in the fatty tissue of non-humans and humans. Adverse effects to health, including the alteration of growth (Burns et al., 2011; Mendez et al., 2011), maturation and development (Denham et al., 2005; Dhooge et al., 2011; Dickerson et al., 2011; Ottinger et al., 2009), and cognitive function (Grandjean and Landrigan, 2006; Newman et al., 2006, 2009; Stewart et al., 2000), have been reported. Polychlorinated biphenyls (PCBs) dichlorodiphenyldichloroethane (*p,p′*-DDE, a metabolite of DDT), and hexachlorobenzene (HCB), are widely recognized as persistent environmental pollutants. Although the use and emissions of these chemicals were severely restricted in the US in the mid-1970s, these toxicants are extremely difficult to destroy by either chemical, thermal, or biochemical processes due to their high thermodynamic stability, and destruction presents the risk of

Abbreviations: AMN, Akwesasne Mohawk Nation; ΣPCB50%, sum of IUPAC#s 52, 70, 74, 84, 87, 95, 99, 101[+90], 105, 110, 118, 138[+163+164], 149[+123], 153, 180, 187; ΣPERPCB8, sum of IUPAC#s 74, 99, 105, 118, 138[+163+164], 153, 180, 187; ΣNONPER6, sum of IUPAC#s 52, 84, 95, 101[+90], 110, and 149[+123]; HCB, Hexachlorobenzene; MAWBs, Mohawk Adolescent Well Being study; *p,p′*-DDE, *p,p′*-dichlorophenyldichloroethylene; PCBs, polychlorinated biphenyls; POPs, persistent organic pollutants; ppb, parts per billion; ppt, parts per trillion

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generating extremely toxic dibenzodioxins and dibenzofurans through partial oxidation (Hansen, 1999; Shibamoto et al., 2007).

Humans continue to be exposed to different mixtures of POPs, especially PCBs, at background levels and via direct routes of intake (ingestion, inhalation, intrauterine transmission, etc.) (Grandjean et al., 2008; Knobeloch et al., 2009; Lignell et al., 2009; Matsumoto et al., 2009; World Health Organization, 2003). Exposure beyond background levels continues primarily through the dietary intake of contaminated animal, fish, or fowl fats. Bottom feeding and long-lived predatory fish can accumulate high levels in their body fat and these residues can be passed on to humans and wildlife that consume them (Startin and Rose, 2003). Exposures to PCBs, *p,p′*-DDE, and HCB have been linked to diabetes (Lee et al., 2010; Turyk et al., 2009), high blood pressure/hypertension (Goncharov et al., 2008; Langer, 2010), thyroid hormone disruption (Chevrier et al., 2007; Langer et al., 2009; Schell et al., 2009; Schell and Gallo, 2010), effects on sexual maturation and differentiation (Denham et al., 2005; Dhooge et al., 2011; Dickerson et al., 2011; Ottinger et al., 2009), and reproduction (Cohn et al., 2010, 2011; Needham et al., 2011; Wigle et al., 2008). These endocrine disruption effects of POPs continue to be of concern, especially during critical times of growth and development such as adolescence and young adulthood, primarily due to their long half-lives (Stehr-Green, 1989) and potentially adverse long-term health consequences.

Although much research has been conducted on health effects of POPs, far less attention has been given to vectors of intake and changes in toxicant levels during the life course. To the best of our knowledge, no other study has utilized sequential measurements of POP levels from adolescence to adulthood, a period of intense change in size and body composition to quantify individual-level change within a community exposed to significant quantities of contaminants over an extended period of time.

We begin by reporting serum levels of persistent organochlorines (PCBs, *p,p′*-DDE, HCB) within Akwesasne Mohawk adolescents (collected between 1995 and 2000) and again in young adulthood (collected between 2000 and 2005), with an average interval of four years between measurements. The Mohawk of Akwesasne have had significant changes in lifestyle following the discovery of contamination of the St. Lawrence River on which they depended for fish, hence reducing their reliance on the traditional fish-based diet to a less healthy one containing more fat and calories (Ravenscroft and Schell, 2014). However, a substantial proportion of our sample evidence an absolute increase in POP levels at follow-up (young adulthood) over their baseline measure (adolescence).

Elimination half-life, the amount of time required to reduce a toxicant to one-half its level at initial measurement, is a conceptually important, and intuitively accessible, pharmacokinetic measure of toxicant elimination, but its conceptual utility is predicated on the assumption that differences in rates of elimination from the body depend primarily on compound-related pharmacokinetics and host-related factors that affect individuals' metabolism rates (Lotti, 2003; Matthews and Dedrick, 1984). Continuing exposure between initial and follow-up measurement violates this assumption and confounds estimation of elimination kinetics. Most of the literature on elimination half-lives is now dismissed as having reported “apparent” elimination half-lives (Shirai and Kissel, 1996; Milbrath et al., 2009; see discussion of apparent versus intrinsic half-lives in Ritter et al., 2011). The concern over bias introduced by ongoing exposure is so severe that it has led some to conclude that it is impossible to offer reliable half-life figures from longitudinal data with ongoing environmental exposure (Lotti, 2003; Yakushiji et al., 1984). At a minimum, the concern has produced a preference for the development of methods for half-life estimation from cross-sectional data (Ritter et al., 2011; Wong et al., 2013) or regression of log serum concentrations on time after primary exposure ceases for longitudinal data (cf. Bartell, 2012). Neither of these approaches is likely to have strong causal inference after extrapolation to the real world (Manski, 1995) or adequately respond to bias introduced by ongoing exposure and host-related factors on elimination (Milbrath et al., 2009).

Accordingly, this analysis has two objectives. First, we determine the factor structure underlying fifteen items representing dietary intake, measures of body burden/storage reservoirs, and other known correlates of toxicant exposure. Second, we estimate intrinsic elimination rates using an exponential decay model and a regression method ideally suited to a population under continuing exposure, with and without adjustment for within-individual effects of the (seven) identified constructs and other host-related covariates. We report the corresponding intrinsic elimination half-lives for PCBs, *p,p′*-DDE, and HCB.

2. Materials and methods

2.1. Sample and site characteristics

The Akwesasne Mohawk Nation (AMN) is a sovereign nation situated on the St. Lawrence River with territory bordering New York State, Ontario and Quebec, Canada. The Akwesasne community is one of several communities comprising the Kahniakehaka/Mohawk nation, and is traditionally known as the keeper of the Eastern Door of the Iroquois Confederacy (the Haudenosaunee Confederacy) with a population approximating 12,000–13,000 people (Akwesasne Task Force on the Environment, 1997; Fitzgerald et al., 1998; George-Kanentiio, 1995).

Industrial development along the St. Lawrence River began in the 1950s, and major industrial facilities located around Cornwall, Ontario, and Massena, New York, discharged significant quantities of contaminants, including PCBs, *p,p′*-DDE, HCB and mirex, into the St. Lawrence River and its three tributaries (Sloan and Jock, 1990). Contamination of the local waters entered the local food chain, and some local species of fish, birds, amphibians and mammals were found to have levels exceeding the US Food and Drug administration's tolerance limits for human consumption (Forti et al., 1995; Sloan and Jock, 1990), leading to the issuance of fish and game advisories in the late 1980s and early 1990s (Fitzgerald et al., 1995, 1998).

2.2. Data collection

The University at Albany, State University of New York's Institutional Review Board approved all study protocols and informed consent procedures. Additionally, assent from minors was obtained from all participants. For both projects, all data collection was performed by project staff, all members of the Akwesasne community, and data were collected without prior knowledge of participants' exposure status. Study protocols and methods have previously been described in detail (Gallo et al., 2005, 2007, 2011; Newman et al., 2006, 2009; Schell et al., 2003, 2008, 2009), and are briefly reviewed here.

Data were collected for the Mohawk Adolescent Well-Being Study (MAWBs) between 1995 and 2000. In brief, 294 mother/youth pairs were recruited, and due to attrition resulted in a final sample size of 271 participants (131 males and 140 females) between 10 and 16.99 years of age (for more detail on recruitment and sampling see Schell et al., 2003, 2008; Gallo et al., 2005, 2007).

In MAWBs, the youth's mother completed interviews and questionnaires to obtain information about the youth's family background including sociodemographic status and sources of