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Exposure and risk assessment of the Czech population to chlorinated pesticides and polychlorinated biphenyls using archived serum samples from the period 1970 to 1990

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ABSTRACT

The serum samples from the years 1970–1990 archived at the temperature of −20 °C in the biobank primarily intended for serological survey performed in the CR since 1960 were pooled and analyzed for DDT, its metabolites, HCB, HCHs, and indicator PCB congeners using up-to-date GC/MS/MS methods to retrospectively assess health risks according to current health guidelines. Samples were pooled based on the decade of sampling, age, gender, and three geographical areas; in adults, one pooled samples consisted of ten and in children of twenty individual samples. Altogether 233 pooled samples were analyzed. For all organochlorine pesticides (OCPs), significant downward trends were observed in the period 1970–1990 ($p < 0.001$). The levels of HCB exceeded the Biomonitoring Equivalent (BE) value. The hazard quotient (HQ) in Prague and Ostrava during the 1970s and 1980s was about 40 and in the 1990s it had dropped to about five. In Uherské Hradiště, the HQ in 1975 was one order of magnitude higher (about 170), and had decreased to approximately 12 by 1987. For both HCB and the DDT sum, the BE-related carcinogenic risk of actual concentrations in the past exceeded significantly the individually accepted cancer risk level of 10^{-4}. The levels of the main PCB congeners in the 1970s through 1990s revealed an upward time trend in all analyzed strata. The highest concentrations were found in the serum of residents from the hot-spot area Uherské Hradiště. Critical PCB sum concentration levels (700 ng/g lipid for vulnerable population groups and 1800 ng/g lipid for other population groups) were substantially exceeded with an increasing time trend. PCB sum had exceeded HBM II values of 7 μg/L of serum since 1980 in all age strata. In conclusion, the body burden of the Czech general population relative to persistent organic pollutants (POPs) in the period 1970 through 1990 significantly exceeded currently existing health based limit values. The past exposure might adversely affect the health status of the Czech population.

1. Introduction

Persistent organic pollutants (POPs) are a diverse group of synthetic chemicals used in the past for various industrial, agricultural, and commercial applications. These compounds persist in the environment for long periods of time. They are lipophilic and bio-accumulate within the food chain into animal and human fatty tissues (Carpenter, 2011; Safe, 1994). The best-known representatives of the POP family are organochlorine pesticides (OCPs), and polychlorinated biphenyls (PCBs) (Rosner and Markowitz, 2013). Organochlorine insecticides, mainly DDT, were widely used in agriculture and in the control and prevention of the spread of infectious disease vectors (WHO, 1979) up until the end of 60s. In the former Czechoslovakia, the production and usage of DDT was banned in 1974; however, it continued to be used, to a limited extent, together with γ-hexachlorohexane (γ-HCH) isomere (lindan) in products for killing hair lice. Hexachlorobenzene (HCB) was widely used as fungicide for several decades in agriculture. HCB was produced in the former Czechoslovakia until 1968, but this compound can also originate as a by-product during the production of other industrial chemicals, e.g. during the electrolytic production of chlorine (ATSDR, 2002; Jacoff et al., 1986).

PCB production started in the eastern part of former Czechoslovakia, in 1959, as technical mixtures under the trade name DELOR (Kocan et al., 2001). Unlike other PCB-producing countries, where the production was prohibited in early 70s, the production in the former Czechoslovakia actually increased in late 70s and peaked around 1980. At the time there were few regulatory controls in

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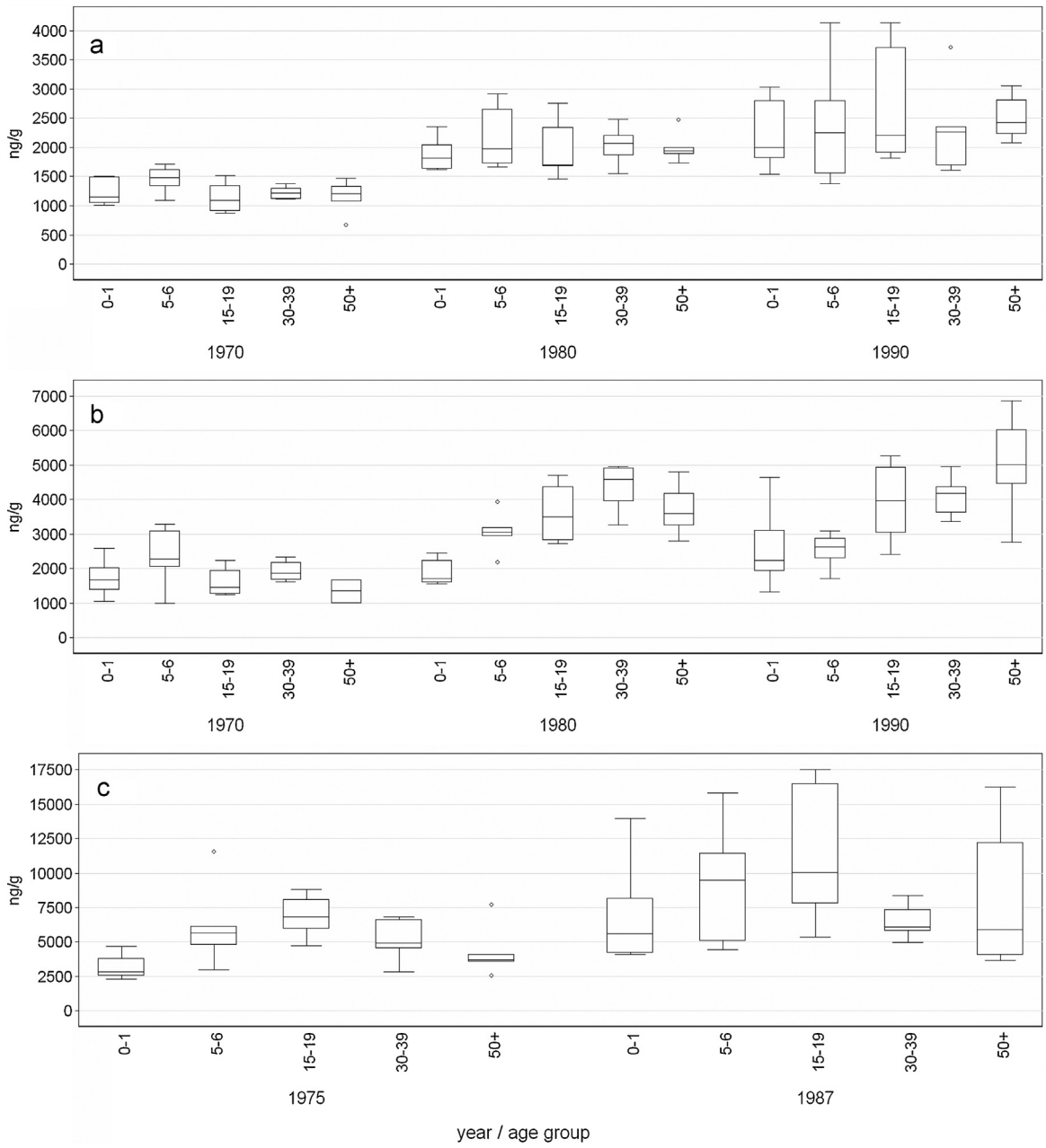

Fig. 1. Box-and-whisker plots of the PCB sum (ng/g lipid) calculated as sum of (138, 153 and 180) multiplied by factor 1.7 in individual age groups: (a) Prague; (b) Ostrava; (c) Uherské Hradiště.

terms of risks to health and environment. The production of PCBs, in the former Czechoslovakia, was banned in December 1984. About 11,613 t were used inside the former Czechoslovakia, as components of many products including paints, which were produced in a former Czechoslovak paint factory in Uherské Hradiště (Jursa et al., 2006).

It is known that human exposure to POPs has been associated with various adverse health effects such as immunotoxicity, endocrine disruption, reproductive effects, neurotoxicity, and carcinogenicity (ATSDR, 2002). The International Agency for Research on Cancer (IARC) long ago classified DDT and HCB as probable and possible human carcinogens (class 2A and 2B, respectively) (IARC, 2015). Recently, PCBs as the whole group were classified as carcinogenic for humans (group 1) (Lauby-Secretan et al., 2013). Because of the long half-life of these compounds, present-day human health effects, as a result of exposure that occurred years before, can still manifest (Ritter et al., 2011). Therefore the knowledge of population exposure and body burden in the past is extremely important also for present-day health risk assessments.

Human exposure to POPs is conveniently assessed using body fluids or tissues containing lipids. Most often, human milk or blood serum/plasma is used for this purpose. Participation of the Czech Republic in the second round of the WHO-coordinated study in WHO-coordinated study in 1992 demonstrated the existence of hot-spot, where the levels of PCB congeners in pooled human milk were much higher than in other participating countries (WHO/ECEH, 1996). This finding initiated systematic biomonitoring of POPs and other environmental contaminants in body fluids of the Czech general population. The levels of indicator PCB congeners and selected OCPs in human milk started to be continuously monitored in 1994 under the auspices of the Czech Environmental Monitoring System (Černá et al., 2003). Since 2001, selected POPs

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