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Intake of DDT and its metabolites through food items among reproductive age women in Bangladesh

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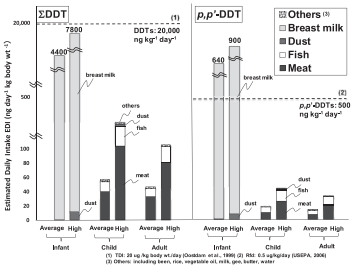
HIGHLIGHTS

- Made clear the major intake route of DDT in Bangladesh people through food.
- Highest levels of DDT were determined in beef and human breast milk.
- EDI on *p,p'*-DDT found in infant was exceeded reference dose (RfD).
- In case of child in Bangladesh, DDT intake through beef consumption occupied 72% of it.

GRAPHICAL ABSTRACT



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ABSTRACT

This study was conducted to make clear the major intake route of DDT in Bangladesh people to develop strategy and policy that could lead to a reduction in body burden especially in the reproductive age women. The concentrations of several POPs (DDT, PCBs, chlordanes, HCHs, HCB, and PeCB) were quantified in food items, human breast milk and house dust collected in Bangladesh in 2011–2012. Among the POPs analyzed in this study, DDT and its metabolites (ΣDDT) showed the highest concentration. The highest median ΣDDT concentration was found in meat (1.3–1100 ng g^{-1} wet weight) and house dust (30–1100 ng g^{-1} dry weight), and followed by human breast milk (20–55 ng g^{-1} wet weight). Estimated daily intake (EDI) was calculated using the DDT concentrations in food items. The highest intake of DDT was found in an infant and 99% of it was via breast milk feeding. DDT intake via consumption of beef accounted for 69% and 72% of that found in children and adults, respectively. The total EDI of DDT did not exceed the tolerable daily intake proposed by the WHO, but the EDI of *p,p'*-DDT exceeded the oral reference dose proposed by the US Environmental Protection Agency. Further research is required to clarify the reason for the high levels of DDT in beef, which seems to be the major intake route of DDT for women of reproductive age in Bangladesh.

1. Introduction

Persistent organic pollutants (POPs) are a group of chemicals whose specific physicochemical properties such as their structures,

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persistence, long half-lives, high lipophilicity, long-range atmospheric transport, and potential toxicity, cause them to bioaccumulate in organisms through the food chain and have adverse impacts on humans and wildlife (AMAP, 2002; IOMC, 2011). DDT is a POP that has been used extensively for agriculture as a pesticide and for malaria prevention worldwide since the 1940s. In Bangladesh, agriculture is the main occupation of the nation, employing 66% of the labor force. The use of DDT as a pesticide in agriculture to increase crop production dates back to the mid-1950s (Rahman, 2003). In Bangladesh, DDT products have been mostly used chemical for public health, particularly for mosquito eradication which was supplied by the World Health Organization (WHO) started in 1960 (ESDO, 2005). In the mosquito control program, the formal use of DDT products as indoor residual spraying (IRS) started in 1965, when every house was sprayed twice a year at a dose of 2 g/m^{-2}. Later, when the mosquito population was significantly reduced, it was applied at 1 g/m^{-2} as a "focal spray" where a significant population of mosquitoes was observed. Gradually the focal spray was reduced and it was used only in areas where there were indigenous cases of malaria. The use of DDT product was banned for agricultural purposes in the early 1980s in Bangladesh. In around 1992/93, all usage of DDT products were banned in every sector and it is currently used only if there is a serious outbreak in some focal areas (ESDO, 2005).

Even after the use of DDT products were discontinued, high concentrations of it were found in food items such as fish (Jabber, 2001; Bhuiyan et al., 2009; Chowdhury et al., 2010; Zamir et al., 2013; Hossain et al., 2016). This not only reflected high levels of DDT in the environment but also suggested DDT spraying of dry fish to protect it from insects (Jabber, 2001; Bhuiyan et al., 2009; Siddique and Aktar, 2012). High DDT concentrations in food items can cause DDT contamination of the human body. Actually, high DDT concentrations were detected in the plasma of people in Bangladesh, which suggested DDT intakes through food items such as dry fish (Zamir et al, 2008). High concentrations of DDT were also detected in human breast milk samples collected in rural villages in Bangladesh in 2002 (Bergkvist et al., 2012). That report suggested that the reason for this was that samples were collected in rural agricultural villages where they used large amounts of pesticides, including DDT, for agriculture purposes (Rahman, 2003). Another reason might be that some of the countries surrounding Bangladesh have shown 10 times higher emission levels of DDT as a consequence of the production and use of organochlorines (AMAP, 2002).

The high levels of DDT found in human blood and breast milk in the general population of Bangladesh suggest that there are high levels of this chemical in women of reproductive age. High levels of DDT in such women would affect fetuses in utero and infants through breast milk. It is known that fetuses and infants are very sensitive to environmental contaminants such as DDT, and in utero exposure to DDT is linked to an increased risk of breast cancer later in life (Cohn et al., 2003). The DDT burden in reproductive women should be reduced to lower the prenatal and postnatal exposures of fetuses and infants. The major intake route of DDT in humans in Bangladesh should be clarified so that counteractions can be taken to reduce its intake. However, there is no information available regarding the intake routes of DDT in Bangladesh. This study was conducted to make clear the major intake route of DDT in Bangladesh people to develop strategy and policy that could lead to a reduction in body burden especially in the reproductive age women.

The objectives of this study were to: 1. quantify the DDT concentrations in various food items and house dust samples, 2. calculate the estimated daily intake (EDI) and evaluate the risks of DDT intake, and 3. propose ways to reduce exposure to DDT, especially for women of reproductive age in Bangladesh.

2. Materials and methods

2.1. Sample collection of food items, breast milk and house dust

The collected food items, human breast milk, and house dust are summarized in Table 1 (detailed information is summarized in Table S1). Collected foods which was the major items in Bangladesh were chosen based on the FAO Food Balance Sheet on Bangladesh (Milton et al., 2006; FAO-STAT, 2009). Food samples were collected from the market in Dhaka and Chittagong in Bangladesh from 2011 to 2012. The food items were divided into 7 food groups. Fish were selected from popular species among Bangladeshi peoples from three different sources (fresh, brackish, and sea water). The meat was comprised of beef, chicken and mutton. It has been reported that almost 60% of beef is imported from India (Khatun et al., 2016), but it was not possible to identify the source of meat samples when those were purchased in the market. The dairy products studied were milk, butter made from milk, and a type of local butter called ghee. The cereals chosen were rice and beans, which are commonly eaten in Bangladesh. Oil consisted of vegetable oil made from soybeans. Drinking water consisted of tap water and that collected from tube wells. The house dust samples were collected from urban type house of apartment houses and hotel located in Dhaka, and rural type of houses in Narayangonj (rural village near Dhaka). Surface fine dusts were collected from top of furniture by using pre-cleaned Kimwipe and stored in a plastic bag. The human breast milk samples were collected from the primipara mothers who were from 18 to 22 years old and had lived in Dhaka more than 10 years. All babies were exclusively breast feeding and aged up to six months since birth. The samples were homogenized and stored at −20 °C until chemical analysis.

2.2. Chemical analysis and quality control

DDT and its metabolites (ΣDDT: sum of *p,p'*-DDT, *p,p'*-DDD, and *p,p'*-DDE), and other POP compounds, polychlorinated biphenyls (PCBs: sum of major peaks of tri-to deca-isomers), chlordane and related compounds (CHLs: sum of *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, *cis*-nonachlor, and oxychlordane), isomers of hexachlorocyclohexane (HCHs: sum of α-hexachlorocyclohexane, β-hexachlorocyclohexane, and γ-hexachlorocyclohexane), hexachlorobenzene (HxCB), and pentachlorobenzene (PeCB) were analyzed following the methods described previously (Kajiwara et al., 2003) with slight modifications.

Briefly, a portion of a food sample (5 g of breast milk or food homogenate) was mixed with surrogate standards comprised of a $^{13}C_{12}$-PCB mix (from mono-to deca-CB) and $^{13}C_6$-HxCB, and ground with sodium sulfate. It was extracted using a Soxhlet apparatus, and the extract was concentrated using a rotary evaporator. An aliquot of the extract was used for determination of crude fat by measuring the total non-volatile extract. Another aliquot of it was subjected to gel permeation chromatography (GPC) for lipid removal. The GPC fraction containing target compounds was concentrated and passed through an activated florisil-packed glass column for further cleanup. Elution was micro concentrated after adding internal standards comprised of a $^{13}C_{12}$-PCB-105 as performance standard and employed for quantification by GC-MS.

Drinking water sample (6 L) was passed through a glass fiber disk (Whatman GMF 150), and extract using solid phase extraction (SPE) cartridge (Aqusis PLS-3200 mg, Waters). SPE cartridge was dried by nitrogen gas flow for 1 h, then eluted by acetone and dichloromethane. Elution was dried by sodium sulfate after adding hexane and micro concentrated after adding internal standards