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Intake estimates of phthalate esters for South Delhi population based on exposure media assessment

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ABSTRACT

An indirect estimation method was followed to derive exposure levels of fifteen phthalate congeners in urban population of Delhi, India. The exposure media samples were collected from Jawaharlal Nehru University (JNU) campus and Okhla industrial area. GC–MS analysis of the samples indicated di(2-ethylhexyl) phthalate (DEHP) to be the most abundant congener and its estimated total daily intake level reached upto 70 μg kg^{-1} d^{-1}. Out of the studied congeners, intake doses for di-n-butyl phthalate (DnBP) and DEHP, reached levels near or above the established exposure limit. In JNU, DEHP, dimethyl phthalate (DMP) and butyl benzyl phthalate (BBP) had 69% share in combined daily intake of Σ_{15} phthalates (CDI_{15}); whereas, in Okhla, DEHP, diethyl phthalate (DEP), diisobutyl phthalate (DIBP), DnBP and DMP shared 64% of the CDI_{15}. Food was found to be the major source of exposure contributing 67% and 74% of the estimated CDI_{15} at JNU and Okhla respectively.

1. Introduction

Phthalates are broadly used as plasticizers in vast range of industrial applications and ultimately they appear in wide spectrum of consumer products. They get their way into the environment either by losses during manufacturing processes or by weathering, leaching or evaporating from and of the products they are used in Wittassek et al. (2011). Thus, phthalates have become a ubiquitous environmental contaminant and as a result, general population is continuously exposed to them. Some human exposure studies during last decade have shown effects on reproduction, DNA damage to sperm (Pant et al., 2010; Rozati et al., 2002), early onset of puberty in females (Wolff et al., 2010), anomalies of reproductive tract (Desdoits-Lethimonier et al., 2012), infertility (Rozati et al., 2002; Tranfo et al., 2012) and adverse outcomes of phthalate exposure during pregnancy including fetal development (Latini et al., 2006; Whyatt et al., 2009). Although far less investigated, there are reports suggesting that phthalates may adversely affect other functions and systems such as thyroid signalling (Meeker et al., 2007), neuro-development (Miodovnik et al., 2011) asthma and allergies (Jaakkola and Knight, 2008), and insulin resistance (Stahlhut et al., 2007). Due to their possible harmful effects, monitoring of phthalate in the environment and human exposure assessment studies have gained momentum during last decade (Clark et al., 2011; Guo and Kannan, 2011; Wittassek et al., 2007; Wormuth et al., 2006).

Humans get exposed to phthalates via multiple exposure pathways, including inhalation, dietary intake and dermal absorption (Clark et al., 2011). Extensive human exposure assessment data are available from Europe (Wittassek et al., 2007; Wormuth et al., 2006), the United States (Blount et al., 2000; Colacino et al., 2010; Kohn et al., 2000), Japan (Itoh et al., 2007), China (Guo and Kannan, 2011; Pei et al., 2013; Wang et al., 2013) and other developed countries. In general, these earlier studies have indicated that the sources and amount of human exposure varies, depending on the country specific consumption pattern of different phthalates. However, in Indian scenario, human exposure assessment data for phthalates still remains scarce. A recent study by Guo et al. (2011) has shown presence of 389 ng mL^{-1} of total phthalate metabolite in urine samples collected from the general populations of a small south Indian town Mettupalayam. Guo et al. (2011) highlighted the concentration to be second highest next to Kuwait, among seven Asian countries including China, India, Japan, Korea, Kuwait, Malaysia, and Vietnam. The human exposure scenario may be found more alarming, if studied, in metro cities of India like Delhi where pollution levels are more critical.

Phthalates are rapidly metabolised into their respective monoesters in human body. The primary monoester then may be further metabolized before getting excreted with urine and faeces (Koch

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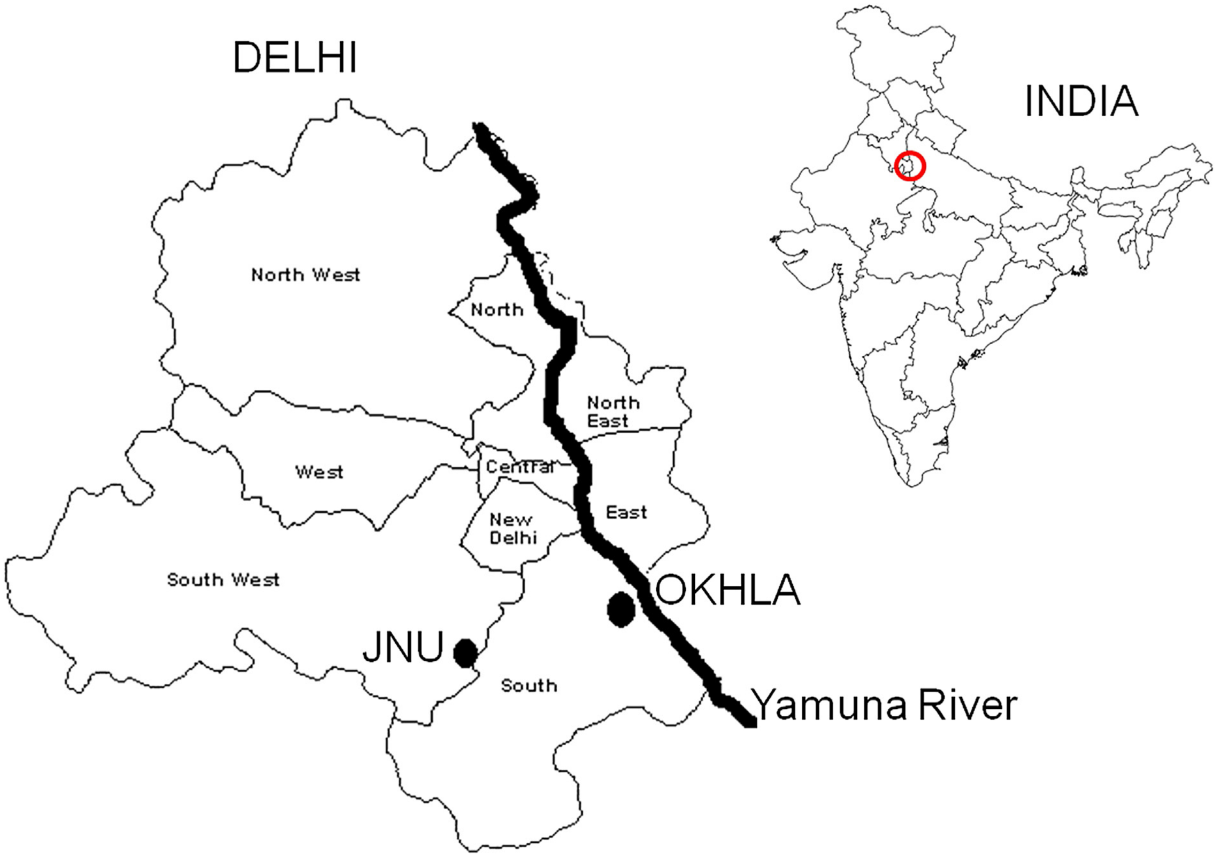

Fig. 1. Sampling sites. Map of Delhi showing sampling location.

et al., 2005). Several studies have monitored urinary phthalate metabolites as biomarkers of human exposure to the mother compounds (Barr et al., 2003; Guo et al., 2011; Latini, 2005). However, urinary metabolite based methods do not provide any information about source(s) and pathways of human exposure. Moreover, they are susceptible to physiological variability among different age, gender and geographical location specific population (Clark et al., 2011). Thus, exposure estimations based on urinary metabolite alone, often underestimate the population's actual exposure to phthalates (Barr et al., 2003; Lyche et al., 2009). Therefore, urinary bio-monitoring studies need to be complimented with exposure source and pathway monitoring data (Wormuth et al., 2006).

Comparatively, fewer studies have so far, estimated human exposure to phthalates by source and pathway monitoring data (Clark et al., 2003; Müller et al., 2003; Pei et al., 2013; Wormuth et al., 2006; Xu et al., 2010). This approach known as "indirect estimation method" uses information about the concentration of phthalates in a particular exposure media (e.g. air, water, food, etc.) and integrates the rate of intake of that media to calculate total daily intake. Müller et al. (2003) have estimated Danish population's exposure to five phthalate members (DEHP, DBP, DINP, DIDP and BBP) from food, water, air and other consumer products by using available concentration data with simulating the exposure situations. Similar simulation based indirect approach has also been used to estimate Canadian population's exposure to phthalate esters (Clark et al., 2003). These indirect estimation studies further help in implementing regulatory actions on production, distribution and disposal limits of phthalates.

Present investigation was designed to estimate an adult individual's daily integrated exposure to phthalate esters in urban South Delhi region. As no prior significant concentration data were available for phthalates in India, initially monitoring was carried out for fifteen different phthalate members in different exposure media after which an indirect estimation method was followed to derive exposure values for each of the fifteen phthalates.

2. Materials and method

2.1. Reagents and precautions

All solvents used in the present work were of HPLC grade and were supplied by Merck (Darmstadt, Germany). The phthalate esters mixture (M-8061 R) used for quantitative analysis were obtained from AccuStandard Inc. (New Haven, USA). All other chemicals were purchased from Sigma–Aldrich (St. Louis, MO, USA) unless stated otherwise.

Analysis of phthalates in samples are often encountered with high blanks problems as many laboratory products, including chemicals and glassware often get contaminated with phthalates due to the latter's ubiquitous environmental presence (David et al., 2003). To avoid phthalates contamination, all glassware used in the study was soaked in acetone for at least 30 min, then washed with acetone, rinsed with hexane, and dried at 120 °C for overnight. Neutral Alumina (pH 6–8) used for cleanup purpose, was heated to 400 °C for 16 h, cooled in a desiccator and activated with 3% of Milli-Q water prior to use.

2.2. Sampling time and site description

Exposure media samples such as food, drinking water, indoor air, indoor dust, outdoor air and outdoor dust were collected from two different locations of South Delhi with varying human activity pattern such as traffic density, industrial and commercial activities etc. (Fig. 1). Sampling was carried out at both the sites simultaneously during 14th–21st March 2011. Further details about the sampling sites were provided in the Supplementary Tables S1 and S2.

The first sampling site was Jawaharlal Nehru university (JNU) campus area with mostly educational and residential buildings, with no industries nearby (3–4 km radius), having very low vehicular traffic and with good vegetation cover. The second sampling site was Okhla, an industrial cum residential area with large number of industries mostly of paints, varnishes, plastic, rubber, cosmetics, chemicals etc. The traffic density was quite high and most significantly, a waste dumping site in the