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Current status of persistent organic pesticides residues in air, water, and soil, and their possible effect on neighboring countries: A comprehensive review of India

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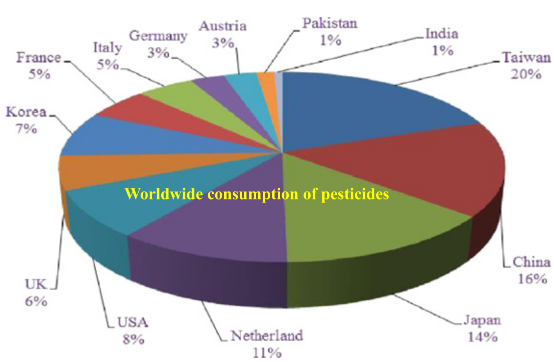
HIGHLIGHTS

- Every environmental component in India is contaminated with POPs.
- Residue level of pesticide is high in air, water and soil despite low consumption.
- Tibetan Plateau in Southwest China is affected with POP emission from India.
- India is one of the major contributors of global POP distribution.

GRAPHICAL ABSTRACT



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ABSTRACT

Though the use of pesticides has offered significant economic benefits by enhancing the production and yield of food and fibers and the prevention of vector-borne diseases, evidence suggests that their use has adversely affected the health of human populations and the environment. Pesticides have been widely distributed and their traces can be detected in all areas of the environment (air, water and soil). Despite the ban of DDT and HCH in India, they are still in use, both in domestic and agricultural settings. In this comprehensive review, we discuss the production and consumption of persistent organic pesticides, their maximum residual limit (MRL) and the presence of persistent organic pesticides in multicomponent environmental samples (air, water and soil) from India. In order to highlight the global distribution of persistent organic pesticides and their impact on neighboring countries and regions, the role of persistent organic pesticides in Indian region is reviewed. Based on a review of research papers and modeling simulations, it can be concluded that India is one of the major contributors of global persistent organic pesticide distribution. This review also considers the health impacts of persistent organic pesticides, the regulatory measures for persistent organic pesticides, and the status of India's commitment towards the elimination of persistent organic pesticides.

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Contents

1. Introduction

The use of chemical pesticides has provided a valuable aid to agricultural production, increasing crop protection and yield. However, the discovery of pesticidal residues in various sections of the environment has raised serious concerns regarding their use; concerns which well-outweigh the overall benefits derived from them (Ali et al., 2014; Sharma et al., 2014). Organochlorine pesticides (OCPs) have been in wide usage across the world to control agricultural pests and vector-borne diseases (Abhilash and Singh, 2009; Zhang et al., 2011). Amongst the OCPs in regular usage, dichlorodiphenyltrichloroethane (DDT), hexachlorocyclohexane (HCH), endosulfan, aldrin, chlordane, dieldrin, endrin, heptachlor, mirex, hexachlorobenzene (HCB) and toxaphene, metoxychlor, and metolachlor are persistent organic pesticides. OCPs are very stable compounds and their half-lives can range from a few months to several years; in some cases decades (Cremlyn, 1991). It has been estimated that the degradation of DDT in soil ranges from 4 to 30 years, while other chlorinated OCPs may remain stable for many years after their use (Afful et al., 2010). Because of their inability to break down in the environment, their degradation is restricted by chemical, physical, biological and microbiological means (Afful et al., 2010; Darko and Acquaah, 2007; NCEH, 2005; Swackhamer and Hites, 1988). They are liposoluble compounds and are capable of bio-accumulating in the fatty parts of biota such as breast milk, blood and fatty tissues (William et al., 2008) in the food chain. As a result, human beings are exposed to the effects of these micropollutants by eating foods in contact with contaminated soil or water (Belta et al., 2006; Raposo and Re-Poppi, 2007). These pesticides not only cause serious diseases in humans but are also highly toxic to most aquatic life (Aiyesanmi and Idowu, 2012) and soil microflora (Megharaj, 2002).

According to a joint report produced by WHO and UNEP, roughly 200,000 people die and around three million are poisoned each year by pesticides, all over the world, though the vast majority (95%) of cases are from developing nations (WHO/UNEP, 1990; FAO/WHO, 2000; Pope et al, 1994). As a result of the severe health effects associated with pesticides, most notably DDT and HCH, their use has been either banned or restricted in many countries (Jit et al., 2011; van den Berg, 2009). In India, pesticide use was banned in 1985, with the exemption of DDT which is still being used to control malaria (UNEP, 2003). Though the use of HCHs was banned for agricultural use in 1997, the Indian government still allows the use of HCHs on specific crops as well as in the health sector (Mukherjee and Gopal, 2003; Tanabe et al., 1994). There are few countries besides India who are still engaged in the production, usage and export of γ-HCH on a large scale (Abhilash and Singh, 2009).

India is home to approximately 16% of the total world's population, but has just less than 2% of the total landmass. Rapid population growth, together with a high emphasis on achieving food grain self-sufficiency has compelled Indian farmers to resort to the substantial use of pesticides. It is estimated that more than 100,000 tons of DDTs has been applied in India alone, primarily for agricultural use and malaria eradication programs, due to their low cost and broad-spectrum toxicity, making them effective in the control of pests and diseases (Kannan et al., 1995; Voldner and Li, 1995; Abhilash and Singh, 2009; Arora et al., 2013). However, some characteristics, such as persistency, volatility and atmospheric distribution through long range transportation (Bentzen et al., 2008; Caldas et al., 1999) has resulted in the contamination of air, water, soil and food (Caldas et al., 1999; Hans et al., 1999; Kim and Smith, 2001; Kumar et al., 1995; Singh, 2002; K.P. Singh et al., 2005; V.K. Singh et al., 2005).

Persistent organic pesticide residues have been broadly distributed in Indian soil (Al-Wabel et al., 2011; Devi et al., 2013; Hoai et al., 2010; Kata et al., in press; Kumar et al., 2014; Kumari et al., 2008; Liu et al., 2009; Senthilkumar et al., 2009), water (Huang et al., 2013; Lari et al., 2014; Malik et al., 2009; Mutiyar et al., 2011; Mutiyar and Mittal, 2012; Singh et al., 2012), air (Chakraborty et al., 2010; Devi et al., 2011; Huang et al., 2013; Srimural et al., in press; Syed et al., 2013; Zhang et al., 2008), living creatures (Bhuvaneshwari and Rajendran, 2012; Devanathan et al., 2009; Subramaniam and Solomon, 2006), and crops (Bajpai et al., 2007; Chowdhury et al., 2011). The fate of OCPs in soils in the areas of land use and cropping patterns has also been extensively studied. The concentrations of these pollutants observed in many agricultural soil samples were very high (Al-Wabel et al., 2011; Hoai et al., 2010; Kumari et al., 2008; Liu et al., 2009; Senthilkumar et al., 2009; Xu et al., 2013a,b). A number of studies revealed the prevalence of OCPs and PCBs in various environmental matrices, including food commodities in India (Aktar et al., 2009; Chakraborty et al., 2010; Chowdhury et al., 2007; Devanathan et al., 2009; Guzzella et al., 2005; IPEN, 2006; Kole et al., 2001; Kumar et al., 2009; Prakash et al., 2004; Senthilkumar et al., 2001; Someya et al., 2009; Zhang et al., 2008). Today, everything from food to groundwater and drinking water is contaminated severely with OCPs, therefore understanding the environmental fate of OCP residues is a key issue. Although OCPs have a long history (over 30 years) of usage in India, very limited information is available on the presence of OCP residues in the air, soil and water systems. Also, study of the health risks associated with OCPs in the Indian environment is lacking (Ali et al., 2014; Sharma et al., 2014). Despite a low consumption pattern of OCPs in India compared to other developed nations, its indiscriminate use has resulted in the sporadic occurrence of the residues in abiotic and biotic compartments (Sarkar et al., 2008). It is worth examining the distribution, behavior and fate of these compounds in various areas of the environment in order to understand the potential sources of persistent organic pesticides. Discovery of these compounds in multiple sections of the environment (water, air and soil) may indicate the extent of contamination and the accumulation in the region.

Here, we review the present levels of persistent organic pesticides in different environmental areas (air, water and soil) in India to assess their possible impact on human health and the neighboring countries/regions; potential hazards and risks associated with saturation; best management practice; and the regulatory system to control persistent organic pesticides.

1.1. Consumption of pesticides in India

Approximately 2 million tons of pesticides is consumed worldwide each year, of which 24% is consumed in the USA, 45% in Europe and the remaining 25% in the rest of the world (Abhilash and Singh, 2009). This indicates that the most developed countries (mostly North America, Western Europe, and Japan, where pesticide application rates are high) consume three quarters of the total pesticide used worldwide (USEPA, 2009). The use of herbicides, which have a lower acute, or immediate, toxicity than insecticides, in these regions is more commonplace (WRI, 1998). In most developing nations, India included, the situation is reversed and insecticide use predominates, posing higher levels of acute risk (Rathore and Nollet, 2012). Though the quantity of pesticides consumed in developing nations is much less compared to developed countries, it is growing gradually and substantially (Wilson and Tisdell, 2001; WRI, 1998). Consumption of pesticides is intense, especially in export crops such as cotton, bananas, coffee, vegetables, and flowers (WRI, 1998).

In India, the use of pesticides began in 1948 with the import of DDT for malaria control and benzene hexachloride (BHC) for locust control (Gupta, 2004). Later, in 1949, the use of both pesticides (DDT and BHC) was broadened to the agriculture sector. At present, India accounts for approximately 3% of total pesticide consumption in the world (Fig. S.1), however this is increasing at the rate of 2–5% per annum (Bhadbhade et al., 2002). Of the total Indian pesticide consumption, 67% is used in agriculture and horticulture (Puri, 1998), while public health accounts for roughly 8.5% (World Bank, 1997). Pesticide consumption has increased several hundred folds, from 154 MT in 1954 to 41,822 MT in 2009–2010. It is estimated that the domestic consumption of pesticides in India is low (0.5 kg ha^{-1}) compared to several other countries (17.0 kg ha^{-1}, 14.0 kg ha^{-1}, 12.0 kg ha^{-1}, 9.4 kg ha^{-1}, 7.0 kg ha^{-1} and 5.0 kg ha^{-1} in Taiwan, China, Japan, Netherland, USA, and United Kingdom respectively) (Fig. 1) (Chauhan and Singhal, 2006). The consumption pattern of pesticides in India is different from that of the rest of the world. It is important to note in Fig. 2 that insecticides account for the largest consumption rate (76%) in India, followed by herbicides (10%) and fungicides (13%). This is a stark contrast to the rest of the world where herbicide (30%) and fungicide (21%) use is higher (Mathur, 1999). Those pesticides in India with the highest consumption rates include monocrotophos, endosulfan, phorate, chlorpyriphos, methyl parathion, quinalphos, mancozeb, paraquat, butachlor, isoproturon and phosphamidon (Bhushan et al., 2013). HCHs and DDTs together account for two-thirds of the total pesticides consumed

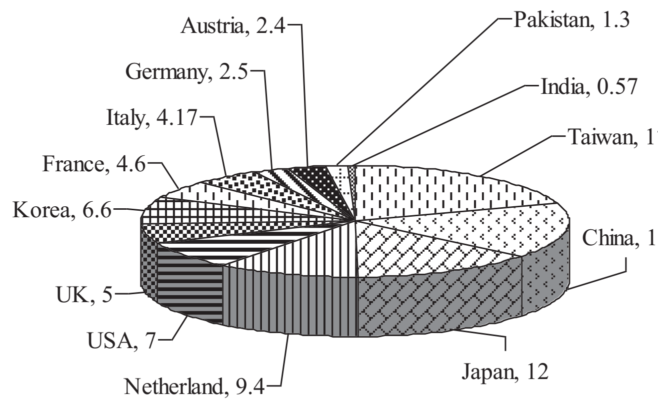

Fig. 1. Worldwide consumption (kg ha^{-1}) of pesticides.

in India (Kumari et al., 2001) for agricultural and public health purposes. In terms of volume, OCPs constitute 40% of pesticide use, followed by organophosphates (30%), carbamates (15%), synthetic pyrethroids (10%) and others (5%) (Fig. 3). In terms of value, organophosphates dominate (50%), followed by synthetic pyrethroids (19%), OCPs (16%), carbamates (4%), bio-pesticides (1%) and so on.

Consumption patterns are highly inconsistent and vary from one state to another (SEEP, 2010). Andhra Pradesh (South India) has the highest rate of pesticide consumption of any state (9289 MT), followed by Uttar Pradesh (North India) (8839 MT), and Maharashtra (West India) (6723 MT) (Fig. 4). The state of Tamil Nadu (South India), with an area of 130,000 km^2 uses 12,500 metric tons of pesticides annually. Maharashtra has an area of 307,000 km^2 and consumes only 6000 metric tons of pesticides (SEEP, 2010). However, the state of Punjab (West India), using 5770 metric tons of pesticides per annum, has an area of just 50,000 km^2. The average usage of pesticide is high in insecticide resistant management (IRM) and non-IRM areas (5.6 and 8 kg ha^{-1}, respectively) of Punjab state compared to the average national use (0.5–1 kg ha^{-1}) (Gupta, 2004; Peshin, 2005). These differences in consumption patterns could be explained by the use of land for cultivation, as well as crops raised. Cotton cultivation covers only 5% of area but consumes more than 50% of the total pesticides used (Rajendran et al., 2000). By contrast cereals and pulses cultivated in 58% of agriculture land, consume only 6 to 7% of the total pesticides used. Thus, cotton-growing states use disproportionately higher volumes of pesticides despite their smaller area. Currently, roughly 230 known pesticides are registered for use in India, of which 40% are organochlorines (FAO, 2005). Only 84 of the 230 pesticides registered, are actually used in the agriculture sector, and only 25–30% of the total cultivated area of the nation (143 million ha) is under pesticide cover. Today, rice has the highest rate of pesticide consumption (29%), followed by cotton (27%), vegetables (9%) and pulses (9%) (Fig. 5). It is evident in Fig. 5 that consumption of pesticides in cotton has been decreasing since 2001–02. This is because of the promotion and increased

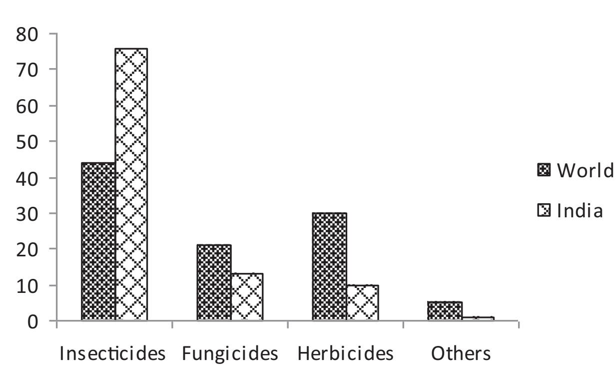

Fig. 2. Consumption pattern of pesticides in India.

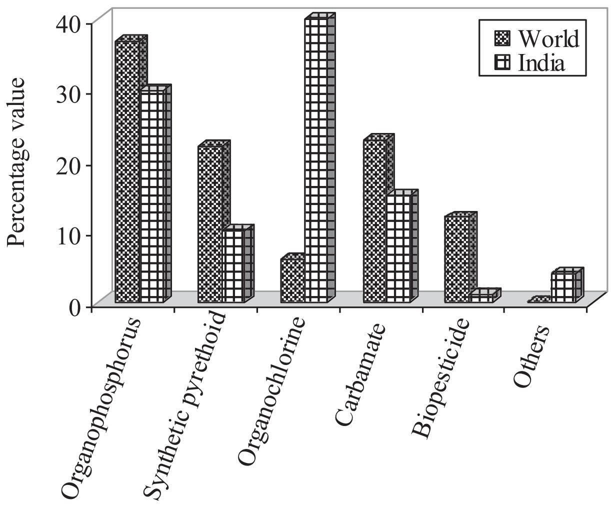

Fig. 3. Percentage chemical composition of insecticides being used in India.

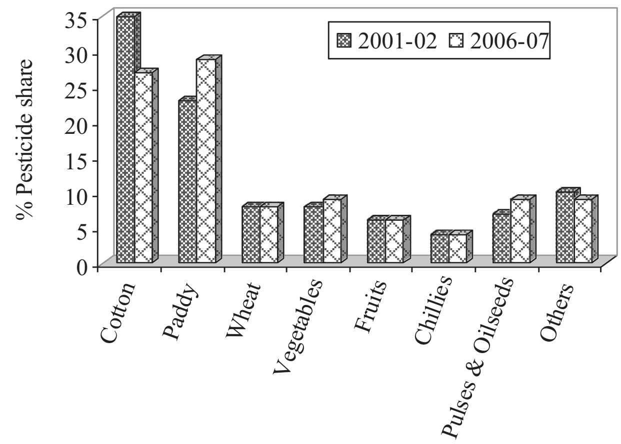

Fig. 5. Crop-wide consumption of pesticides in India during 2001–02 and 2006–07.

awareness of bio-pesticides amongst Indian farmers, replacing the use of existing pesticides.

Several global reports have ranked India amongst the leading pesticide consuming countries (Gupta, 2004; Mehrotra, 1993). The report, 'Pesticide Residues in Indian Food and Agricultural Products' (REF), disclosed that Indian food and agricultural products contained substantial quantities of pesticide residues. This is because Indian farmers use pesticides indiscriminately, due to lack of adequate education. Another reason for high pesticide residues is the use of sub-standard pesticides and incorrect advice issued to farmers by pesticide dealers. In most developed countries, such as the USA, many European nations, Taiwan, Japan and Korea, farmers are better informed and the spraying of pesticides is done in a meticulous manner to the target crop, thereby minimizing loss. However, in India this is not the case. To increase yield, Indian farmers use pesticides indiscriminately (Sarkar et al., 2008). To combat these issues, the Indian government should consider providing a training program for farmers, clarifying the correct dosages and methodologies of pesticide application (Kumari and Reddy, 2013).

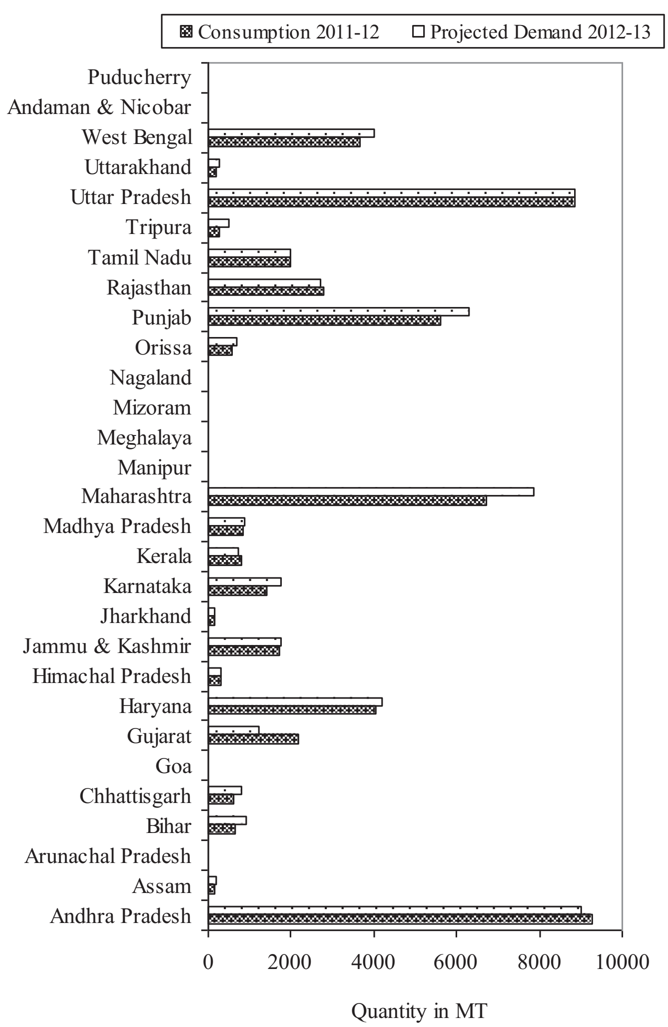

Fig. 4. State-wide consumption and projected demand for 2012–13 of pesticides in India.

1.2. Production of pesticides in India

Before 1970, the nine chemicals targeted by the Stockholm Convention were formulated and consumed internationally, and were used in agricultural crops or in the public health sector (Pandey et al., 2011). These pesticides were subsequently either banned or restricted in many countries in 1970s. Although banned in developed nations, these chemicals were permitted for use in many developing countries because of their low cost and versatility in industry (Tanabe et al., 1994). India started pesticide production in 1952 with the establishment of the BHC technical plant for the production of BHC at Rishra, near Kolkata (East India). Later, two more DDT manufacturing units were setup by Hindustan Insecticides Limited. Thereafter, several pesticide manufacturing companies, such as Union Carbide India Ltd. (1969), were set up to manufacture pesticides. Today, the Indian pesticide industry produces more than 500 pesticides through roughly 125 large and medium scale pesticide enterprises. A sharp increase in the number of Indian pesticide manufacturers led to over production of many technical grade pesticides, causing a decrease of 50% in the price of several key insecticides. This made pesticide use an economically viable option for Indian farmers to increase their crop production.

India is currently the second largest producer of pesticides in Asia, after China. It ranks as the fourth largest pesticide-producing nation in the world after the USA, Japan and China (Agnihotri, 2000; Kumar et al., 2012). India has more than 139,000 MT pesticide production capacity annually, with more than 219 technical grade and manufacturing units, and over 4000 formulation units. A steady growth in the production of technical grade pesticides was observed during the period 1958 (total production 5000 MT) to 1997 (total production 102,600 MT) (Table 1). However, pesticide production lowered to 80,900 MT in 1998 due to a drop in the sale of pesticides by 13%, as a result of crop failures due to adverse weather. This decline during 1998 can also be attributed to the poor financial status of those cotton farmers who suffered

Table 1
Pesticide production, consumption, import and export of India (in MT).

Year	Production	Consumption	Import	Export
1966	–	16.1	–	–
1972	–	25.8	–	–
1977	–	34.5	–	–
1982	–	47	–	–
1987	–	67.3	–	–
1992	84,170	84.8	–	–
1996	96,400	61,260	–	–
1997	102,600	56,114	–	–
1998	80,900	52,239	–	–
1999	80,900	49,157	–	–
2000	46,195	46,195	5379	39,042
2001	92,300	43,584	5934	47,930
2002	81,800	47,020	7240	50,085
2003	69,600	48,350	6795	55,693
2004	85,100	41,020	12,647	68,981
2005	94,000	40,672	18,102	79,100
2006	82,240	39,773	1711	94,500
2007	84,701	41,515	7080	110,700
2008	79,756	43,630	6360	96,268
2009	85,338	43,860	2668	184,537
2010	82,185	41,822	6269	125,818

heavy crop losses that year. Likewise, the reduced data requirements for registration of pesticides for exportation, by the Registration Committee of Pesticides, Government of India, encouraged pesticide production in the country. After 1998, the production trend of pesticides in India is almost stable, with total production limited to around 82,000–85,000 MT in 2009–2010. This is because the costs of discovering, developing and registering new molecules have increased substantially over the years.

The Indian pesticide industry produces two categories of pesticides: technical grade and formulated. Technical grade pesticides are extremely toxic and contain hazardous material; formulations are produced by the processing of technical grade pesticides with some emulsifiers. More than 60 technical grade pesticides are manufactured in India, mainly by multinational corporations, while the formulation market is highly fragmented and includes small formulators. At present, 30 technical grade pesticides have been banned (Table S.1), while seven, including DDT, are restricted (Table S.2) (Pandey et al., 2011). Aldrin, chlordane and heptachlor were banned in September, 1996, while DDT has been restricted since July 1989. Dieldrin was banned in July, 2003, Endrin in May 1990, and HCH in April 1997. Endosulfan and methoxychlor are allowed for use in India, however, HCB has never been registered as a pesticide (Pandey et al., 2011; UNEP, 2003).

1.3. Maximum residual limits (MRLs)

Residues of some persistent pesticides may remain longer in target crops and find their way into humans through the food chain (Bhushan et al., 2013). The residues of these pesticides should not exceed extreme limits as this may cause a threat to human health. Hence, maximum residue limits (MRLs), acceptable daily intake (ADI) and theoretical maximum daily intake (TMDI) have been proposed and developed to monitor residues of these pesticides in the food chain (Bhushan et al., 2013). The MRL is the maximum residue limit of pesticides, which may be considered normal, found in a product treated with them if good agricultural practices have been followed. An ADI is the maximum acceptable intake of pesticides from all dietary sources in a day, without their posing any chronic health risk. The TMDI is an estimate of the maximum intake of pesticides with the existing MRLs for a person, resulting from a particular dietary practice.

There are no globally accepted standards for pesticide residues available, however, FAO and WHO have recommended that the standard acceptable rate for POPs in any sample should be zero tolerance (FAO/WHO, 2011). The most widely accepted and adopted safety limits for pesticides are those set by the FAO and WHO, resulting from a Joint Meeting on Pesticide Residues (JMPR) and the Codex Alimentarius Commission. JMPR recommendations are based on a peer review of international data, together with an examination of pesticide occurrence, treatability, detectability and effect (FAO and WHO, 2011; Fishel, 2010). It is also opined that pesticide limits should not be lower than the analytical limits of quantification (LOQ) achievable in qualified laboratories under routine operating conditions. To minimize the risk of pesticides at reasonable cost, the majority of nations have enforced regulatory limits, either in the form of standards (which are enforceable) or guidelines (which are desirable levels).

Central Insecticides Boards and Registration Committee of India (CIBRC) is the regulatory body of the Indian government responsible for the registration of pesticides, while the Food Safety and Standard Authority of India (FSSAI) sets the MRLs of registered pesticides. Indian standards allow considerably increased levels of persistent organic pesticides compared to European Union (EU) standards. For example, the regulatory limit for lindane is 100 times higher than those of the EU (IPEN, 2006); acceptable levels of aldrin and dieldrin allowed are 10 times higher in Indian food than in the Czech Republic (IPEN, 2006); and acceptable limits permitted for DDT and its metabolites, heptachlor and endosulfans, are seven times, five times, and 40 times higher respectively in Indian food than in the Czech Republic. In Indian drinking water, the residual limit for lindane is roughly 20 times higher than in the Czech Republic; heptachlor is more than 30 times than in the EU; and 200 times more aldrin and endosulfan is permitted than in the Czech Republic (IPEN, 2006). These figures indicate the alarming variation of MRLs for pesticides in India. It appears that the levels are being wrongly formulated, while their implementation status is much worse.

2. Persistent organic pesticide levels in the environment

2.1. Persistent organic pesticide levels in air

Pesticides from non-target agricultural crops or volatilization from the treated area may contaminate air, soil, and water. Various instances of pesticide drift occur during every application, even from ground equipment (Glotfelty and Schomburg, 1989). Pesticide drift accounts for approximately 2 to 25% chemical loss during application; this can spread over a wide area, from a few yards to several hundred miles. There are many cases of pesticides drifting, in the US alone, every year (Que et al., 1975). Many pesticides volatilize (i.e. evaporate from the soil and foliage, and move away from the initial application area) and reach into every area of the environment, thereby contaminating them adversely (USGS, 1995). More than 80–90% of pesticides are volatilized within a few days of application on crops or in the health sector (Majewski and Capel, 1995).

Despite the limited information available on persistent organic pesticide residues in Indian air, every study found high levels to be present in samples taken (Table 2). Rajendran et al. (1999) analyzed the concentration of DDT and HCH in air samples from the tropical coastal environment at Parangipettai (Southeast India). They found that the concentration of DDT and HCH ranged from 0.16 to 5.930 ng m^{-3} and 1.45 to 35.6 ng m^{-3}, respectively. Zhang et al (2008) conducted monitoring of persistent organic pesticides in urban, rural, and background (mangrove or wetlands) sites along the coastal region of India. They found the concentration of DDTs, HCHs, and chlordanes ranging from 16 to 2950 pg m^{-3}; from 66 to 5400 pg m^{-3}; and from 0.45 to 1120 pg m^{-3} respectively. The highest levels of all detected persistent organic pesticides (except endosulfans) were observed at the urban sites, indicating the dominant areas of usage and emissions. Chakraborty et al. (2010) studied the concentrations of different kinds of OCPs in major Indian cities, using passive and active samplers. They found that the concentration of HCHs observed in Indian cities was the highest reported across the globe and ranged from 890 to 17,000 pg m^{-3} (mean 5400 pg m^{-3}). DDTs ranged from 250 pg m^{-3} to 6110 pg m^{-3} (mean 1470 pg m^{-3}); chlordanes from 290 to

Table 2
Persistent organic pesticide residues (pg/m^3) in air from regions across India.

Location	ΣDDTs	ΣHCHs	α-Endosulfan	References
Tamil Nadu	31	375	–	Srimural et al. (in press)
Imphal	159	169	30	Devi et al. (2011)
Thoubal	188	194	89	Devi et al. (2011)
Waithou	175	242	80	Devi et al. (2011)
Laksar	619	902	972	Pozo et al. (2011)
Sukanda Devi	8	110	184	Pozo et al. (2011)
Patna	1832	2809	2530	Pozo et al. (2011)
Mudhol	91	1728	25,630	Pozo et al. (2011)
New Delhi	1980	3190	230	Chakraborty et al. (2010)
Agra	1520	5730	340	Chakraborty et al. (2010)
Goa	2310	13,240	390	Chakraborty et al. (2010)
Kolkata	994	1140	372	Zhang et al. (2008)
Mumbai	1637	1549	498	Zhang et al. (2008)
Baroda	566	2275	317	Zhang et al. (2008)
Chennai	2901	5253	680	Zhang et al. (2008)
Bangalore	284	227	3	Zhang et al. (2008)
Cuddalore	116	289	185	Zhang et al. (2008)
Portonovo	647	1096	992	Zhang et al. (2008)
Pichavaram	132	73	4	Zhang et al. (2008)
Gulf of Kutch	112	131	12	Zhang et al. (2008)
Thane Creek	55	17	13 l	Pandit and Sahu (2001)
Parangipettai	5930	3550	–	Babu Rajendran et al. (1999)

5260 pg m^{-3} (mean 1530 pg m^{-3}); endosulfans from 240 to 4650 pg m^{-3} (mean 1040 pg m^{-3}); and hexachlorobenzene from 120 to 2890 pg m^{-3} (mean 790 pg m^{-3}). Chlordanes and endosulfans were lower than levels reported in southern China. γ-HCH dominated the HCH signal, reflecting the widespread use of lindane in India.

Another study monitored the level of various OCPs in urban, rural and mountain sites of Manipur, Northeast India, and found the maximum concentration of HCHs in the mountain region during the summer season (403 pg m^{-3}), followed by that of the rainy season (349 pg m^{-3}) (Devi et al., 2011). DDTs had a high concentration with 384 pg m^{-3} at the rural site and 379 pg m^{-3} at the urban site during the summer season. Endosulfans and chlordane were found high in concentration during the summer season (260 pg m^{-3}) and low during the monsoon seasons (44 pg m^{-3}) at the rural site. Pozo et al. (2011) assessed the concentration of persistent organic pesticides in the air across several Indian agricultural regions, using PUF-PAS for the first time. The project was conducted as a sub-project of the Global Atmospheric Passive Sampling (GAPS) Network. They found high concentrations of HCH (292 pg m^{-3}), endosulfans (2770 pg m^{-3}) and DDT (247 pg m^{-3}).

Recently, Srimural et al. (in press) conducted passive air sampling in urban, suburban, coastal, and agriculture areas in Tamil Nadu (Southern India). They found the total concentration of 13 OCPs ranging from not detected (ND) to 41,400 pg m^{-3}. DDTs, DDD, heptachlor, and mirex were predominant during monsoon season. The elevated α/γ isomer ratio of HCH (5.03 pg m^{-3}) during summer revealed fresh or recent usage of HCHs in coastal areas.

2.2. Persistent organic pesticide levels in water

At present, every freshwater body (rivers, lakes and estuaries) is contaminated with pesticide. Only about 10% of the wastewater generated is treated; the rest is discharged untreated into the water system. The pollutants then find their way into groundwater, rivers and other water bodies, which ultimately ends up being used in households. As a result this water is highly contaminated with chemicals and disease-causing microbes. Subsurface run-off from agricultural fields contains a variety of OCPs, and may carry chemical fertilizers and pesticides into local rivers (Barbash and Resek, 1996). Because of the mobility of pesticides in soil, groundwater becomes susceptible to pesticide contamination. There are several factors, such as the solubility of pesticides in water, environmental factors (soil, weather and seasons), and distance of water sources from the area of application, that may influence the spread of pesticide contamination (Gustafson, 1993).

A number of researchers have reported pesticide residues in groundwater and drinking water across India (Bansal and Gupta, 2000; Bouwer, 1989; Dikshit et al., 1990; Jani et al., 1991; Kumar et al., 1995; Ray, 1992) (Table 3). HCHs and DDTs were detected in various water sources, such as wells, hand pumps and ponds in Bhopal (Central India). Well water samples in Bhopal contained 4640 μg/l of HCHs, and 5794 μg/l of DDT (Bouwer, 1989). Drinking water analysis results from Ahmedabad (West India) showed 23.90–2488.70 ng/l of total HCHs and about 10.90–314.90 ng/l of total DDT (p,p-DDE, o,p′-DDT and p,p′-DDT) (Jani et al., 1991). Ray (1992) found high levels of pesticide residues in groundwater samples from irrigation wells, domestic wells and canals in Aligarh (North India). Aldrin and dieldrin residues have been also detected in soil, water, fish and clam samples from various locations in Delhi.

Even surface water samples from five lakes of Nainital (North West India) contained 6.054–31.336 μg/l of DDT and 3.121–8.656 μg/l of HCH, in places where insecticide has never been used for vector control (Dua et al., 1998). This indicates high mobility of DDTs and HCHs. BHC and DDT residues were also detected in the waters of Keoladeo National park and Bharatpur, Rajasthan (West India) in the range of 0.58 μg/l and 3.86 μg/l (Murlidharan, 2000).

Pesticide contamination of groundwater is of vital concern for the people of India, especially in those areas where the main source of drinking water is groundwater aquifers. It is estimated that around 260 million l of industrial waste, together with runoff from the 6 million tons of fertilizers and 9000 tons of pesticides, may enter into the river system each year (K.P. Singh et al., 2005). More than 90% of water and fish samples from all streams sampled contained often, one, or more pesticides. Analysis of drinking water samples from across India showed the presence of HCHs and endosulfan isomers and DDT metabolites in high concentrations (Bakore et al., 2004; Sankararamakrishnan et al., 2005; V.K. Singh et al., 2005).

Akhil and Sujatha (2012) studied the prevalence of OCP in the groundwater of the Kasargod district in Kerala (South India). High concentrations of OCPs were observed for endosulfan, followed by hexachlorobenzene (BHC). Groundwaters in the Ambala and Gurgaon districts, and surface water in the Hisar district of Haryana (Central India), were found to be contaminated with isomers of HCH and endosulfan, as well as metabolites of DDT. The total concentrations of HCH, DDT and endosulfan were 87.6 ng/l, 848.2 ng/l, and 27.4 ng/l and 99.8 ng/l, 275.3 ng/l and 164.2 ng/l respectively, for the Ambala and Gurgaon districts. In the case of the Hisar district, the values were 78.5 ng/l, 115.9 ng/l, and 53 ng/l, respectively (Kaushik et al., 2012).

Lari et al. (2014) analyzed OCPs in the surface water and groundwater from Maharastra India (West India). They found surface water to be more highly contaminated with OCPs than groundwater. The concentration of those OCPs which were more frequently found in groundwater were: α-HCH (0.39 μg/l in Amravati region),α-endosulphan (0.78 μg/l in Yavatmal region), chlorpyrifos (0.25 μg/l in Bhandara region) and parathion-methyl (0.09 μg/l in Amravati region), whereas α,β,γ-HCH (0.39 μg/l in Amravati region),α,β-endosulphan (0.42 μg/l in Amravati region), dichlorovos (0.25 μg/l in Yavatmal region), parathion-methyl (0.42 μg/l in Bhandara region), phorate (0.33 μg/l in Yavatmal region) were more prominent in surface water.

2.3. Persistent organic pesticide levels in soil/sediments

Presently, most pesticides used in agricultural fields are synthetic organic compounds which may pass into the soil by missing their intended target, through surface and subsurface runoff from treated plants or by spillage during application. Dissipation of chemical pesticides in the soil depends on the characteristics of the soil, the nature of the chemical compound, cropping system, irrigation pattern, and the ambient climatic conditions (Agnihotri et al., 1994). Pesticide

Table 3
Persistent organic pesticide residues (ng/l) in water from regions across India.

Location	Type of water body	DDTs	HCHs	Endosulfan	References
Bhandara	Groundwater	Nd	Nd–60	Nd–720	Lari et al. (2014)
Bhandara	Surface water	Nd	Nd–60	Nd–80	Lari et al. (2014)
Amravati	Groundwater	Nd	Nd–39	Nd–60	Lari et al. (2014)
Amravati	Surface water	Nd	Nd–39	Nd–42	Lari et al. (2014)
Yavatmal	Groundwater	Nd	Nd–80	Nd–78	Lari et al. (2014)
Yavatmal	Surface water	Nd	Nd	Nd	Lari et al. (2014)
Devprayag	Ganges river	nd	7.24	Nd	Mutiyar and Mittal (2013)
Tamiraparani	River	0.01–0.72	0.01–0.78	–	Kumarasamy et al. (2012)
Ambala	Groundwater	940.3	147.1	13.1	Kaushik et al. (2012)
Gurgaon	Groundwater	357.1	123.8	266.4	Kaushik et al. (2012)
Hisar	Tap water	83.5	13.1	51.7	Kaushik et al. (2012)
Nagaon	River, ponds	6121	4911	–	Mishra and Sharma (2011)
Dibrugarh	River, ponds	5402	4403	–	Mishra and Sharma (2011)
Nagaon	Borewell/dug well	6904	5574	–	Mishra and Sharma (2011)
Dibrugarh	Borewell/dugwell	6549	5168	–	Mishra and Sharma (2011)
Haryana	Ghaggar river	587.30	119.74	–	Kaushik et al. (2010)
Pilbhit	Gomti River	5.97	46.69	9.64	Malik et al. (2009)
Karnataka	Cauvery river	1750	2430	–	Abida Begum et al. (2009)
Kolkata	Tank water	193	1157	–	Ghose et al. (2009)
Kolkata	Groundwater	10	100	–	Ghose et al. (2009)
Chennai	Ennore creek	10.07	15.86	29.21	Sundar et al. (2010)
Jaipur	Tubewells	–	7240	–	Sharma and Khan (2009)
Delhi and Haryana	Yamuna River	387.90	310.25	–	Kaushik et al. (2008)
Haryana	Groundwater	280	1086	180	Kumari et al. (2008)
Thiruvallur	Open wells	4743	10,014	4518	Jayshree and Vasudevan (2007)
Unao	Streams, ponds	Nd–230	1880–1950	Nd–130	Singh et al. (2007)
Lucknow	Rainwater	0.53	23.48	2.99	Malik et al. (2007)
Mumbai	Seawater	12.45	5.42	–	Pandit et al. (2006)
Hyderabad	Groundwater	613–742	–	5917–8879	Shukla et al. (2006)
Unao	Dug wells	17	209	–	K.P. Singh et al. (2005), V.K. Singh et al. (2005)
Tamil Nadu	Vellar river	0.057–4.8	26–3900	–	Ramesh et al. (1990)
South India	Cauvery river	2.4	102.2	–	Rajendran and Subramaniyam (1997)

residues in the soil may subsequently pollute groundwater aquifers through leaching, thereby affecting the quality of the agricultural crops and product. This in turn may affect the health of the consumers in the region (Singh et al., 2004). The persistent organic pesticides found in the crops are likely to have been derived from soil, water or air. Pesticide pollution in soil is the most important source of exposure.

Volatilization (and subsequent diffusion), decomposition, retention by the soil, and the transportation by water are the main factors which influence the cost potential of pesticides from soil to groundwater. Synthetic organic pesticides may decompose in different ways. If applied to plant foliage or the soil surface, pesticides may be broken down rapidly by sunlight. Unlike, organochlorine compounds (chlordane, DDT and dieldrin) decompose very slowly and may live for years. These pesticides, however, are of little concern as they are relatively insoluble in water and are retained strongly by the soils.

Several cases of pesticide pollution, including DDTs, in soil have been reported across the globe. Pesticide residues have been widely distributed in all types of soil (Vassilev and Kambourova, 2006) (Table 4). Abhilash and Singh (2009) studied the seasonal variation of HCH in an open soil–plant rhizospheric soil system of a contaminated industrial area of Uttar Pradesh (North India). The mean concentration of total HCH in open soil and rhizospheric soil samples were found in the range of 38.64–104.18 mg kg^{-1} and 8.38–26.05 mg kg^{-1} respectively. Sediment samples collected from in and around Keoladeo National Park, Bharatpur Rajasthan (West India), showed OCP contamination in park soil (Bhadouria et al., 2012). Total concentration of OCPs in the sediments varied from 0.1173 ppm (dieldrin) to 5.55 ppm (γ-HCH) in the samples from inside the park, while samples from outside the park ranged from 0.1245 ppm (4,4′-DDD) to 7.54 ppm (γ-HCH).

Kata et al. (in press) assessed the distribution pattern and potential source of OCP residues in soil samples from Hyderabad city (South India). The results reveal that δ-HCH, p,p-DDE, endrin ketone and endosulfan sulfate were most frequently detected, while heptachlor epoxide and p,p-DDD were less present. The total concentration of organochlorine pesticides in soil samples varied from 129 μg kg^{-1} to 1001 μg kg^{-1} dry weight. The ratio of (DDD + DDE)/DDT in the soils range from 0.74 to 7.81, which indicate the usage of DDT in the recent past.

Kumar et al. (2014) found that the concentration of OCPs in Korba (Central India) is to be average. The concentration of HCH and DDT ranged between 0.9–20 μg kg^{-1} and 2–315 μg kg^{-1} respectively. However, it was lower than recommended soil quality guidelines, indicating low eco-toxicological risk.

Samples from surface sediment (113), ponds and wetland sediments (43), and river and stream sediment (70) were collected from Nagaon and Dibrugarh (North East India), showing that many sediments are contaminated with OCPs, exceeding the sediment quality guidelines (Mishra et al., 2013). The mean concentration of $\sum$HCH and $\sum$DDT in sediments was found to be 287 ng/g (71.2–834 ng/g) and 321 ng/g (30.1–918 ng/g) for Dibrugarh, while 330 ng/g (39.2–743 ng/g) and 378 ng/g (72.5–932 ng/g) were measured for Nagaon, respectively. DDTs and HCHs in sediments were well influenced by the total organic carbon, clay and silt content of sediments.

High levels of OCPs are also reported in agricultural soils from the Northern states of India (Kumar et al., 2011). The average concentration of OCPs was 37.67 ± 0.33 ng/g (dry weight — DW) while HCHs alone accounted for 93%, followed by DDT (4.27%) and endosulfan (2.51%). The α/γ ratio of HCH (<0.01–8.64) reflects the use of technical as well as lindane formulations. The ratio of p,p′-DDT/p,p′-DDE (0.16) and o,p′-DDT/p,p′-DDT (<0.01) indicates the contamination of soils with the past use of technical DDT. The mean concentrations of endosulfan and dieldrin were 0.95 ± 0.53 ng/g (DW) and 0.16 ± 0.07 ng/g (DW), respectively.

Kumari and Reddy (2013) analyzed DDTs and HCHs in urban soil from Kurukshetra (Central India) and found low levels of HCHs and DDTs. The average concentrations of total HCHs and total DDTs were well below guidelines and ranged from 0.56 to 8.52 mg kg^{-1} and from 0.54 to 37.42 mg kg^{-1} respectively. The compositional analysis of HCH isomers suggests contaminations from recent usage.

Table 4
Persistent organic pesticide residues (ng/g) in soils/sediments from regions across India.

Location	Types of soil	DDTs	HCHs	Endosulfan	References
Hyderabad	Surface soil	61.72	75.89	18.05	Kata et al. (in press)
Korba	Residential soil	2.1–315	0.9–16	–	Kumar et al. (2014)
Assam	Forest soil	0.042–0.286	0.007–0.025	0.019–0.041	Devi et al. (2013)
Tripura	Forest soil	Nd–0.09	0.003–0.049	0.009–0.057	Devi et al. (2013)
Manipur	Forest soil	0.017–2.68	Nd–1.15	0.02–0.47	Devi et al. (2013)
Kurukshetra	Surface soil	0.5–37.4	0.6–8.5	–	Kumar et al. (2013)
Andaman Island	Surface soil	1.31–5.90	–	4.79	Murugan et al. (2013)
Dibrugarh	River sediments	291	255	–	Mishra et al. (2013)
Dibrugarh	Pond sediments	366	335	–	Mishra et al. (2013)
Nagaon	River sediments	339	279	–	Mishra et al. (2013)
Nagaon	Pond sediments	449	422	–	Mishra et al. (2013)
Keoladeo National Park	National park Sediment	1321.8	9039.9	4569	Bhadouria et al. (2012)
NCR Delhi	Surface soil	1.61	34.96	0.95	Kumar et al. (2011)
Yamuna river	River sediments	16.87–136.35	1.03–104.6	–	Pandey et al. (2011)
Gomti River	River sediments	49.84	16.52	0.13	Malik et al. (2009)
Sunderban	Wetland soil	0.05–11.5	0.05–12.4	–	Sarkar et al. (2008)
Hissar	Agricultural soil	1–66	2–51	2–39	Kumari et al. (2008)
Unnao	Surface soil	Nd–74	0.1–7.3	Nd–13.07	Singh et al. (2007)
Hugly estury West Bengal	Surface sediments	0.957	0.243	–	Guzzella et al. (2005)
Thiruvallur	Surface soil	10.3	75.3	–	Jayashree and Vasudevan (2005)
Dehradun	Surface soil	117	326	–	Babu et al. (2003)
Hugly estury, Sunderban	Surface soil	58	134	–	Bhattacharya et al. (2003)
Aligarh	Agricultural soil	34	88.9	–	Nawab et al. (2003)
Kasimedu	Agricultural soil	0.1	0.1	–	Senthilkumar et al. (2001)
Ennore	Agricultural soil	35	2.1	–	Senthilkumar et al. (2001)
Cochin	Agricultural soil	3.8	4.8	–	Senthilkumar et al. (2001)
Visakapattnam	Agricultural soil	0.1	0.21	–	Senthilkumar et al. (2001)
Agra	Agricultural soil	0.93	0.50	0.03	Singh (2001)
West coast of India	Surface sediments	1.47–25.17	0.85–7.87	–	Sarkar et al. (1997)
Farrukhabad	Agricultural soil	337	158	–	Agnihotri et al. (1996)
Haridwar		270.5	61.12	–	Dua et al. (1996)

3. The role of India in the global distribution of persistent organic pesticides

Due to the severe human health risk associated with persistent organic pesticides, both researchers and politicians have shown a deep interest in understanding the fate and behavior of persistent organic pesticides in the environment, as well as the discovery of persistent organic pesticides in pristine environments far from emission source. The long range atmospheric transportation (LRAT) movement of persistent organic pesticides has led to the negotiation of protocols in various countries (e.g. UN/ECE, UNEP) for their reduction or elimination, reducing the risks to regional and global environments. As these pesticides are volatile and resistant to photolytic, biological and chemical degradation, they become persistent throughout the world. The distribution of persistent organic pesticides around the world has been discussed in many studies (Ali et al., 2014; Zhang et al., 2011). Researchers have reported the ubiquitous nature of these chemicals in their studies.

There are several factors that play a significant role in the dispersal of persistent organic pesticides on a global scale. First, industrial activities are increasingly moving to Asia, resulting in major emissions from countries like China and India (Lohmann et al., 2007). Due to shifting phenomenons, such as monsoons, certain emissions will get transported into the southern hemisphere (Dachs et al., 1999; Semeena et al., 2006). Secondly, POP emissions linked to low-temperature combustion processes will increase for Asia, Africa and S. America, where biomass burning and forest fires are common (Crutzen and Andrea, 1990; Kasischke and Penner, 2004). Thirdly, many currently used pesticides are increasingly being used across the globe, indicating their major use in agricultural areas of S. America, Africa and Asia (Pozo et al., 2006).

Tanabe et al. (1994) assumed that the large quantities of persistent organochlorines used in tropical regions were released into the atmosphere and dispersed through 'long-range atmospheric transport' in global terms. HCHs and DDTs transported through the atmosphere are absorbed and deposited in seas and oceans near point-source contamination on land. However, PCBs, as well as CHLs, are likely to be more mobile over the oceans than HCHs and DDTs.

Interestingly, India did not sign the Convention on Long Range Transboundary Air Pollution (CLRTAP), which is aimed at limiting and gradually preventing the discharge of air pollutants (Bull, 2003). Information on the source and emission of persistent organic pesticides in the Indian region and the fate of these compounds in global environment is not fully understood. A number of researchers around the world have reported that persistent organic pesticides originating from India adversely affect the neighboring countries (especially the southwest of China and other South Asian countries) leading to its distribution on a global scale. Further, some researchers have opined that the Indian monsoon system plays an important role in the transportation of persistent organic pesticides across major Indian cities.

Iwata et al. (1993) reported that tropical Asia, including India, is a significant emission source of global persistent organic pesticide contamination. Rajendran et al. (1999) found low level of OCPs in air samples collected from the coastal regions of southern India. They suggested that the low level of OCPs in the air may be due to more volatile HCH compounds being transported from the Indian tropics to colder regions and their resultant rapid condensation, thereby leading to global distribution. Recently, Zhang et al. (2008) also found that coastal Indian cities are polluted by OCPs, and the source is due to transportation of air mass originating from Indian major cities through coastal areas. This might affect global POP concentration.

Pandit and Sahu (2001) also suggested that OCPs are removed rapidly from inland water and agricultural fields, and washed out, in the majority of cases, to the ocean. The concentration of DDTs, HCHs and endosulfans in the air is dependent on concentrations in the surface seawater. Further, they theorized that the volatilization process enhances the active transfer of OCPs from seawater into the atmosphere. Today, levels of organochlorine pesticides in India remain exceptionally high (Shunthirasingham et al., 2010). The highest levels of α-HCH, together with high γ-HCH levels, suggests that lindane use is now the major source of α-HCH in the global atmosphere (Shunthirasingham et al., 2010). Chakraborty et al. (2010) studied the concentration of OCPs in several major cities in India and found that the concentration of HCH observed in India was the highest concentration reported across the globe, and that the cause of this could be local and regional sources of OCPs within India.

Pozo et al. (2011) analyzed the different types of persistent organic pesticides in the agricultural regions of India and found wide spread use of OCPs, with the potential for most OCPs to undergo long range atmospheric transport. In a recent study of air samples from Manipur (Northeast India), Devi et al. (2011) found elevated levels of OCPs, the source of which could be long range atmospheric transport (LRAT) of OCPs from Indian cities located on coastal region, crossing through Kolkata (East India) to reach Manipur.

Persistent organic pesticides are known to undergo LRAT, reaching remote regions where they have never been produced or used. In order to better understand the key processes that control persistent organic pesticide distribution, environmental data is needed from across the world. Various studies based on modeling and simulation analysis have indicated that persistent organic pesticides originating from Indian sources may affect the neighboring countries, especially the Tibetan Plateau (TP). Several previous studies have revealed contamination of HCH in TP is largely attributed to emissions from Indian sources (Gong et al., 2010; Wang et al., 2010).

Gong et al. (2010) reported high levels of γ-HCHs in the Lhasa atmosphere and advised that the source could be due to current usage of lindane in India and atmospheric transportation via Indian monsoons. In contrast to this, Li et al. (2006) found the source of OCPs in Lhasa to be current use and local emission. Xu et al. (2011) studied the transportation characteristics of OCPs and their potential sources at remote stations, such as Tengchong Mountain (TM) in the south-western part of China, and found elevated levels of HCHs and DDTs in the air. Potential source contribution function (PSCF) analysis suggests that high levels of OCPs at TM are mainly caused by LRAT from India and Southern China.

Multiple modeling simulation studies conducted between 2005 and 2007 to assess the cancer risk in China subject to γ-HCH emissions sources across China and India showed that current use of lindane in India strongly contributed to the risk of cancer in Western China (Xu et al., 2013a). The contribution was almost one order of magnitude higher than that from the reemission of γ-HCH from previously contaminated soils in India. However, Indian sources only made a minor contribution to human risk of cancer in China (Xu et al., 2013a). Recently, Xu et al. (2013b) studied the environmental fate of α-HCH in Asia during the period 1948 to 2008, using the Chinese Griddled Pesticides Emission and Residue Model (ChnGPERM) simulation model. They found that the environmental fate of a-HCH in the Southern TP was influenced by the atmospheric outflow from Indian sources; whist sources in China played a major role for its accumulation in eastern TP. Some persistent organic pesticides originating from distant sources have been transported and accumulated in TP due to orographic cold trapping. Based on backward air mass trajectory analysis, Tao et al. (2011) reported that the accumulation of OCPs and PAHs in the soils of the west and northwest Qinghai-Tibetan Plateau is due to LRAP from East Asia.

The transportation and fate of persistent organic pesticides to the TP might also be influenced by Indian monsoon systems. Several studies conducted in the TP on persistent organic pesticides have reported the role of Indian sources, driven by Indian monsoons. Wang et al. (2006) evaluated the source and transport pathways of OCPs and PAHs in the Zhangmu–Nyalam (NZ) region (Central Himalayas) and found the transportation and fate of pollutants is influenced by the air mass movement dominated by the Indian monsoon from south Asia. Wang et al. (2010) found that persistent organic pesticide concentrations in the air from regions impacted by the Indian monsoon are higher than in other regions of the TP. Tian et al. (2009a) employed the Canadian Model for Environmental Transport of Organochlorine Pesticides (CanMETOP) simulation model to assess the contributions of γ-HCH in Chinese air and soil from four major γ-HCH residual regions (India, USSR, Europe and China). They found that Chinese sources contributed 75% of γ-HCH in the north-eastern area, followed by Indian sources whose largest contribution in the north-western region was 63%. Li et al. (2006) found high levels of OCP concentration in air samples from the Mt. Everest region (the south Tibetan Plateau) in 2002. The back trajectory analysis showed the OCP emissions to be from the Indian subcontinent.

Xu et al. (2009) observed good agreement between soot emissions in the India subcontinent and black carbon in southern Tibetan glaciers. Recently, Yang et al. (2013) hypothesized that some semi-volatile compounds, such as OCPs and PAHs, in southeast TP could be transported from the densely populated and intensely agricultural Indian subcontinent, brought by Indian monsoon. Yang et al. (2010) studied the transportation and fate of OCPs and PCBs in TP and found that the sources are mostly attributed to the southwest or southeast air mass from the Indian subcontinent.

Tian et al. (2011) investigated the long-term environmental fate of α-HCH in China from 1952 to 2007 using the ChnGPERM simulation model. They found that α-HCH contamination in southwest China is mostly influenced by foreign sources, notably sources in India and other Southern Asian countries. Tian et al. (2009b) assessed the impact of the East Asian summer monsoon on the temporal and spatial distribution of α-HCH in China and other East Asian countries. They opined that the East Asian summer monsoon provides a major atmospheric pathway and summer outflows to α-HCH over East Asia. Yang et al. (2008) believed that the OCPs in southeast TP are strongly influenced by the summer Indian monsoon, as OCPs are mainly used during summertime. The distribution pattern throughout the TP strongly suggests that the main sources of OCPs may be from the Indian subcontinent.

Xu et al. (2012) assessed the environmental fate and source–receptor relationships of β-HCH from 1948 to 2009 over Asia, and its association with environmental factors using ChnGPERM. Modeling results suggested that contamination of β-HCH in soil residue of the TP is due to Indian, Southeast Asian and Chinese sources, driven by the Indian summer monsoon and East Asian monsoon respectively. Further, they found that residue levels β-HCH from Indian sources was higher than from South Asian countries (Xu et al., 2012). Likewise, Xu et al. (2013a) found a significant influence of India γ-HCH emissions on western China, assisted by the Indian monsoon, leading to a north-eastward migration of contaminants from India. However, in contrast to this, Li (1999) reported that Indian source contribution was not considerable due to the significant wet disposition during Indian monsoon season.

Recently, Sheng et al. (2013) studied the source of persistent organic pesticides in TP using the PSCF model and found air circulation driven by the Indian monsoon to be responsible for the transportation of persistent organic pesticides to the TP rather than local source emissions. The back trajectory analysis indicated that p,p′-DDT is arriving at the sampling site from the east coast region of India, where metropolitan cities, such as Kolkata, Bhubaneswar, Chennai, are located.

4. Health impact and ecological toxicity

Around the world, pesticides are considered important public health tools, used to prevent vector borne diseases and to increase food production. However, extensive use of such pesticides results in substantial health and environmental threats (Forget, 1993; Jeyaratnam, 1985). Pesticides constitute an important component in agricultural development and the protection of public health in India. The tropical climate of India is conducive to pest breeding thereby making the population increasingly vulnerable to pest-borne diseases. So far, roughly 20 major diseases in India have been brought under control by the application of pesticides; major amongst them are malaria, filariasis, dengue, Japanese encephalitis, cholera and louse-borne typhus. Likewise, synthetic organic pesticides are preferred to control weeds, insects and other organisms in agricultural and non-agricultural settings.

The hydrological system is one of the important pathways for the distribution of pesticides because it supplies water to both humans and natural ecosystems. Water is a primary medium through which pesticides are transported from an application area to other areas of the environment. Because of their slow decomposition rates, long half-life and high stability in the environment, OCPs are the most harmful class of pesticides. They can remain and accumulate in the upper trophic levels of food chains (Gallo and Lawryk, 1991; Kocan and Landolt, 1989; Raju et al., 1982). Mild cases of pesticide poisoning can result in headaches, dizziness, gastrointestinal disturbances, numbness and weakness of the extremities, apprehension and hyperirritability. Chlorinated hydrocarbons when absorbed into the body, are not metabolized rapidly and instead are stored in fatty tissues. Some pesticides have been proven to be endocrine disruptors. They are known to elicit their adverse effects by mimicking or antagonizing natural hormones in the body. It has been assumed that long-term low-dose exposure is linked to human health effects such as immunosuppression, hormone disruption, diminished intelligence, reproductive abnormalities and cancer.

Organophosphate, organochlorines and aluminum phosphide (AlP) compounds are common pesticides used worldwide. In most developing countries, AlP is used as both an outdoor and indoor pesticide due to its low cost and effectiveness. Also, it is free from toxic residue and does not affect seed viability (Chugh, 1992). It is worrying to know that today, not a single section of the population is completely protected against pesticide exposure and the resultant possible health effects. High risk groups (with low socioeconomic status), especially those in developing countries, are more prone to adverse pesticide effects. An estimate showed that globally, as many as 3 million cases of acute, severe poisoning which occur due to intoxication of pesticides are recorded annually. Pesticide poisoning accounts for the deaths of some 220,000 people annually, with many more cases going unreported (Gunnell and Eddleston, 2003; WHO/UNEP, 1990). Cases of pesticide poisoning are more frequent in developing countries (about 13 fold more than in developed nations) compared to highly industrialized nations which consume 85% of world's pesticide production (Forget, 1989). The tropical and temperate climate of many countries in Central and South America, Africa and South East Asia is becoming “breadbaskets” for pesticides. Most developing nations have yet to develop strict strategies for controlling the use of pesticides (Mbiapo and Youovop, 1993).

Pesticide poisoning is a significant health issue in India. However, no national data is available on the total cases of pesticide poisoning. Also, there is no network of poison control centers in India. The pattern of pesticide suicide is dominant in India, whose population of 1050 million accounts for 81% of the 1293 million people living in the South-eastern Asia region (Gunnell et al., 2007).

4.1. Risk of residual pesticides

Pesticide poisoning, whether intentional and occupational or not, is an important health issue because pesticides are used in a large number of industries, putting many categories of the workforce at risk. The chance of pesticide exposure through occupational source is higher in India than in most industrialized nations because of the low level of protection to workers. Even though proper regulations are available, several shortcomings in their enforcement and inadequate resources, together with limited infrastructure, put workers at high risk of pesticide exposure. Further, inadequate availability of protective equipment; improper application conditions and practices (e.g. lack of suitable washing facilities for workers, routine application of highly toxic pesticides by untrained workers, or lengthy application hours); unsafe storage and disposal facilities; poor management of hazardous pesticides; untrained pesticide dealers; use of faulty equipment, and the poor calibration and maintenance thereof; inadequate health centers, medical facilities, and lack of trained health personnel may all elevate the health risk to workers.

5. Preventative measures

The implementation of appropriate preventive measures helps to minimize cases of pesticide poisoning and other health risks linked to pesticide use. The government of India is concerned about the health risks of pesticides and has implemented various measures, such integrated pest management (IPM), prohibition of highly hazardous pesticides, restricting the use of toxic compounds, and the development of a national implementation plan (NIP).

5.1. Integrated pest management (IPM)

Development of pest resistance, pest resurgence, the outbreak of secondary pests, and the presence of pesticide residues in food, fodder, soil, air and water ultimately leads to human health hazards and ecological imbalances. Looking into the severe side-effects of chemical pesticides, the Indian government has proposed and adopted an IPM approach, strengthening and modernizing pest management approaches. IPM is an eco-friendly method for managing pests without greatly affecting the health of man or the environment. It utilizes cultural, mechanical and biological methods and need-based use of chemical pesticides, preferably bio-pesticides, bio-control agents and indigenous innovation potential.

Development of the IPM approach was initially introduced in cotton and rice crop by Swaminathan (1975) under the operational research project of the Indian Council of Agricultural Research (ICAR), which helped develop location-specific IPM technologies for the two crops. Since 1981, the Indian government has adopted IPM as the dominant principle of plant protection, to reduce dependency on chemical control. The Commonwealth Agricultural Bureau International (CABI) in the UK has supported the IPM regional program in cotton since 1993. A significant governmental investment in the cotton sector was made through the Technology Mission on Cotton (TMC) launched in 2000. The TMC plan was initiated by the government of India to increase cotton production and improve the quality of cotton, thereby increasing the income of cotton growers. In 2002, the Central Institute of Cotton Research (CICR) in Nagpur (West India) launched the IRM program in ten cotton growing states of India, targeting 26 districts, which were consuming 80% of insecticides for cotton crops. IRM programs were implemented by the State Agriculture Universities (SAU) (Kranthi and Russell, 2009; Russell, 2004). The FAO of the United Nations has been instrumental in the development of these ecological approaches for crop management. Subsequently, the Indian government has implemented a number of IPM regional programs in three phases. In the first phase emphasis is given on authentication of IPM technology and the expansion of pilot extension activities with farmers. The second phase deals with human resource development and the introduction of a fulltime farmer training program known as ‘IPM FFS’. This approach has proven to be very popular amongst Indian farmers. The last phase emphasizes the institutionalization of the IPM approach into government structures.

At present, 31 central IPM centers are operating across India. IPM performs several tasks, such as the monitoring of pests and diseases, production and release of bio-control agents and bio-pesticides, and conservation of bio-control agents. It also imparts training to agriculture/horticulture extension officers and farmers at the grass root level by organizing farmers' field schools (FFS). The IPM package for pest and disease management in 77 major crops has been developed and implemented in all Indian states, in consultation with the State Department of Agriculture, Horticulture, ICAR Institutions and SAU. The government of India has provided grants to state-level government offices for the establishment of 29 state bio-control laboratories across India.

5.2. Banned and restricted pesticides

The Indian government banned the production, import and use of 25 pesticides of the total 230 currently registered. Two pesticide formulations were banned for their use but allowed for export, while another four pesticide formulations were banned for import, manufacturing and use, and eight pesticides were withdrawn.

5.3. National implementation plan

As stated in article 7 of the Stockholm Convention on persistent organic pollutants (SCPOPs) every nation party to the convention is required to develop a NIP for SCPOPs to demonstrate how the obligations of the convention would be implemented. Fulfilling this requirement is another preventive measure instituted by the Indian government to reduce the exposure of pesticides. Based on the recommendation of the NIP development team, the government of India has identified the following priorities for the successful implementation of the NIP:

- Non-persistent organic pesticide alternatives to DDT should be developed and encouraged
- Separate mechanisms should be designed to deal with new persistent organic pesticides
- Best available techniques (BAT) and best environmental practice (BEP) approaches should be implemented for the elimination and reduction of unintentional persistent organic pesticide emissions from industry as identified in the NIP
- Production and demonstration of neem based bio-pesticides should be encouraged
- Persistent organic pesticide contaminated sites should be identified and potential hotspots be restored
- Raised awareness about persistent organic pesticide and pesticide management in India
- A national persistent organic pesticides monitoring program should be implemented throughout India
- For effective and efficient implementation of NIP, institutions involved in pesticides research and capacity building should be strengthened.

6. Government of India's efforts to eliminate persistent organic pesticides

The international community has been greatly concerned about the health risks associated with persistent organic pesticides and have sought mutual cooperation and action from all nations to reduce and eliminate the production, use and release of persistent organic pesticides. To date, six international legally binding instruments have been negotiated and concluded. India has signed and ratified three of them (Table 5). The executive body of the UNECE Convention on Long-Range Trans boundary Air Pollution (CLRTAP) adopted the UNECE protocol on POPs in June 1998 and this came into force on 23 October 2003. As of February 2007, the convention has a total of 51 supporting parties, though India has yet to become one of them.

The SCPOPs is the most important legally binding international agreement to protect human health and the environment from some of the most dangerous chemical on earth. The SCPOP highlights the prohibition or severe restriction of the production and use of intentionally produced POPs; restrictions on the export and import of intentionally produced POPs; provisions on the safe handling of stockpiles; procedures for the proper and safe disposal of waste containing POPs; and guidelines for the reduction of emissions from unintentional POPs (e.g. dioxins and furans).

Since 1998, India has been actively involved in various negotiations regarding the SCPOPs. India has set up a National Steering Committee (NSC) under the Ministry of Environment and Forests (MOEF) to facilitate its full participation in the Intergovernmental Negotiating Committee (INC) for implementing international action on persistent organic pesticides. The NSC has been formed to harmonize the interests and standpoints of different ministries, state administrations and commissions, and also to determine the position of the Indian government on persistent organic pesticides. Further, the NSC is also responsible for planning, guidance and monitoring of all actions needed for compliance with the provisions of the SCPOP. India has shown its desire to play a full role as a member of the Stockholm Convention by signing the SCPOP on the 14th May 2002, which was subsequently ratified on the 13th January 2006. As a signatory, India has demonstrated an understanding that compliance with the obligations set out in the convention will have a significant and positive influence, not only on India's own chemical management regime, but also on the ultimate global success of the convention's aim to protect human health and the environment. Moreover, successful efforts in moving towards compliance will also serve as a model for other developing countries.

7. Regulatory system for controlling persistent organic pesticides in India

Though the use of pesticides is necessary to protect agricultural crops from pests and to control vector borne disease, the regulatory system has had to take into consideration its residues in food and water. In India, a number of government agencies are involved in the regulation of pesticides. The Ministry of Agriculture regulates the manufacture, sale, transport and distribution, export, import, and use of pesticides through the 'Insecticides Act 1968'. CIBRC directs the central and state governments on technical matters. The formulation and use of new pesticides to control pests in various crops is approved by the Registration Committee (RC), while the Union Ministry of Health and Family Welfare monitors and regulates pesticide residue levels in food. It also sets MRLs for pesticides in food commodities. Likewise, pesticide regulation in India is governed through various rules and acts (Table S.3).

Table 5
International conventions on POPs and India's status.

International instruments	Parties	Rationale	India's status	Adopted	Ratified
Virtual Elimination of Persistent Toxic Substances in the Great Lakes	Canada, United States	To promote emissions reductions of toxic substances	–	–	–
North American Agreement on Environmental Cooperation (NAAEC)	United States, Canada, and Mexico	To develop regional initiative on the sound management of chemicals	–	–	–
Basel Convention on the Transboundary Movement of Hazardous Waste and the Subsequent 1995 Ban Amendment (Basel Ban),	163 nations	Import and export of hazardous waste	Signed and ratified	1989	1992
United Nations Economic Commission for Europe (UNECE) on POPs under the Convention on Long-Range Transboundary Air Pollution (LRTAP)	United States, European countries, Canada, and Russia	Elimination of production and reduction of emissions of POPs in the UNECE region	–	–	–
Rotterdam Convention on the Prior Informed Consent (PIC) Procedure for Certain Hazardous Chemicals and Pesticides in International Trade	United States, along with 71 other countries	Import and export of hazardous chemicals	Signed and ratified	2002	2004
Stockholm Convention on Persistent Organic Pollutant	149 countries	Production, use and disposal of persistent organic pollutants	Signed and ratified	2002	2006

States and UT authorities are empowered to grant licenses for the manufacturing and sale of insecticides. Additionally, state officials assist the CIBRC in conducting analysis (including MRLs of the pesticide) to report and enforce on public safety matters. The CIBRC periodically reviews the use of pesticides and recommends bans on registration if it is considered necessary. It is clear from Table S.3 that despite the numerous rules and acts instituted in India for regulating pesticides, these have failed to end pesticide contamination in water, air and soil. India needs a strong bill that will control and regulate the use of pesticides and lead to a lower impact on human health and the environment. Looking into the problem of persistent organic pesticide management, the Indian government has introduced 'The Pesticide Management Bill 2008', approved on April 24, 2008. The purpose of bill is to introduce new, safe and effective pesticides, and to improve the quality of existing pesticides. The bill establishes more effective regulation on the import, manufacture, export, sale, transportation, distribution and use of pesticides, to curb the risk to human beings, animals, or environment. Additionally, the bill provides strict action against offenders and maximum punishments for greater deterrence to violators. However, the bill is still pending for ratification in the Indian Parliament, though once it is passed it will become law. The bill will replace the existing Insecticide Act 1968 and Insecticides Rule 1971.

8. Concluding remarks

Many studies have focused on the monitoring and analysis of persistent organic pesticides, especially OCPs and Ops, showing that every environmental area (air, water and soil) in India has been contaminated by persistent organic pesticides; its residues are found above MRL levels in air, water and soil across the nation. The majority of studies reviewed suggest that residue levels of pesticides in the Indian air, water and soil are high, despite the present low consumption in India. This may be because many chemical pesticides were extensively used in the past, before most of them were banned. Another reason for the presence of residue levels in the Indian environment is due to the incorrect dosage of pesticides during application, leaving behind an excess, off-target quantity. Use of the correct spraying machines would prove more economical and minimize this wastage. Farmers should also be made aware, through education and training programs, to use certified sprayers and nozzles.

Analysis of past data showed an increasing trend of pesticide use in India between 1958 and 2004 (from 5000 to 85,000 MT), however its production remained constant thereafter (2004–10). Hence there is need to sensitize and educate the general population, especially end-users, on the sound management of pesticides. These practices lead to potential health risks amongst the general population. The introduction of courses on persistent organic pesticides in the curriculum of Indian colleges and universities is also recommended. More environmentally friendly and economical approaches, such as IPM and BIMP, should be adopted and encouraged. Likewise, information regarding the correct application of pesticides and the use of advanced technologies for target delivery of pesticide, as well as intensive training on selective application of the correct pesticides at the correct time for the correct pests, should be disseminated to the user group. Plant health clinics to offer guidance to farmers could be established. Development of a medical surveillance program and protocols for the management of pesticide exposure, as well as facilities for investigating cases of poisoning in leading hospitals, will help in the reduction of health risks from persistent organic pesticides.

Several modeling simulation studies, mostly carried out in the TP of southwest China, suggest the prominent role of persistent organic pesticide emissions from Indian sources in harming environment. Further studies have revealed that India is one of the major contributors of persistent organic pesticides on the TP, leading to POP contamination on a global scale. India, therefore, needs to enact an umbrella policy and strictly enforce the legislation addressing all aspects of pesticide control. Any policy must support the newer concepts of IPM, with eco-friendly and multi-disciplinary approaches to pest control and the minimal use of pesticides.

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Appendix A. Supplementary data

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