

Occurrence of Congener Specific Polychlorinated Biphenyls in Soils from Roadside Agricultural Fields

Bhupander Kumar^{*}, V. K. Verma, S. K. Singh, Sanjay Kumar and C. S. Sharma

National Reference Trace Organics Laboratory, Central Pollution Control Board, East Arjun Nagar, Delhi, India

ABSTRACT

Twenty eight polychlorinated biphenyls (PCBs) congeners including twelve dioxin-like PCBs were measured in soils from roadside agricultural lands of National Capital Region (NCR), India. The concentration of Σ PCBs ranged between <0.01 - 38.32 ng g^{-1} (dw) with the mean of $8.40 \pm 1.27 \text{ ng g}^{-1}$ (dw), where, Σ 12dl-PCBs ranged between <0.01 - 13.59 ng g^{-1} (dw) and their average value was $2.86 \pm 0.46 \text{ ng g}^{-1}$ (dw). Group homolog of total 28 PCBs was dominated by tetra-chlorinated biphenyls (4-CBs) and hexa-chlorinated biphenyls (6-CBs) which accounted for 56% and 23.6%, respectively. Toxic equivalent (TEQ) of dl-PCBs was between <0.01 - $140.24 \text{ pg WHO}_{2005}\text{-TEQ g}^{-1}$ (dw) with a mean of $31.86 \pm 5.25 \text{ pg WHO}_{2005}\text{-TEQ g}^{-1}$ (dw). PCBs contaminations in studied soils of agricultural areas were lower than soil quality guideline values.

Keywords: polychlorinated biphenyls (PCBs), PCB Toxic Equivalent, soil.

INTRODUCTION

Polychlorinated biphenyls (PCBs) are long range air transport (LRAT) pollutants and ubiquitously found worldwide, regions far from their original sources [1-2]. The properties of hydrophobicity and resistance to degradation make them to accumulate in soil, sediments and biota [3]. PCB compounds have been recognized for wide range of human health effects and are classified as probable human carcinogens. Although, atmospheric air is the primary medium of global transport for persistent organic pollutants but soil and sediment acts as important reservoirs for them globally, vegetation acts as scavenging medium and as a major vector of organic pollutants into terrestrial food chain [4-5].

As a party to the Stockholm Convention, India is obligated to abide by the objectives of the treaty with the intention of reducing, and ultimately eliminating these pollutants, and encouraged to support research on POPs. Recent studies carried out in India, and PCBs contaminations has been reported for different matrices, includes water [6], soils [7-8], sediments [9-11], atmospheric air [12] and human milk [13-14]. In continuation of support research on POPs in India, this study was aimed to evaluate polychlorinated biphenyls (PCBs) including dioxin like PCBs (dl-PCBs) concentrations in soils from roadside agricultural fields of three north Indian states in the vicinity of National Capital Region of India.

MATERIALS AND METHODS

Study Area and Sampling

National Capital Region (NCR), India comprises by the National Capital Territory (NCT) Delhi and delineated area of the surrounding states of Haryana, Uttar Pradesh, and Rajasthan with the population of 22.157 million. The entire

National Capital Region spread over an area of around 30,240 km². The cultivated land is accounted a large proportion of 79.53 per cent area in the NCR, Delhi region [15], where wheat, mustard, sugarcane, maize, millets, sorghum, cereals, paddy and commercial agricultural crops, such as, vegetables, flowers, mushroom etc are the main crops grown regularly.

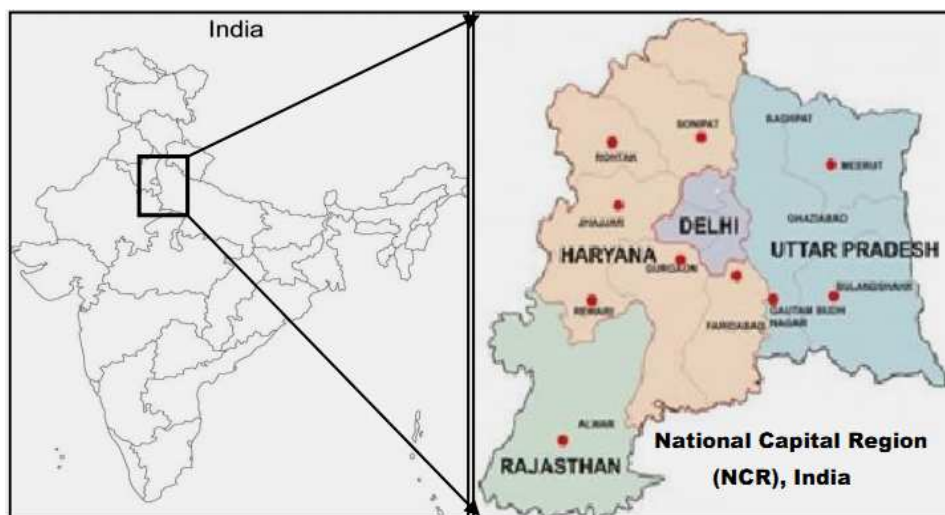


Figure 1: Map showing study area in National Capital Region, India

Sampling locations were selected in agriculture lands near main roads of Delhi, Uttar Pradesh and Haryana in NCR (Figure 1). A total number of 83 samples collected during November 2011-January 2012. Approximately 500 grams of soil in duplicate was collected, pebbles and wood sticks removed manually and mixed thoroughly to ensure that the soil collected was truly representative of each location. Then, aliquot of the soil was taken and stored in clean wide mouth amber glass bottle which were then transported to laboratory and kept at -4°C until further extraction.

PCBs Extraction and Cleanup

EPA Method 3550C [16] was followed for sample extraction. Briefly, 15-20 g wet sample was mixed with cleaned anhydrous sodium sulfate until a free-flowing powder was obtained. Activated copper was used to remove the sulfur. The sample was then extracted in 3 cycles (50, 25, 25 ml) with hexane/acetone (1/1v/v) solvent mixture by ultrasonication for 30 min each cycle. The extract was concentrated using Rotatory Vacuum evaporator (Eyela, Japan) for further cleanup.

The extract clean-up was performed with multilayered silica gel column chromatography on a tri-functional column with neutral, basic and acid silica to remove interfering organic and polar compounds. Briefly, multilayered silica gel packed in glass column (300 mm x 30 mm) from bottom to up with 2.5 g silica gel, 4.0 g silver nitrate silica gel, 2.5 silica gel, 4.0 basic silica gel, 2.5 g silica gel, 12.0 g acid silica and on top 5.0 g anhydrous sodium sulphate was added. 100 ml hexane was used for column rinse before sample loading. After loading the sample extract with three rinsing pollutants were eluted with hexane. The eluted extract was concentrated under gentle stream of purified nitrogen gas using TurboVap (Caliper, USA) to 1.0 ml and transferred to vial for quantification with gas chromatography equipped with an electron capture detector (GC-ECD).

PCBs Quantification

The separation and quantification of polychlorinated biphenyls (PCBs) was performed on Shimadzu 2010 gas chromatography (Japan) attached with autosampler and equipped with an ECD (⁶³Ni), on HP-5MS capillary column (60 m x 0.25 mm x 0.25 µm film, Agilent). The temperature program of the column oven was initially maintained at 170°C for 1 min and increased with 3°C min⁻¹ to 270°C, where it temperature was hold for 1 min, then further ramped with 10°C min⁻¹ to 290°C at and kept for 3 min. The injector and detector temperature were maintained at 225°C and 300°C respectively. Laboratory grade nitrogen gas was used as carrier @ 1.0 ml. min⁻¹.

Analytical Quality Control

Certified reference standards procured from Dr. Ehrenstorfer (GmbH, Germany) and used for the instrument calibration and quantification of individual PCB congeners. The PCB congeners were identified in the sample extract by comparing the accurate retention time from the standard and quantified using the response factors from five level calibration curves of the standards (r^2 value, 0.999). Adequate quality control was performed, including analysis of procedural blanks alongwith real samples (concentrations were <MDL 'method detection limit'), random duplicate samples and calibration verification analysis (Standard deviation <5). Matrix spike recovery study was undertaken and percent recovery was 100±15. Each sample was analysed in duplicate and the average was used in calculations. Toxic equivalent quantities (TEQ) were calculated by multiplying the concentration of individual dl-PCB congener with the corresponding 2, 3, 7, 8-TCDD substituted toxicity equivalent factors (TEFs) [17].

Moisture content was determined gravimetrically to report results as dry weight. The results of PCBs in soil were reported in ng g⁻¹ and their toxic equivalency as pg WHO₂₀₀₅-TEQ g⁻¹ dry-weight (dw). Concentrations above reporting limit (> 0.01 ng g⁻¹) was taken for calculation and concentrations below reporting limit or below MDL (<0.01 ng g⁻¹ dw) were not included in the calculations.

RESULTS AND DISCUSSION

Distribution of PCBs in Soil

The observed concentrations of PCBs in soils of roadside agricultural lands are presented in Table 1. The total concentration of PCBs were range between <0.01 – 38.32 ng g⁻¹ (dw) with the mean of 8.40±1.27 ng g⁻¹ (dw). The concentration of PCBs in soils from Uttar Pradesh was comparatively higher than soils from Delhi and Haryana states. The average concentration of PCBs in soils from Delhi, Uttar Pradesh and Haryana was 0.86±0.15 ng g⁻¹, 20.77±1.82 ng g⁻¹ and 3.97±3.33 ng g⁻¹ with the range of <0.01-4.69 ng g⁻¹, 0.26-38.32 ng g⁻¹ and <0.01-30.58 ng g⁻¹, respectively. The PCBs contamination levels in soils from National Capital Region (NCR) were lower than soil quality guideline value of 500 ng g⁻¹ [18] (CCME, 1999). Earlier studies reported significant levels in of PCBs in different environmental matrices from Delhi and adjoining areas [6,8,10-12]. Results of this study for PCBs in soils were in agreement with the observations by other workers [1,19-20]. However, the concentrations of PCBs from this study were higher than those PCBs reported in soils from China [21], Turkey [22]. Comparatively higher concentrations of PCBs in agricultural soil have been reported in literature from Estonia [23], Romania [24], China [25-27] and Mexico [28].

Table 1: ΣPCBs and Σdl-PCBs (ng/g) in Roadside Agricultural Soils from NCR, India

Study area (n)*	PCBs	Range	Mean	SE*	%
Delhi (44)	PCBs	<0.01-4.30	0.67	0.14	77.86
	dl-PCBs	<0.01-0.66	0.19	0.03	22.14
	ΣPCBs	<0.01-4.69	0.86	0.15	100
Uttar Pradesh (30)	PCBs	0.26-24.72	13.5	1.21	64.98
	dl-PCBs	<0.01-13.59	7.27	0.70	35.02
	ΣPCBs	0.26-38.32	20.77	1.82	100
Haryana (9)	PCBs	<0.01-22.48	2.80	2.46	70.41
	dl-PCBs	<0.01-8.09	1.18	0.87	29.59
	ΣPCBs	<0.01-30.58	3.97	3.33	100
All study area (83)	PCBs	<0.01-24.72	5.54	0.84	65.96
	dl-PCBs	<0.01-13.59	2.86	0.46	34.04
	ΣPCBs	<0.01-38.32	8.40	1.27	100

Note: <0.01=below detection limit, *n=number of samples, **SE=standard error (SD/√n)

Congener Profile and Group Homolog of PCBs

The individual PCB congener and their group homolog in soils from roadside agricultural fields of NCR, India was presented in Table 2 and Table 3, respectively. Among the 28 PCB congeners the PCB-44 was the dominant congener (1.58±0.27 ng g⁻¹), followed by PCB-49 (1.19±0.20 ng g⁻¹), PCB-81 (0.77±0.17 ng g⁻¹), PCB-151 (0.62±0.11 ng g⁻¹), PCB-156 (0.47±0.09 ng g⁻¹), PCB-77 (0.45±0.11 ng g⁻¹), and PCB-74 (0.44±0.12 ng g⁻¹), other congener concentration were comparatively low (<0.01 to 0.30 ng g⁻¹). PCBs were generally used as technical mixtures and 70% of PCBs produced globally were trichlorinated to tetrachlorinated biphenyls [29].

Table 2: PCBs congeners (ng g⁻¹) in Roadside Agricultural Soils from NCR, India

Congeners	Range	Mean	SE*	%
PCBs				
PCB - 18	<0.01-2.42	0.31	0.06	3.65
PCB - 37	<0.01-6.59	0.17	0.08	2.03
PCB - 44	<0.01-9.38	1.58	0.27	18.8
PCB - 49	<0.01-5.81	1.19	0.20	14.12
PCB - 52	<0.01-3.65	0.11	0.05	1.31
PCB - 70	<0.01-3.80	0.17	0.06	2.03
PCB - 74	<0.01-4.22	0.44	0.12	5.27
PCB - 119	<0.01-3.15	0.07	0.05	0.87
PCB - 128	<0.01			
PCB - 138	<0.01-1.42	0.27	0.05	3.18
PCB - 151	<0.01-3.36	0.62	0.11	7.33
PCB - 168	<0.01-2.49	0.15	0.05	1.81
PCB - 170	<0.01-1.50	0.08	0.03	1.01
PCB - 177	<0.01-1.75	0.11	0.04	1.26
PCB - 187	<0.01			
PCB - 207	<0.01-3.19	0.28	0.06	3.29
dl-PCBs				
dl-PCB - 77	<0.01-4.26	0.45	0.11	5.40
dl-PCB - 81	<0.01-6.97	0.77	0.17	9.12
dl-PCB - 126	<0.01-1.20	0.26	0.04	3.14
dl-PCB - 169	<0.01-1.04	0.17	0.03	2.05
dl-PCB - 105	<0.01-1.31	0.09	0.03	1.02
dl-PCB - 114	<0.01-0.91	0.05	0.02	0.65
dl-PCB - 118	<0.01-1.90	0.19	0.04	2.27
dl-PCB - 123	<0.01-2.71	0.08	0.04	1.00
dl-PCB - 156	<0.01-3.33	0.47	0.09	5.56
dl-PCB - 157	<0.01-0.04	<0.01	<0.01	<0.01
dl-PCB - 167	<0.01-1.49	0.30	0.05	3.62
dl-PCB - 189	<0.01-0.49	0.02	0.01	0.20

Note: <0.01=below detection limit, *standard error=SD/√n

The group homolog of PCBs in soils from NCR was dominated by tetra and hexa chlorinated biphenyls. Tetra-CB (56%) homolog was the main contributors to the total PCB homolog in average followed by hexa-CBs (23.6%) (Figure 2). The pattern of homolog was observed as tetra-CBs > hexa-CBs > penta-CBs > tri-CBs > hepta-CBs. Table 3 shows that the percentage (average >60% for tri- to tetra-PCBs) of lighter-weighted molecular PCBs (LWM-PCBs) were higher than those higher-molecular weight PCBs (HMW-PCBs) (penta to hepta-CBs). It is reported that LMW-PCBs were primarily used in power capacitors and transformers, while HMW-PCBs were mainly used as an additive [30]. This indicates that PCBs used in heat transfer equipments and as additives found their way to the environment of national capital region.

Table 3: PCB group homolog (ng g⁻¹) in Roadside Agricultural Soils from NCR, India

PCB Group Homolog	Delhi		Uttar Pradesh		Haryana		All study area	
	Range (mean±se)	%	Range (mean±se)	%	Range (mean±se)	%	Range (mean±se)	%
3-CBs	<0.01-1.85 (0.20±0.10)	22.5	<0.01-2.42 (0.75±0.12)	3.6	<0.01-7.78 (0.96±0.85)	24.1	<0.01-7.78 (0.48±0.11)	5.7
4-CBs	<0.01-4.33 (0.51±0.12)	60.9	<0.01-21.02 (11.83±1.04)	56.9	<0.01-10.73 (1.39±1.17)	35.0	<0.01-21.02 (4.70±0.71)	56.0
5-CBs	<0.01-0.60 (0.13±<0.01)	9.0	<0.01-4.99 (1.84±0.23)	8.9	<0.01-1.34 (0.40±0.15)	10.1	<0.01-4.99 (0.75±0.12)	9.0
6-CBs	<0.01-0.21 (<0.01)	3.1	<0.01-9.43 (5.17±0.52)	24.9	<0.01-7.81 (0.89±0.56)	22.4	<0.01-9.43 (1.98±0.34)	23.6
7-CBs	<0.01-0.10 (<0.01)	0.6	<0.01-2.21 (0.50±0.12)	2.4	<0.01-1.98 (0.22±0.22)	5.7	<0.01-2.21 (0.21±0.05)	2.5

<0.01=below detection limit, se=standard error (SD/√n)

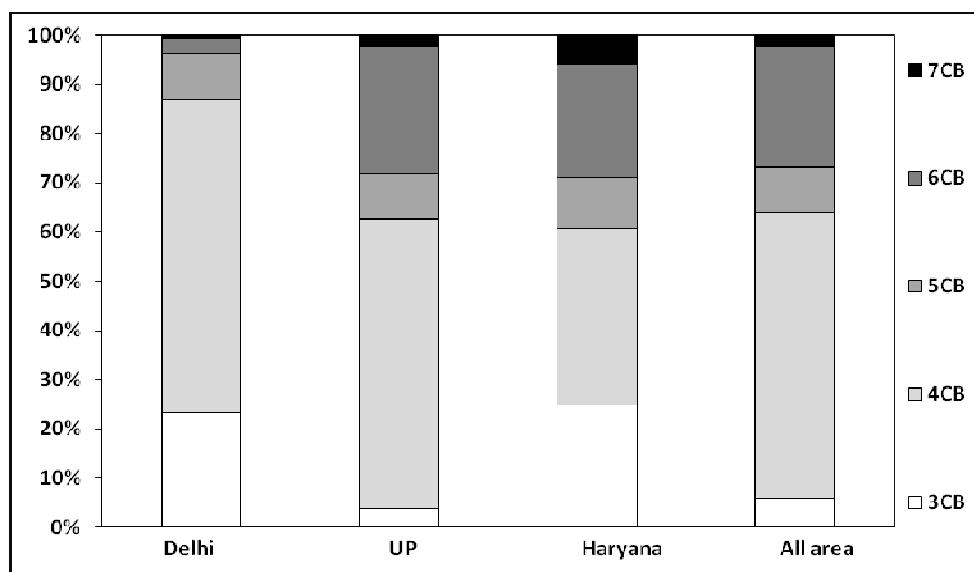


Figure 2: Percent of PCB homolog in Roadside Agricultural Soils from NCR, India

Toxic Equivalency of dl-PCBs

The concentration of \sum dl-PCBs in this study was ranged between <0.01 - 13.59 ng g⁻¹ with an average of 2.86 ± 0.46 ng g⁻¹, and quantity of 12 dl-PCBs accounts for 34.04% of total 28 PCBs. DL-PCBs have toxic responses similar to those caused by 2, 3, 7, 8-tetra-chlorodibenzo-*p*-dioxin (TCDD) the most potent congener within the dioxins/furans group of compounds [31]. As a result the concept of toxic equivalency factors (TEFs) has been developed to assess the impact of these compounds on human health and concentrations of dl-PCB congener can be converted into 2, 3, 7, 8-TCDD substituted TEQ concentrations.

Table 4: Toxic Equivalent of dl-PCBs (range and mean) (pg WHO-TEQ g⁻¹) in Roadside Agricultural Soils from NCR, India

Study area (N)*	dl-PCBs	Range	Mean	SE	%
Delhi (44)	non ortho	<0.01-0.18	0.03	<0.01	92.7
	mono ortho	<0.01-0.02	<0.01	<0.01	7.1
	total	<0.01-0.18	0.03	0.01	100
Uttar Pradesh (30)	non ortho	<0.01-140.07	84.36	7.29	99.9
	mono ortho	<0.01-0.21	0.09	0.01	0.1
	total	<0.01-140.24	84.45	7.30	100
Haryana (9)	non ortho	<0.01-108.69	12.11	12.07	99.8
	mono ortho	<0.01-0.11	0.02	0.01	0.2
	total	<0.01-108.80	12.13	12.08	100
All study area (12)	non ortho	<0.01-140.07	31.82	5.25	99.9
	mono ortho	<0.01-0.21	0.04	0.01	0.1
	total	<0.01-140.24	31.86	5.25	100

*N=number of samples

TEQ concentrations of PCBs with established dioxin-like activity especially the non- and mono-ortho substituted PCBs in soils NCR India were calculated by multiplying the concentration of each dioxin-like PCB congener by its 2, 3, 7, 8-TCDD substituted TEF values (Toxic Equivalency Factors) for human and mammals. The toxicity equivalency (TEQ) for 12 dl-PCBs was presented in Table 4. Total TEQ levels ranged from <0.01 to 140.24 pg WHO₂₀₀₅-TEQ g⁻¹ with the mean of 31.86 ± 5.25 pg WHO₂₀₀₅-TEQ g⁻¹. The TEQ of non ortho-PCBs (CB-77, CB-81, CB-126 and CB-169) were higher and contributed 92-99% for total TEQ, while the TEQ of mono ortho PCBs (CB-105, CB-114, CB-118, CB-123, CB-156, CB-157, CB-167 and CB-189) were <1 for all the samples. CB-126 and CB-169 congeners represent the higher TEQ values which both had the high toxic potency (toxic equivalency factor proposed WHO-TEF=0.1 and 0.03 respectively) thus significantly increasing the \sum dl-PCBs with the contribution of 99% for \sum TEQ. Congener specific concentration of dl-PCBs in NCR were in agreement with the observations of

other workers [27]. The concentrations of Σ dl-PCBs in agricultural soils from NCR India were lower than Tailake region and Southern Jiangsu region of China [20-21], Pancevo, Serbia [32], Brescia city, Italy [5], but, higher than Yellow river delta, China [26], Tailke region, China [33]. The observed concentrations of 12dl-PCBs TEQ in soil samples studied in this study was lower than environmental quality standard (1000 pg WHO-TEQ g⁻¹) [34].

There is no reported data, that PCBs produced in India but technical mixture of PCBs have been used in industry and electrical appliances. It has been reported that around 9837 tons of PCBs exist with transformers in India [35]. Here it is suggested that PCBs sources to the environment may be from biomass burning, depositions of emissions from wood, paint and dying processing, chemicals and PVC (polyvinylchloride) manufacturing units and from electrical and electronic waste recycling. These PCB sources also include off gassing from closed system such as older equipments (e.g. transformers that contain large quantities of PCB fluids).

CONCLUSION

This study that PCBs contaminations in agricultural soils from north Indian states in the vicinity of NCR are not alarming, because observed levels are much lower than soil quality guideline values. Intensive study on bioaccumulation of PCBs including other persistent organic pollutants is recommended due to human health and environment concerns.

Acknowledgements

The authors express their sincere gratitude to the Member Secretary and Chairman of Central Pollution Control Board, Ministry of Environment & Forest Government of India for encouragement and guidance to conduct the study.

REFERENCES

- [1] Aichner B., B. Glaser, W. Zech, **2007**. *Organic Geochem.*, 38: 700-715.
- [2] Alison, L. S. & Jason, D. W. **2008**. *Rev. Biol. Trop. (Int. J. Trop. Biol.)*, 56 (4):1-9.
- [3] Baldassarri, L. T., Abate, V., Alivernini, S., Battistelli, C. L., Carasi, S., Casella, M., Iacovella, N., Iamiceli, A. L., Indelicato, A., Scarcella, C., Rocca, C. L. (**2007**). *Chemosphere*, 67:1822–1830
- [4] Binelli, A., Sarkar, S.K., Chatterjee, M., Riva, C., Parolini, M., Bhattacharya, B.D., Bhattacharya, A.K., Satpathy, K.K. (**2009**), *Environ. Moni. Assess.*, **153** (1-4), 221-234
- [5] Breivik K., Sweetman A., Pacyna J., Jones K. **2002**. *Science of Total Environ.* 290: 181-198.
- [6] CCME (Canadian Council of Ministers for the Environment). **1999**; Canadian Soil Quality guidelines for the Protection of Environmental and Human Health, Polychlorinated Biphenyls (Total). Health branch, Ottawa, Canada
- [7] Devanathan, G., A. Subramanian, M. Someya, A. Sundaryanto, T. Isobe, S. Takahashi, P. Chakraborty and S. Tanabe, (**2008**), *Environ. Poll.* 157 (1): 148-154.
- [8] Doina, D., Simona, C., Alin, C. D., Raluca, M., Luc Van Vaeck, A. C. **2006**. *Int. J. Environ. Anal. Chem.* 86 (11): 833-842.
- [9] Giesy, J. P. and Kannan, K. (**1998**). *Critical Reviews in Toxicology*, 28 (6):511–56.
- [10] Hyun Park, S. H. Lee, M. Kim, J. H. Kim and H. S. Lim. **2010**. *Antarctic Science*, 22 (1): 31-38.
- [11] Kaisarevic S., K. Hilscherova, R. Weber, K. L. Sundqvist, E. Voncina, S. Bobic, A. K. Pogrmic, M. V. Miloradov, J. P. Giesy, R. Kovacevic. **2011**. *Environ. Sci. Pollut. Res.* 18 (4): 677-686
- [12] Kumar, B., Kumar, S., Gaur, R., Goel, G., Mishra, M., Singh, S. K., Dev Prakash and Sharma, C. S. (**2011**)^A, *Soil & Water Research* **6** (4), 190–197
- [13] Kumar, B., Mishra, M., Goel, G., Gaur, R., Singh, S. K., Prakash, D., Kumar, S. and Sharma, C. S. (**2011**)^B, *Adv. Life Sci. Technol.* **1**: 1-8.
- [14] Kumar, B., Singh, S. K., Kumar, S. and Sharma, C. S. (**2012**)^A, *J. Natural Sci. Research* **2** (1):26-37.
- [15] Kumar, B., Gargi Goel, Richa Gaur, S. K. Singh, Sanjay Kumar and C. S. Sharma (**2012**)^B, *Adv. Appl. Sci. Research*, **3** (2):1012-1019.
- [16] Liu J., Z. J. Cui, H. Y. Xu, F. X. Tan. **2009**. *Soil Sediment Cont.: An Int. Journal.* 18 (2): 144-154.
- [17] Minh N. H., Minh T. B., Kajiwarra N., Kunisue T., Subramanian A., Iwata H., Tana T. S., Baburajendran R., Karupiah S., Viet P. H., Tuyen B. C. and Tanabe S. **2006**. *Arch. Environ. Conta. Toxicol.*, 50: 474-481.
- [18] MoE, Japan. (**2009**), Brochure Dioxin: www.env.go.jp/en/chemi/dioxins/brochure2009.
- [19] NCRPB (National Capital Region Planning Board), Delhi. **2012**. Accessed on 24th Feb **2012**: ncrpb.nic.in

- [20] Rogelio C. S., T. A. Antonio, R. A. Diana, G. R. Octavio, D.B. Fernando, P.M. Ivan. **2011**. *Bull. Environ. Conta. Toxicol.*, 86 (2):212-216.
- [21] Salihoglu G, Salihoglu NK, Aksoy E, Tasdemir Y. **2011**. *J. Environ. Manage.*, 92(3):724-32.
- [22] Senthil Kumar, K., Priya, M., Sajwan, K. S., Killic, R. and Roots, O. (**2009**). *Estonian J. Earth Sci.*, 58 (2):109-123.
- [23] Shen, C., Chen, Y., Huang, S., Wang, Z., Yu, C., Qiao, M., Xu, Y., Setty, K., Zhang, J., Zhu, Y., Lin, Q. (**2009**). *Environment International*, 35:50–55.
- [24] Someya, M., Ohtake, M., Kunisue, T., Subramanian, A., Takahashi, S., Chakraborty, P., Ramachandran, R. Tanabe, S. (**2009**). *Environment International* **36** (1): 27-35.
- [25] SC (Stockholm Convention), (**2012**), <http://chm.pops.int/Implementation/NIPs>.
- [26] USEPA, **2007**. Method 3550C, Ultrasonic Extraction. EPA SW-846, U.S. Government Printing Office, Washington, DC.
- [27] Van den Berg M., Linda S. Birnbaum, Michael Denison, Mike De Vito, William Farland, Mark Feeley, Heidelore Fiedler, et al. **2006**. *Toxicol. Sci.* 93 (2): 223-241.
- [28] Wang, D.G., Meng, Y., Jia, H.L., Lei, Z. Li, Y.F. **2008**. *Chemosphere*, 73 (1): 38-42.
- [29] Wang P., Zhang, Q., Wang, Y., Wang, T., Li, X., Li, Y., Ding, L., Jiang, G. **2009**. *Chemosphere*, 76:1498-1504.
- [30] Wang, T., Wang, Y., Fu, J., Wang, P., Li, Y., Zhang, Q., Jiang, J. (**2010**)^A. *Chemosphere*, 81:1045–1051.
- [31] Wang, H., Quiong, A., Dong, Y., De Chen Li, and B. Velde, **2010**^B. *J. Hazardous Materials*, 176 (1-3): 1027-1031.
- [32] You, H., Ding, J., Zhao, X. S., Li, Y. F., Liu, L. Y., Ma, W. L., Hong, Q. H., Shen, J. M. (**2011**), *Environ. Geochem. Health*, **33**, 291–299.
- [33] Zhao X., M. Zheng, B. Zhang, Q. Zhang, W. Liu (**2006**). *Science of Total Environment*, 368: 744–752.
- [34] Zhang, G., Chakraborty, P., Li, J., Sampathkumar, P., Balasubramanian, T., Kathiresan, K., Takahashi, S., Subramanian, A., Tanabe, S. and Jones, K. C. **2008**. *Environ. Sci. Technol.*, 42 (22): 8218-8223.
- [35] Zhang, J., Qui, L., He, J., Liao, Y. and Luo, Y. **2007**. *J. Environ. Sci.*, 19 (3): 338-342.