Quantification of PBDEs and PCBs in indoor dust collects (soils) from E-waste industry and End of Life vehicle (ELV) at major cities in South India

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1. Aim

Pollution by polybrominated diphenyl ethers (PBDEs), the popular brominated flame retardants (BFRs) incorporated into commercial materials, is a worldwide environmental problem. Polychlorinated biphenyls (PCBs) were once widely deployed as dielectric and coolant fluids in electrical apparatus, carbonless copy paper and in heat transfer fluids. PBDEs and PCBs are Persistent Organic Pollutants (POPs) widely used in electronics, polymers, and fabrics as BFRs. In the last two decades, they have attracted special environmental concern because of their persistence, bioaccumulative profile, and potential toxicity to humans and wildlife worldwide. PCDD/Fs, PBDEs, and PCBs have recently been detected in many environmental and human samples (air, soil, dust, sediment, freshwater, fish, cow milk, human milk, blood, umbilical cord blood) in the areas contaminated by E-waste. Three major PBDEs commercial mixtures have been commonly used: Penta-, octa-, and decaBDE. PentaBDE is primarily composed of BDE-47, -99, and -100, with smaller contribution of BDE-153, -154, and -85. OctaBDE contains 12 congeners (BDE-194 to BDE-205) whereas DecaBDE contains BDE-209, with minor nanoBDE and octaBDE congener impurities.

In developing countries, PBDEs and PCBs and their byproducts have been found to leach out from discarded electrical and electronic equipment (e-waste) dumping site, thus becoming pivotal issue for the international community. An important area of concern of PBDEs is their presence in e-waste and the challenges they posses for waste management particularly for end-of-life vehicles (ELVs).

The present study has revealed to determine and quantify PCBs and their biotransformation products in End of Life Vehicles soil samples from major cities of Tamil Nadu state in India. The current study was focused on the levels of PCBs in soils from ELV sites and their distribution based on the activity at particular site locations. Further, this study was noticed the levels of 56 PCB congeners, and their concentration range, maximum, and minimum level of PCBs at specific sites. This is the first report of PCBs level in End of Life Vehicle sites in India.

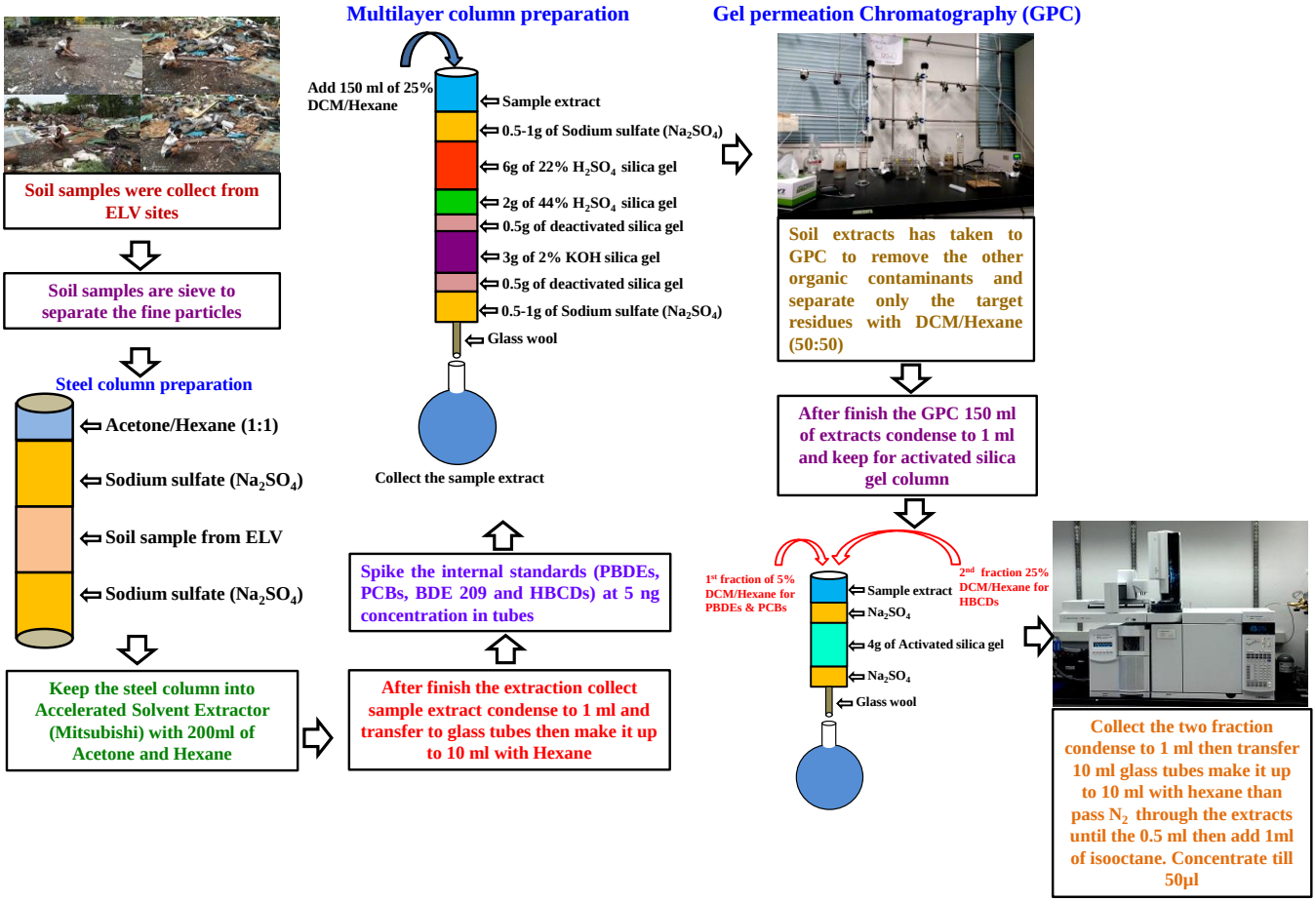
2. Objectives

✓To find the distribution and concentration of PCBs in End of Life Vehicles (ELV) in soil particles from various locations and also to find the hotspots of PCBs contaminated sites.

✓ To find out the distribution of PCBs from ELV soil samples and depends upon the sources than manmade activities.

✓ To assessof PCBs maximum and minimum levels in each sites at particular sites and find to discussed the background data on scraping of vehicles.

3. Methodology for PCBs extraction in soil samples



4. Result and Discussion

4.1 Level of PCBs in End of Life Vehicles (ELV) sites in Chennai and Coimbatore

Concentration of PCBs in soil from Chennai and Coimbatore were given in Table 1. PCBs concentration were observed with large range from ND(22.9) to -1258 ng/g dw with mean concentration of 2.2 – 88.9 ng/g dw,respectively (Table.1). Higher PCB concentration was quantified at CN3, where truck engines were dismantled. PCBs were mostly used in capacitors, electromagnets and miscellaneous electrical equipment for various purpose (Table.2). On the other hand, dielectric fluids, hydrulic fluids, break fluid,ignition coils and lubricants were used to avoid friction in engine. All the above mentioned components play a major role in the release of PCBs. According to EIP Associates, (1997) PCBs usage was 60% for closed system and heat transfer fluids like transformers, capacitors, fluorescent light ballasts. PCBs uasge in plastizers were 25%, hydraulic fluids and lubricants containing 10% and 5% for miscellaneous uses. This could be the possible source for higher PCBs concentration at CN3. The next higher total PCBs concentrtaion were quantified at CN1 ranged of 344 ng/g dw, where engines were dismantled and dumped at this site (Fig.1). Engine dismanding sites were observed with higher PCBs concentration due usage of various fluids and PCBs containing engine systems. The leaset concentration was observed at CN5 where the body of the vehiles were scrapped. The source of PCBs was lower at this site, coating materials are the source for PCBs which results in lower concentration.

Table.1 Concentration ranges, mean, maximum and minimum levels of PCBs in End of Life Vehicles (ELV) sites from India

Sampling Locations	Chennai					Coimbatore	
	CN1	CN2	CN3	CN4	CN5	MP1	MP2
Total $\sum_{56}$PCBs	344.3	520.6	4189.9	203.9	75.2	2.2	6.4
Concentration Range (ng/g dry wt)	ND - 73.8	ND - 73.1	ND - 1258	ND - 77.6	ND - 22.9	ND - 0.3	ND - 1.4
Mean	10.4	12.7	88.9	7.1	2.2	0.1	0.4
Maximum	73.8	73.1	1258.1	77.6	22.9	0.3	1.4
Minimum	0.0 3	0.1	0.1	0.1	0.1	0.0	0.1

The present study was compared to PCBs concentration in the floor dust samples from ELV-processing households in Thuyen village (Bac Giang, Vietnam) ranged from 19 to 2200 ng/g, which was comparitively higher than PCBs concentration in the soil samples from CN3 site (Takahashi et al.,2016). They suggesting ELV materials containing PCBs could be the source for PCBs in the floor dust. They also quantified PCBs from the indoor dust (Thuyen, Vietnames) from living areas (Median: 100ng/g) and

workshops (Median: 190 ng/g), which were an order of magnitude higher than those reported in house dust (Hanoi) (10 ng/g) and e-waste recycling sites (Hai Phong city (Median: 23 ng/g) and Hung Yen province (Median: 12 ng/g)), respectively. Contamination of PCBs in the living area in Thuyen village was mainly contribute by ELV- related activities (Tue et al.,2013).

PCB-28 congener was detected with higher concentration at all sampling site except CN1. PCB-52 congener was detected with higher concentration at CN1 site among other congeners (Fig.1). Some literatures corroborate with the present study, i.e. Anh et al,(2019) reported the predominance of PCB-28and PCB-52 in the industrail street dusts from Thai Nguyen and outdoor dust were found with abundance of PCB-28 from Guangzhou and Hong Kong,China (Wang et al.,2013). Some other reports also showed elevated concentration of PCB-28 and 52, which may due to the sampling area was close proximity to the port and ship dismandling area. This clearly showed that ship breaking and port activities could be the reason for abundant PCB-28 level (Chakraborty et al.,2013). Breivik et al,(2002) showed the dominat PCB-52 congener among others in Indian surface soil from in and around the open dumping sites. PCB-52 has higher half-life which retained in soil for long period and it was extensive global production and usage compound.

Table.2 Background activity of Soils samples in different ELV sites

S.No	ELV sites	Sample ID	Sources of scrapped vehincels	Date of collection
1	Chennai	CH1	Engine dismantle and dumped	17.09.2018
2		CH2	Lorry engine dismantle and storage	
3		CH3	Large types of truck engine dismantle and oil spilied noticed	
4		CH4	Bus engines was dismantle and spares was dumped	
5		CH5	Bus and Van body scrapped and burned (soil contained some ash)	
6	Coimbatore	MP1	Bunk of car engines are stored	26.08.2019
7		MP2	Car bodies are scrapped and dumpped	

The present study found with higher PCBs concentration than e-waste recycling households (Vietnamese) (1.0-2.2 ng/g), common houses in Japan (median: 1.6 ng/g) and USA (median: 5.6 ng/g), respectively

(Tue et al.,2010; Suzuki et al.,2010; Tue et al.,2013). Application of PCBs were considered to be closed application, such as hydraulic and heat transfer and cooling systems. These could not allow PCB leakout from the system. Dielectric fluids in capacitors and transformers were represented with 50-60% of sales of the PCBs in the USA (IARC,1978). Leung et al,(2006) reported that improper handling of e-waste was the source of PCBs in environment, specially old equipment dismandling results in the PCBs release.

PCBs concentration in soil from Mettupalayam (Coimbatore) ELV sites were range from 0.3 to 1.4 with the mean concentration of 0.1 to 0.4 ng/g dw, respectively (Fig.2). Higher PCBs concentration was observed at MP2 site than MP1, where four wheeler vehicles bodies were scrapped and dumped. PCBs have been used in various paint pigments (Hu and Hornbuckle, 2010; Kuusisto et al.,206). MP1 site was observed with lower total PCBs concentration, where vehicle engines were stored in closed manner. Hence the release of PCBs from the engines could be prevented. MP2 site have been observed with higher PCBs concentrtaion due to the dumping of vehicle bodies containing PCB containing oil-based paints, light ballasts, foam, cork, fibreglass, etc (Basel Convention, 2003). ELV site in Mettupalayam were proceeding the dismandling and scrapping in appropriate way (Table.2). Hence, contamination level was controlled at this site. UNEP, 2009 reported that e-waste and increase of e-waste recycling results in higher PCBs contamination in Ghana, Senegal, Nigeria,Kenya and the United Republic of Tanzania. Ali et al,(2012) reported the PCBs concentration in indoor floor dust from rural homes and mosques in Gurarat, Pakistan ranged between 0.3-6.1 ng/g. Concentration of PCBs in (209 PCBs) street dusts from northern Vietnam industrial, urban and rural areas were 14, 11, and 0.25 ng/g,respecively. PCBs concentration were higher in samples from industrail (Thai Nguyen) and urban areas (Hanoi) than rural sites (Giang). The reason behind this was samples collected near to desiel engine manufacturing company and the central area of the industrail park. Street dust from rural areas in Giang (Vietnam) were found with lower PCBs concentration (Anh et al.,2019). Street dusts from e-waste recycling facility (760-16000 ng/g) and some industrail sites (3600-63000 ng/g) in North Rhine- Westphalia, Germnay were found with extremely high PCB concentrations (Kless et al.,2017, 2015), which were relatively higher than those in the present study. Soil sample from Hanoi was detected with PCBs concentration ranged from 15 to 190 ng/g (Taon et al.,2007) and industrail soil was observed with PCBs concentration ranged from 18 to104 ng/g (Hue et al.,2016). PCBs in house dust from urban (5.6-85 ng/g) and suburban areas (3.6-20 ng/g) of Hanoi (Tue et al.,2013). The samples from industrail site (soil sample) and dust from urban and suburban areas showed lower PCBs concentration than those in the ELVs sites in cities of TamilNadu, which conclude that PCBs sources were lower in urban and suburban areas. Tue at al,(2016) reported the DL-PCBs levels in soil samples from e-waste recycling site in Agbogbloshie ranged between 3.4-83 ng/g, which was relatively lower than the present study. They also found DL-PCBs in non-burning areas were

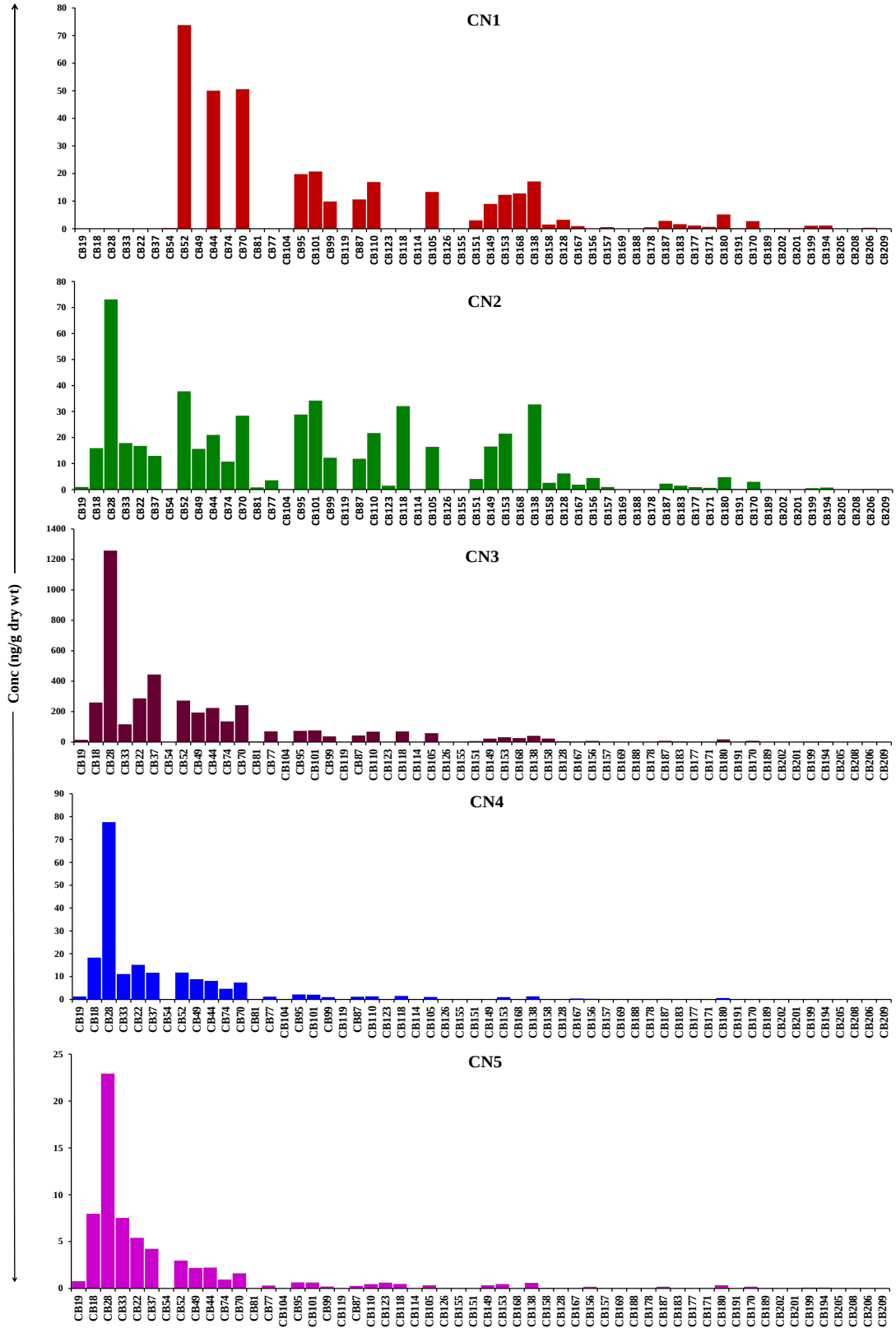


Fig.1 Levels of PCBs in ELV sites soils from Chennai, Tamil Nadu, India

ranged between 1.4-7.5 ng/g,respectively. DL-PCBs in open burning areas were found with higher level, which indicating the e-waste origing results in the higher DL-PCBs level (Hosoda et al.,2014).

Most of the studies quantified PCBs in samples from e-waste recycling sites. To our knowledge, this the first study to report PCBs levels in soils from ELVs sites in Chennai and Coimbatore, Tamil Nadu, India. PCBs concentration were higher in soil samples from Chennai ELVs sites than those in Mettupalayam ELVs sites, may due to the uasge of vehicles are vast and results in dumping of huge vehicles in such metropolitan city. Dismandling and scrappingof engines and vehicale bodies were not followed in proper procedure in unlicensed scrapping sites, could lead to higher PCBs contamination at the dumpig yard. Soil organic carbon (SOC) content was considered as important factor for binding of hydrophobic pollutants (Ribes et al.,2002). To better understand the PCBs distribution at contamination site, we should study the SOC of the particular site. In future, correlation between SOC and PCB concentration at particular site act an indicator to know the PCB source.

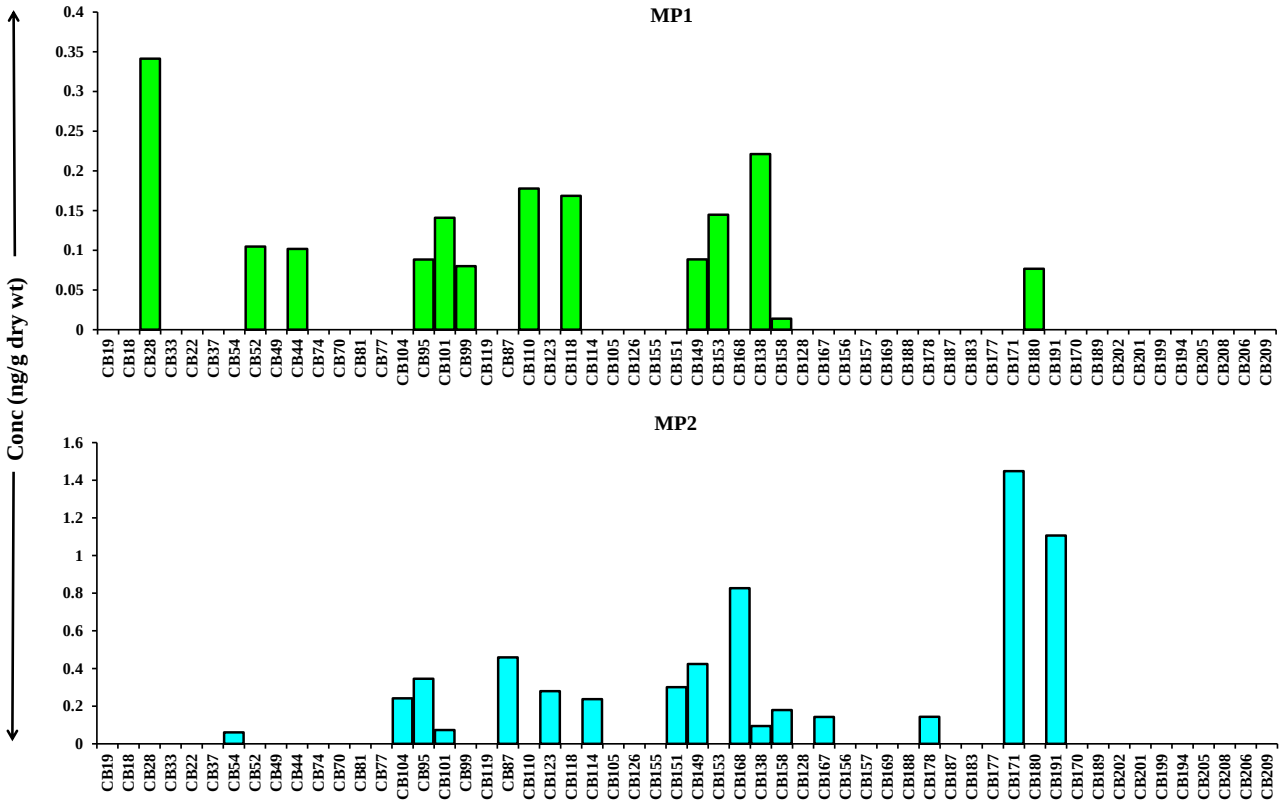


Fig.2 Levels of PCBs in ELV sites soils from Coimbatore, Tamil Nadu, India

5. List of publication and Conference presentation

5.1 Publications:

1. S. Sangeetha, **K. Vimalkumar**, A. Pugazhendhi (2019). Fate of Triclocarban (TCC) in aquatic and terrestrial systems and human exposure. *Journal of Chemosphere* (under review) (**IF 4.427**).

2. R. BabuRajendran, G. Preethi, R.K. Poopal, Nishikant Patil Nikhil**, K. Vimalkumar**, A. Subramanian, S. Krishna Kumar (2018). GC-MS determination of phthalate esters in human urine: a potential biomarker for phthalate bio-monitoring. *Journal of Chromatography B* 1079 (2018) 15–24(**IF 2.603**).

3. **Vimalkumar K**, ArunE,Krishna Kumar S, Poopal R, N. N. Patil**,** Subramanian A, BabuRajendranR. Occurrence of triclocarban and benzotriazole ultraviolet stabilizers in water, sediment, and fish from Indian rivers. *Science of the Total Environment*625 (2018) 1351–1360 (**IF 4.900**).

4. SrimuraliS,Krishna Kumar S, Govindaraj S, **Vimalkumar K**, Paromita C, BabuRajendranR(2016). Evaluating Spatial Distribution and Seasonal Variation of Phthalates Using Passive Air Sampling in Southern India. *Environmental Pollution* 221 (2017) 407-417. (**IF 5.099**).

5.2 Book Chapter:

1. Nikhil Nishikant Patil, S. Krishna Kumar, **K. Vimalkumar**, E.Arun, *BabuRajendran, R*. Organochlorine pesticide contamination in the Kaveri (Cauvery) river, India: A review on distribution profile, status, and trends. In: Water Challenges and Solutions on a Global Scale (Ed. Satinder Ahuja), ACS Books, Washington, DC, 2015: 447p.

5.3 Conference presentation:

1. Babu Rajendran R, **Vimalkumar K**, Patil Nikhil Nishikant, Arun E and PoopalRKentitledPerfluorinated compounds (PFCs) in Indian environment. 27th Symposium on Environmental Chemistry. (22-25, May 2018) at Okinawa, Japan.

2. **Vimalkumar K*,**Srimurali S, Krishna Kumar S, Govindaraj S, Paromita C, BabuRajendranRentitled Spatial Distribution and Seasonal Variation of Phthalates from atmospheric air using Passive Air Sampler in Southern India. International conference on Biodiversity and Sustainable Resource Management (ICBSRM 2018) (12th – 13th March, 2018) at Madras University, Chennai.

3. **Vimalkumar K***,E. Arun,S. Krishna Kumar, Nikhil NishikantPatil, RK. Poopal and R. BabuRajendranentitled Determination of Triclocarban (TCC) and Benzotriazole UV stabilizers (BUVSs) in surface water from South Indian Rivers. 19th International Conference Symposium on “Pollutant Responses in Marine Organisms” (PRIMO 19). (30th July – 3rd June, 2017) at Ehime University, Matsuyama, Japan.

4. **Vimalkumar K*,** E. Arun,S. Krishna Kumar, Nikhil NishikantPatil, RK. Poopal and R. BabuRajendranentitled Toxicity of metal and metal oxide nanoparticles to the Asian common Toad *DuttaphrynusMelanostictus*. 19th International Conference Symposium on "Pollutant Responses in Marine Organisms" (PRIMO 19). (30th July – 3rd June, 2017) at Ehime University, Matsuyama, Japan.

5. Subramanian A, BabuRajendran R, Takafumi Matsushita, **Vimalkumar K***, Tatsuya Kunisue and Shinsuke Tanabe entitled PCDD/Fs, DL-PCBs and PBDEs in the dumping site soils of Tiruchirappalli town, South India. 19th International Conference Symposium on "Pollutant Responses in Marine Organisms" (PRIMO 19). (30th July – 3rd June, 2017) at Ehime University, Matsuyama, Japan.

6. **Vimalkumar K***,Govindaraj, S. Krishna Kumar, S. Srimurali, Nikhil NishikantPatil, E. Arun and R. BabuRajendranentitledNonsteroidalanti inflammatory drugs (NSAIDs) in major rivers of Tamilnadu, India**.** International Conference on "Recent Trends in Bioscience". (07th – 09th February, 2016) at Alagappa University, Karaikudi, Tamilnadu, India.

7. **Vimalkumar K***, Bhuvaneswari R, Govindraj S, Arun E and BabuRajendran R entitled Human and Environmental Risk Assessment of Organochlorine pesticides in Water and Fish from River Cauvery, Tamilnadu. Two days workshop on "Micro pollutants in water and their hazards" (12th – 13th January, 2015) at IIT, Madras, Tamil Nadu, India.

8. **VimalkumarK***, Srimurali S, Krishna Kumar S, Govindaraj S, BabuRajendran R entitledDetermination of PCDDs, PCDFs and dioxin like PCBs in bovine milks and ash samples by using CALUX Assay in Tamilnadu, India. International Symposium on Halogenated Persistant Organic Pollutants (IEEP-2014) (January 16-17 2014) at CSIR-NEERI, Nehru Marg, Nagpur, Maharashtra, India.

9. **Vimalkumar.K*** present paper entitled "Study on effect of elevated CO_2 on growth and nutrient accumulation in important indigenous tree species" in International Symposium on Environmental Risk Assessment at Department of Zoology, School of Life Sciences, Bharathiar University, Coimbatore, Tamil Nadu, India (17th – 19th October, 2011)**.**

***Presenter**