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Note

Assessment of organochlorine pesticides in different seaweed species of Thoothukudi coast, Tamil Nadu, South India

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ABSTRACT

Five species of macroalgae, namely *Sargassum wightii*, *Padina tetrastromatica*, *Ulva lactuca*, *Caulerpa racemosa* and *Gracilaria edulis* collected from three sampling sites, *viz*. Harbour Beach (site 1), Tharuvaikkulam (site 2) and Hare Island (site 3), situated along the Thoothukudi region of the Gulf of Mannar were analysed for 17 organochlorine pesticides (OCPs) by gas chromatography-mass spectrometry (GC-MS) to assess their suitability as biomonitors of pesticide contamination. The total OCP concentrations were several folds higher in all the macroalgae collected from site 1 than in the other two sites. Brown alga *S. wightii* accumulated more OCPs (297 ng g^{-1}) than the other sampled algal species in site 1. The OCPs assessed in this study were endrin aldehyde (51 ng g^{-1}), endrin (48 ng g^{-1}), β-hexachlorocyclohexane (45 ng g^{-1}), heptachlor epoxide (43 ng g^{-1}), dichlorodiphenyldichloroethylene (37 ng g^{-1}) and endosulfan sulfate (11 ng g^{-1}) in *S. wightii*. It was observed that the green alga *C. racemosa* had selective sorption of endosulfan I (43 ng g^{-1}) and red alga *G. edulis* had selective sorption of endosulfan II (44 ng g^{-1}). The ability of seaweeds to accumulate pesticides was found to be species specific and not class specific. The study indicated that *S. wightii* could be used as a biomonitor of OCP residues in the marine environment, with the exception of endosulfan.

Keywords: Endosulfan, Macroalgae, OCPs, Pesticides

Macroalgae are widely used as food and animal feed in Asian countries. They are low-calorie food rich in minerals, vitamins, essential unsaturated fatty acids (particularly long-chain n-3 polyunsaturated fatty acids), as well as soluble and insoluble dietary fibers (Dhargalkar and Verlecar, 2009). Besides having appreciable food value, they are often contaminated with chemical contaminants such as pesticides, heavy metals and polyaromatic hydrocarbons that enter the marine environment through either agriculture or allied activities (Pavoni *et al.,* 2003; Harnando *et al.,* 2007; Burridge *et al.,* 2010; Gran-Scheuch *et al.,* 2020). The biological sensitivity of seaweeds to chemical contaminants differs from one species to another (Lytle and Lytle, 2001). Their selective accumulation makes them suitable bioindicators of chemical contamination in coastal waters (Pavoni *et al.,* 2003).

The presence of pesticide residues in macroalgae has received more attention in the past few decades (Maroni *et al.,* 2000). Organochlorine pesticides (OCPs) tend to associate with macroalgae due to their hydrophobicity and persistence nature (Mwevura *et al.,* 2002). They affect the human health through either direct consumption or ingestion by fish or other marine organisms (Moreno *et al.,* 2007). Hence, monitoring of pesticides has become one of the most important aspects in minimising potential hazard to human health (Garcia-Rodriguez *et al.,* 2012). Macroalgae such as *Fucus*, *Ulva*, *Gracillaria*, *Porphyra*, *Grateuloupia*, *Undaria* and *Cystosseira* are reported as environmental biomonitors, as they show contamination of organic micropollutants such as polychlorinated biphenyls, chlorinated pesticides and polycyclic aromatic hydrocarbons (Pavoni *et al.,* 2003; Nunes *et al.,* 2008). Nevertheless, studies pertaining to the presence of pesticides in the wild and edible macroalgae are scarce.

Gas chromatography (GC) and high performance liquid chromatography (HPLC) are widely used for the analysis of pesticide residues (Boer and Law, 2003). The preparation of sample before chromatographic analysis is one of the most critical steps in the analytical process. QuEChERS (quick, easy, cheap, effective, rugged and safe) kit was used for the extraction of pesticides from

seaweeds before GC analysis (Anastassiades *et al.,* 2003). The kit consists of primary and secondary amines (PSAs) that remove sugar, fatty acids and organic acids; carbon-18 (C-18) resin, which removes long-chain fatty compounds, sterols and nonpolar interference; graphitised carbon black (GCB), which removes carotenoid pigments and magnesium sulfate ($MgSO_4$), which separates the solvent portion to improve the recovery of pesticides. The advantage of QuEChERS method is that it does not require any special equipment for the separation of pesticides unlike other procedures.

In India, 650 species of marine macroalgae are documented, including 320 species of Rhodophyta (red algae), 165 species of Chlorophyta (green algae) and 150 species of Phaeophyta (brown algae) (GoMBRT, 2015). In the Gulf of Mannar, there are 147 species comprising 42 green, 31 brown and 69 red macroalgae (Ramesh, 1996). *Sargassum wightii*, *Ulva lactuca*, *Caulerpa racemosa*, *Padina tetrastromatica* and *Gracilaria edulis* are macroalgae most commonly found in this region. In this study, the OCP residue contamination in the macroalgae available widely in Thoothukudi coast was investigated to identify their suitability as biomonitors of pesticides.

Macroalgae were collected from three sampling sites *viz*., (1) Harbour Beach (8.74°N; 78.17°E), (2) Tharuvaikkulam (8.89°N; 78.17°E) and (3) Hare Island (8.78°N; 78.19°E) located in the Thoothukudi region during October-November 2017. Two green algal species, *C. racemosa* (*mookutthi paasi*) and *U. lactuca* (*pattu paasi*); two brown algal species, *S. wightii* (*vaeppelai paasi*) and *P. tetrastomatica* (*elikkathu vagai paasi*) and one red algal species, *G. edulis* (*kanji paasi*), were chosen for the study. Macroalgae were collected during low tide, washed thoroughly in seawater to remove extraneous materials, packed in polyethylene bags and brought to the laboratory immersed in seawater in order to prevent drying. They were once again washed with potable freshwater to remove residual salt, epiphytes, animal castings, calcareous and other adhering detritus matter and dried under shade.

Analytical techniques, including sample extraction, purification, instrumentation and quality control and quality assurance, have been reported earlier (Sundhar *et al.,* 2019; 2020). Briefly, OCPs from seaweed were extracted by the QuEChERS extraction and purification method given by AOAC (AOAC 2007.01, 2007) and analysed by Gas Chromatography-Mass Spectrometry (GC-MS) (Thermo Fisher Trace 1300 and ISQ LT Single Quadrupole Mass Spectrometer, Germany). The recovery and detection limit was checked by spiking the seaweed with 100 ng g^{-1} OCP standard mix and a six-point (1 to 250 ng g^{-1}) linear curve was plotted for each pesticide. The lowest point (1 ng g^{-1}) of the calibration curve is the detection limit (LOD). In the same way as environmental samples, blank samples (solvent only) were prepared and analysed. Each sample was analysed in triplicate. A blank sample was performed to search for pollutants that could have derived from reagents and apparatus.

The selected macroalgae were abundant during October and November in the Gulf of Mannar, corresponding to the fruiting period of these algae (Rani *et al.,* 2015). Among the three selected sampling sites, sites 1 and 3 were located within the industrialised city limits and experienced greater influx of contaminated water from land-based agrochemical industries and occasional agricultural runoff into the estuarine region. All the analysed 17 OCPs were present in the macroalgae sampled. The total OCPs recorded at site 1 ranged from 19.89 to 297.04 ng g^{-1}, at site 2 from 0.60 to 27.72 ng g^{-1} and at site 3 ranged from 0.64 to 35.56 ng g^{-1} (Table 1). Analyses of the macroalgae samples from site 1 showed high absorption of heptachlor epoxide, endrin, endrin aldehyde, endosulfan I, endosulfan II, endosulfan sulfate, α-, β-, γ- and δ-HCH than the other two sites. The presence of agrochemical industries and agricultural runoff from the nearby estuary might have contributed to the high concentrations of OCPs at this site. Site 2 was comparatively less polluted with OCPs, possibly due to lack of agrochemical industries and agricultural runoff nearby. Site 3 was also comparatively less polluted than site 1, as it did not receive any direct estuarine discharge containing agricultural contaminants.

S. wightii is considered to adsorb more pesticides from the marine environment (20-297 ng g^{-1}) followed by *C. racemosa* and *P. tetrastromatica*. In this investigation, *S. wightii* had accumulated the highest level of OCPs at site 1 (297 ng g^{-1}) and site 3 (36 ng g^{-1}), whereas *P. tetrastromatica* accumulated the highest level of OCPs at site 2 (28 µg g^{-1}). Brown algae were observed to accumulate higher pesticides than red or green seaweeds (Maroli *et al.,* 1993). Some earlier reports have also stated that DDT concentration was higher in Dictyota than *U. lactuca* and *G. edulis* (Maroli *et al.,* 1993).

Concentration of OCPs recorded at site 1 was higher in *S. wightii* mainly because of the sorption capacity of heptachlor epoxide (43 ng g^{-1}), endrin (48 ng g^{-1}), endrin aldehyde (51 ng g^{-1}) and endosulfan derivatives (43 ng g^{-1}). In addition, concentrations of α-, β-, γ- and δ-HCH (46 ng g^{-1}) were also high in *S. wightii* compared to other seaweeds. In the samples from site 2, the OCPs present at higher concentration in *P. tetrastromatica* were β-HCH (11 ng g^{-1}), endosulfan I (6.6 ng g^{-1}) and endrin (6 ng g^{-1}). In the samples from site 1, *C. racemosa* was found to possess the tendency to uptake more of

Table 1. Pesticide accumulation (ng g^{-1}) in different seaweeds collected from the sampling sites in Thoothukudi

	S.wightii			*P. tetrastromita*			*C. racemosa*			*U. lactuca*			*G. edulis*		
	Site -1	Site -2	Site -3	Site -1	Site -2	Site -3	Site -1	Site -2	Site -3	Site -1	Site -2	Site -3	Site -1	Site -2	Site -3
Alpha-BHC	5.03	0.00	0.00	0.02	0.01	0.00	0.38	0.05	0.03	0.03	0.00	0.00	0.07	0.00	0.00
Beta-BHC	29.60	5.35	5.24	22.20	10.85	7.78	16.30	3.38	3.85	2.18	0.18	0.13	3.00	0.12	0.07
Gamma-BHC	5.97	0.16	0.11	0.86	0.23	0.12	1.18	0.15	0.20	0.39	0.00	0.01	0.54	0.01	0.00
Delta-BHC	5.05	0.10	0.11	0.02	0.01	0.01	0.44	0.05	0.08	0.14	0.00	0.00	0.05	0.00	0.00
∑ BHC	45.64	5.61	5.47	23.10	11.10	7.91	18.30	3.63	4.17	2.74	0.19	0.14	3.67	0.13	0.07
Heptachlor	1.85	0.12	0.13	0.27	0.07	0.05	0.15	0.00	0.00	0.02	0.04	0.01	0.17	0.00	0.01
Heptachlor-epoxide	43.11	0.00	0.00	0.08	0.00	0.00	0.35	0.00	0.00	0.35	0.00	0.00	0.03	0.00	0.00
∑ Heptachlor	44.96	0.12	0.14	0.36	0.07	0.05	0.50	0.00	0.00	0.37	0.04	0.01	0.20	0.00	0.01
Endosulfan I	14.91	2.53	6.51	17.68	6.60	8.45	43.75	3.23	2.13	4.96	0.03	0.00	19.54	0.23	0.22
Endosulfan II	16.87	0.03	0.02	7.16	0.11	0.01	1.01	0.05	0.08	0.18	0.04	0.04	43.46	0.02	0.03
Endosulfan-sulphate	11.10	0.02	0.01	0.52	0.05	0.01	1.69	0.07	0.22	0.48	0.00	0.02	0.67	0.00	0.00
∑ Endosulfan	42.88	2.58	6.54	25.36	6.75	8.47	46.45	3.36	2.43	5.62	0.08	0.05	63.67	0.25	0.24
Aldrin	0.25	0.05	0.54	0.53	0.54	2.47	5.68	2.99	3.49	0.63	0.03	0.02	1.06	0.07	0.09
Dieldrin	11.57	0.02	0.02	0.05	0.10	0.01	0.33	0.04	0.19	0.33	0.00	0.00	0.08	0.02	0.02
∑ Aldrin	11.82	0.07	0.56	0.57	0.64	2.48	6.01	3.03	3.68	0.96	0.03	0.02	1.14	0.09	0.11
Endrin	48.03	0.87	1.54	0.11	5.81	2.91	8.00	1.25	1.18	7.68	0.21	0.33	1.02	0.18	0.17
Endrin-aldehyde	50.98	11.07	20.91	6.61	3.23	2.05	6.93	2.93	3.03	1.23	0.00	0.04	1.51	0.01	0.02
∑ Endrin	99.01	11.94	22.44	6.72	9.03	4.95	14.93	4.18	4.21	8.91	0.21	0.37	2.53	0.19	0.19
PP'-DDE	37.13	0.00	0.00	0.16	0.01	0.01	0.47	0.00	0.00	0.35	0.00	0.00	0.19	0.00	0.00
PP'-DDD	11.31	0.13	0.16	0.02	0.00	0.00	0.63	0.08	0.09	0.10	0.00	0.00	0.01	0.00	0.00
PP'-DDT	0.00	0.03	0.02	0.00	0.05	0.01	0.00	0.05	0.03	0.04	0.02	0.02	0.34	0.01	0.01
∑ DDT	48.44	0.16	0.18	0.18	0.06	0.02	1.10	0.12	0.11	0.49	0.02	0.02	0.54	0.01	0.01
Methoxychlor	1.12	0.12	0.08	0.02	0.02	0.02	0.12	0.02	0.03	0.01	0.02	0.00	0.29	0.00	0.00
Cypermethrin-1	1.75	0.01	0.01	0.79	0.00	0.00	0.05	0.00	0.05	0.73	0.01	0.01	0.29	0.02	0.00
Cypermethrin-2	0.00	0.07	0.14	0.15	0.03	0.02	0.13	0.12	0.12	0.00	0.02	0.02	0.20	0.01	0.01
Cypermethrin-3-4	1.42	0.00	0.00	0.00	0.00	0.00	0.15	0.16	0.13	0.06	0.00	0.01	0.24	0.01	0.01
∑ Cypermethrin	3.17	0.09	0.16	0.94	0.03	0.02	0.33	0.28	0.30	0.79	0.03	0.03	0.73	0.04	0.02
Total	297.04	20.67	35.56	57.24	27.72	23.92	87.74	14.62	14.93	19.89	0.60	0.64	72.77	0.71	0.66

β-HCH, endosulfan I, aldrin and endrin derivatives and the accumulation of endosulfan I (43 ng g^{-1}) was the maximum. Similarly, *G. edulis* adsorbed higher concentrations of endosulfan II (44 ng g^{-1}) and endosulfan I (20 ng g^{-1}), whereas the uptake of other OCPs was minimal. In *U. lactuca*, the total OCPs detected were very low (<1 ng g^{-1}), with the exception of site 1 (20 ng g^{-1}), due to a sorption of β-HCH (2 ng g^{-1}), endosulfan (5 ng g^{-1}) and endrin (8 ng g^{-1}).

The potential of different species of macroalgae to accumulate OCPs was individually assessed to examine their suitability as biomonitors of pesticide contamination in the marine ecosystem. Lindane (γ-HCH), formerly known as γ-BHC **(**benzene hexachloride**)**, is an insecticide that interferes with GABA neurotransmitters as a neurotoxin. HCH consists of eight isomers, but only γ, α-, β- and δ-HCH are of commercial significance. In samples from site 1, β-HCH was predominant compared to other isomers. *S. wightii*, *P. tetrastromatica* and *C. racemosa* had accumulated 30, 22 and 16 ng g^{-1} β-HCH, respectively. Their presence in macroalgae is a concern as it has been recently classified as group I human carcinogen (IARC, 2017). *U. lactuca* and *G. edulis* had accumulated 10 times less β-HCH than *S. wightii.* In the Venetian Lagoon (Italy), *Ulva* spp accumulated 11 ng g^{-1} HCH (Pavoni *et al.,* 2003) and such a high concentration was not found for *U. lactuca* (2.74 ng g^{-1}) in this study.

Heptachlor insecticide has been banned for over 40 years. Heptachlor epoxide is a derivative of heptachlor, which is more toxic to humans. In this study, *S. wightii* had specifically accumulated heptachlor epoxide and the concentration was 43 ng g^{-1} at site 1. The presence of heptachlor epoxide in macroalgae even after the long period of ban gives an indication of their persistence nature. The specific accumulation of heptachlor epoxide by *S. wightii* alone makes it a biomonitor of heptachlor contamination.

The use of endosulfan is restricted in agriculture due to its acute toxicity. On the basis of Stockholm Convention 2011, more than 80 countries have banned the use of

endosulfan and its derivatives in agricultural activities. In some parts of the world, endosulfan is still illegally used in tea and rubber plantations. In this study, endosulfan I, II and their sulfate derivatives were detected in macroalgae, dominated by endosulfan I. Sorption of endosulfan by macroalgae was very distinct at site 1. *S. wightii* absorbed endosulfan I, II and endosulfan sulfate at concentrations ranging between 11 and 17 ng g^{-1}. *P. tetrastromatica* (18 ng g^{-1}) and *C. racemosa* (44 ng g^{-1}) absorbed more endosulfan I whereas *G. edulis* absorbed more endosulfan II (44 ng g^{-1}). The differential accumulation pattern by the macroalgae suggested that *C. racemosa* and *G. edulis* are the best biomonitors of endosulfan I and II, respectively, in the marine environment.

Aldrin is a highly carcinogenic substance banned for 20 years due to its persistence and toxicity. It readily metabolises to dieldrin in plants and animals. In this study, dieldrin was detected only in *S. wightii* sample from site 1 (12 ng g^{-1}). However, aldrin was found in *C. racemosa* at concentrations of 3-6 ng g^{-1} at all the sampling sites. Specific absorption of aldrin by *C. racemosa* and dieldrin by *S. wightii* indicates that these macroalgal species could serve as best biomonitors for these pesticides.

Endrin is an insecticide, rodenticide and piscicide that binds very strongly to organic matter in aquatic sediment due to its high adsorption coefficient (Zitko, 2003). It is noncarcinogenic to humans; however, it affects the central nervous system (IARC, 1987). At site 1, *S. wightii* had accumulated more endrin (48 µg g^{-1}) and endrin aldehyde (51 µg g^{-1}) than the other seaweeds. *S. wightii* had shown special affinity towards sorption of endrin aldehyde than endrin and the same was also evidenced at sites 2 and 3. Breakdown of endrin depends on the local environmental conditions. For instance, sunlight leads to more rapid breakdown of endrin into endrin aldehyde and endrin ketone (www.epa.gov). Thoothukudi region has an average temperature of 29.2°C (maximum 35.9°C; minimum 22.6°C), which might have contributed for the breakdown of endrin compound. *P. tetrastromatica* and *C. racemosa* had also accumulated both the endrin derivatives in similar concentrations. *U. lactuca* had accumulated endrin at site 1, whereas *G. edulis* showed very less sorption for endrin derivatives.

DDT is an insecticide widely used to combat malaria, typhus and other insect-borne diseases. Owing to its lipophilic nature, it is accumulated in the fat of all living organisms. It has been banned in many countries since the early 1970s because of its toxicity to humans. DDE is a metabolite formed by the loss of hydrogen chloride from DDT, which is a reproductive toxicant but rarely excreted from the body. DDD is another metabolite used to kill insects and treat cancer of the adrenal gland. DDT and its derivatives can persist for nearly 150 years in the aquatic environment. In our study, the amount of ∑DDT was very high in *S. wightii* found at site 1 (48 ng g^{-1}) and was very minimal in other macroalgae. DDT derivatives such as DDD (11 ng g^{-1}) and DDE (37 ng g^{-1}) were detected in higher amounts in *S. wightii* at site 1. This shows that DDE and DDD were not used directly as pesticides and might have been formed as breakdown products of DDT and persisted in the environment. Specific accumulation of DDT metabolites by *S. wightii* was also noticed, because the other algae at site 1 did not accumulate them. Amico *et al.* (1979) recorded DDT residues in the red alga *Liagora viscida* (2.1 ng g^{-1}) and brown alga *Cystoseira stricta* (20.1 ng g^{-1}) collected near the industrial area in the East Coast of Sicily, Italy. Maroli *et al.* (1993) also reported higher amount of DDT in *Dictyota* (10.6 ng g^{-1}) compared to *Ulva* (0.8 ng g^{-1}) and *Gracilaria* (2.0 ng g^{-1}) from the lagoon of Venice, Italy.

The anecdote of high accumulation of DDT by the brown algae than green and red algae need not always be true, because in this study, *P. tetrastromatica* did not accumulate DDT as did *S. wightii*. In an earlier study, a very low concentration (0.1 ng g^{-1}) of DDT metabolites (DDD and DDE) has been reported in *Cystoseira* present in the Venetian Lagoon (Pavoni *et al.,* 2003). The other macroalgae, *U. lactuca*, *P. tetrastromatica*, *G. edulis* and *C. racemosa*, accumulated DDT derivatives only at low levels. Thus, accumulation of the pesticides by the macroalgae was found to be species-specific rather than class-specific. This study revealed that *S. wightii* could serve as a biomonitor of DDT-derived pesticides (DDE and DDD).

Methoxychlor is mainly produced as the replacement of DDT because of its acute toxicity and endocrine disruptor characteristics and has been banned since 2004. It has no carcinogenic effect on humans but causes liver damage in animals (Kim and Lee, 2017). In the present study, methoxychlor and cypermethrin derivatives were present at very low levels (<2 ng g^{-1}) in macroalgae collected from site 2. *S. wightii* accumulated methoxychlor slightly higher than *G. edulis* and other algae.

It could be inferred from this study that the macroalgae available at site 1 (Harbour Beach) are more polluted with OCP residues. Among the macroalgae investigated, *S. wightii* accumulated more OCPs, followed by *C. racemosa* and *P. tetrastromatica*. *U. lactuca* accumulated the least. *S. wightii* could be a suitable biomonitor of pesticides such as heptachlor epoxide, dieldrin, endrin aldehyde and DDT derivatives. Similarly, *C. racemosa* could be a selective biomonitor for endosulfan I and *G. edulis* for endosulfan II. Macroalgae could also serve as biomonitors of specific derivatives of OCPs. Particularly,

S. wightii, *C. racemosa* and *G. edulis* indicated β-HCH contamination. So, in order to monitor pesticide pollution in the marine environment, the regulatory authorities can selectively identify the macroalgae to be used as biomonitors.

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