

# Persistent Organic Pollutants (POPs) in Locally Made Antifouling Agent (*Sifa*)

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**Abstract-** This paper reports the level of persistent of organic pollutants (POPs) from locally made antifouling agent (*Sifa*) in Zanzibar. A total of 9 samples of the antifouling agent prepared from decayed mixture of blubber and liver of common bottlenose (*Tursiops truncatus*) dolphins were analysed for a wide range of acid stable organohalogen compounds. Organochlorine pesticides mostly DDT metabolites and methoxylated brominated diphenyl ethers were found well above the detection limits. The levels of DDTs measured in this study were highly dominated by p,p'-DDE reflecting the effect of metabolism due to the decay process. The concentrations of two MeO-BDEs found in this study showed significant correlation ( $R^2 = 0.952$ ) indicating that they originate from the same source and they have same stability to microbial degradation. Pollutants of industrial origin, such as polychlorinated biphenyls (PCBs) were not detected showing that the sampled areas are free from persistent industrial pollutants. The results indicate that users of local antifouling agent are exposed to aged chlorinated contaminants though the levels have no detrimental effects on their health. On the contrary, HCHs were found in trace levels in fresh samples of dolphin liver, indicating their relatively low persistence compared to DDTs.

**Keywords-** Local antifouling; methoxylated brominated diphenyl ethers; organohalogen; Persistent Organic Pollutants (POPs); *Sifa*

## I. INTRODUCTION

Marine fouling is undesirable attachment or accumulation of macroorganisms, plants, and animals on artificial surfaces immersed in sea water [1]. Once attached to a surface, these life forms rapidly divide and form a slime film of great importance to the emerging fouling community. About 5000 biological species have been listed as causing the fouling of structures exposed to or immersed in water. Animals involved in macrofouling consist largely of barnacles, mussels, bryozoans, hydroids, tunicates and serpulid worms [2].

Marine fouling is most common on ship hulls, dhows, navigational buoys and underwater equipment. The fouling process is associated with various problems including development of friction which reduces the speed of a vessel thereby increasing fuel consumption, reducing desalination and performance of power plants fixed in sea water. Fouling may sometimes destroy the timber surface of the boat [3]. There has been a significant investment in researches worldwide, to understand the phenomenon of biofouling and develop appropriate strategies for its prevention and control. The marine industry is estimated to incur an expenditure of 10 billion sterling pounds annually to combat the problems associated with fouling [4].

Historically, various methods including firing, application of tar and wax as well as metals (Zn, Pb, As, Ni) sheathing techniques were commonly used to control fouling. Types of material and techniques used to combat fouling varied from place to place and time. Unfortunately, most of the developed methods were found to be inefficient in controlling the fouling and consequently the last century witnessed significant advancements in the development of new antifouling agents. In the 19<sup>th</sup> century, tributyltins (TBTs) have been most often used as antifouling agents and are very effective against both soft and hard fouling organisms. However, in spite of their performance, these chemicals are associated with negative detrimental impacts on the marine environment and long half life in the environment [5]. More efforts to look for nonpersistent alternatives have resulted in introduction of booster biocides commonly Irgarol<sup>®</sup> and diuron. Again, these chemicals have been found to be equally harmful to marine environment because they are photosynthetic inhibitors capable of bleaching coral and deforming aquatic organisms [6]. Booster chemicals have been also reported to be among the common pollutants in ground water [7].

In Zanzibar, evidence shows that TBT has been used and amount of their residues along the harbour are still above the detectable levels. The measured residues were associated with past use of TBT in fumigating ships and boats around the harbour [8]. On the other hand, the local fishermen were not able to shoulder the costs of chemical antifouling for protecting their dhows and mostly used local antifouling commonly known as *sifa*. *Sifa* is prepared by decaying rich fat organs of marine fish and mammals

The existing challenge which has not been addressed is to find a material that will disrupt the life cycle of these organisms, and thus prevent their accumulation and growth on the surfaces on which they accumulate, while not impacting the environment.

*Sifa* is often used and considered as very effective in combating marine fouling in Zanzibar, however, its potentiality to be used as a veritable way of circumventing this challenge has not been investigated. Interviews with the locals showed that high quality *sifa* is obtained by using fat-rich organs of carnivorous fish and mammals. But researches have shown that fat-rich organs from carnivorous organisms accumulate higher levels of POPs [9].

This calls for the need to determine types of POPs in *sifa* and to investigate the extent to which they survive degradation during the decay process. It is also of utmost importance to investigate safety issues related with use of *sifa*. This study was therefore intended to determine the type, levels and composition of POPs in *sifa* and compare measured levels with fresh samples. The study also investigated local community awareness as well as health and environmental risks associated with the use of *sifa*.

## II. METHODOLOGY

### A. Collection of Sample and Sample Storage

Blubber and liver of bottlenose dolphins stranded along the coast of North Unguja Island at Tazari were used by fishermen as raw material for preparation of *sifa* which was used in this study. The samples were collected from 9 local fishermen at the preparation grounds. *Sifa* was thoroughly stirred in preparation containers before sampling, to ensure homogeneity. The sampled portion was then kept in sealed glass bottle and stored at  $-20^{\circ}\text{C}$  until extraction.

### B. Sample Preparation

*Sifa* samples were defrosted and homogenized by stirring with electrical mixer. A sub-sample (5 g) was mixed and ground with anhydrous sodium sulphate and internal standard (hexabromobenzene - HBB) to give free floating powder. The powder was extracted successively by shaking for 10 min with 50 + 20 + 20 + 20 ml of dichloromethane using an overhead shaker. The extracts were filtered through anhydrous sodium sulphate and the solvent reduced to 2 ml volume by using rotary evaporator.

### C. Cleanup

Sulphuric acid treatment cleanup of extract was done by partitioning technique [10]. The extract in cyclohexane ( $2\text{ cm}^3$ ) was directly mixed with acid in a test tube. The content of the test tube was cautiously mixed by inverting the tube for half a minute before centrifugation for 5 minutes. The upper organic phase was transferred to a pre-cleaned graduated test tube using a Pasteur pipette and its composition changed to the appropriate solvent (cyclohexane/acetone 9:1) for injection into the GC.

### D. Instrumental Analysis

Organochlorine pesticides (OCPs) were quantified with gas chromatography (GC) on an Agilent 6890 (Agilent, Wilmington, Delaware, USA) equipped with two electron capture detectors. Separation was performed simultaneously on two capillary columns of different polarity (CP-Sil 5CB and CP-Sil 19CB, 30 m x 0.32 mm x 25  $\mu\text{m}$ , Chrompak, Middelburg, The Netherlands) attached to the same split-less injector, but with separate detectors. The column temperature program employed was  $90^{\circ}\text{C}$  (equilibrium time 1 min), rising to  $180^{\circ}\text{C}$  at  $30^{\circ}\text{C}/\text{min}$ , then to  $260^{\circ}\text{C}$  at  $4^{\circ}\text{C}/\text{min}$  and held for 10 min. The injector and detector were held at  $250^{\circ}\text{C}$  and  $300^{\circ}\text{C}$ , respectively. The analytes were identified and quantified by comparison of the retention times and peak heights with authentic reference standards relative to HBB on the two columns.

Brominated compounds were identified by gas chromatography-mass spectrometry (GC-MS) with electron capture negative ion (ECNI) ionization in the full scan mode. Mass spectra and retention times were compared with the spectra of MeO-BDEs reference standards. The brominated compounds were identified by the presence of the fragment ions  $m/z$  79, 81, 159, 161 516 and 530 [11].

### E. Data Analysis

Statistical analyses were performed using the Student's t-test to examine the difference between means of the two MeO-BDEs. Simple regression analysis ( $R^2$ ) was used to examine the relationships between accumulation of MeO-BDEs.

Level of community awareness on the functioning of *sifa* and possible risks associated with chemical pollutants in it were investigated by interview method. All 9 fishermen who kindly provided *sifa* samples were interviewed during the study.

## III. RESULTS AND DISCUSSION

A total of 9 *sifa* samples were screened for a wide range of acid stable persistent organic pollutants. The analysis indicated presence of two major groups of compounds, namely OCPs and methoxylated brominated diphenyl ethers (MeO-BDEs). These substances were confirmed by using chromatograms from two capillary columns (SE-30 and OV-1701) of different polarities. The concentrations, method recovery and detection limits of all measured residues are given in Table 1.

TABLE 1 CONCENTRATION ( $\mu\text{g/g}$ ) OF ORGANOCHLORINE COMPOUNDS IN IN *SIFA* SAMPLE

SAMPLE NAME	HCB	ALDRIN	Alfa-HCH	Delta-HCH	p,p'-DDD	p,p'-DDE	p,p'-DDT	o,p'-DDT
SF1	BDL	BDL	BDL	BDL	36	1151	64	BDL
SF2	56	BDL	BDL	BDL	39	1976	121	6
SF3	BDL	BDL	BDL	BDL	5	160	30	BDL

SF4	BDL	2	2	BDL	3	25	4	1
SF5	26	BDL	BDL	BDL	41	1217	120	BDL
SF6	BDL	BDL	BDL	BDL	41	2120	166	11
SF7	BDL	BDL	BDL	BDL	16	459	59	11
SF8	25	BDL	BDL	BDL	11	106	20	BDL
SF9	88	BDL	BDL	8	17	191	41	BDL

**Note:** BDL stands for Below Detection Limit.

#### A. Organochlorine Compounds

The OCs group found to be above detection limit includes hexachlorobenzene (HCB), hexachlorocyclohexanes (HCHs) and DDTs.

Types and levels of organochlorine found in *sifa* samples differed in many ways from fresh organ samples of dolphin accidentally caught at the same location. The fresh samples were rich in different types of POPs [12] while *sifa* samples had very few POPs. The difference was obviously attributed to the decaying process for formation of *sifa*. This demonstrates that POPs are largely resistant to microbial degradation but some of them can be degraded through the decaying process.

In general, organochlorine residues were dominated by DDTs metabolites and the other residues were detected in very low frequency and concentration. Concentration of DDTs contributed to 98% of the total measured organochlorine residues. DDTs dominance indicates their strong refractory nature of being resistant to microbial degradation and thus survived the whole decaying process [13]. On the other hand, HCH despite being resistant, can be microbial degraded to give other simpler products [14], cyclodienes such as aldrin and dieldrin are well known stable POPs but only aldrin was detected in one sample from this study and dieldrin which is more resistant to degradation [15] was not detected. The absence of dieldrin is not surprising because the samples were cleaned using sulphuric acid treatment which can degrade aldrin [10].

#### B. DDTs

The DDT compounds measured in this study are composed of parent molecules p,p'-DDT and o,p'-DDT as well as metabolite molecules p,p'-DDE and p,p'-DDD. Both parent molecules were detected in fewer samples and lower concentrations than metabolites. p,p'-DDT is a major isomer in technical product and it was measured in all samples. Its concentration ranged between 4 to 166 µg/g w/w (Average 69.4 µg/g w/w). The minor isomer o,p'-DDT was detected in 44 % of the analysed samples and its concentration was from BDL to 11 µg/g ww. The p,p'-DDE was by far the dominant metabolite in terms of frequency of detection and concentrations. It was found in all *sifa* samples with mean concentrations of 822.8 µg/g w/w ranging from 25 to 2120 µg/g w/w. The other metabolite, p,p'-DDD was found in all analysed samples with concentrations ranging from 3 to 41 µg/g w/w.

Fresh technical DDT which is applied on the field is a mixture containing more than 70% p,p'-DDT and less than 15% o,p'-DDT [15] and the presence of other metabolites indicates occurrence of metabolism. DDT undergoes degradation to give DDE as a major and most stable metabolite favoured under aerobic conditions and DDD which mainly formed under anaerobic conditions. Compositions of DDT are given in Fig. 1. The minor isomer of o,p'-DDT contributed to 0.34% of total DDT while the major isomer p,p'-DDT contributed 7.37% of the total DDTs concentration. On the other hand, the metabolite p,p'-DDE dominated the total DDT residues by contributing 87.37% of total concentration of DDTs while p,p'-DDD had minor contribution of 2.47% of total DDT. The contribution of p,p'-DDT was much lower in *sifa* samples than of fresh technical mixture.

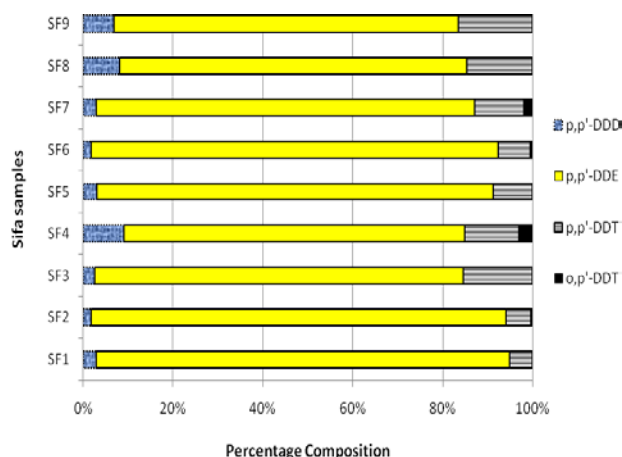


Fig. 1 DDTs composition in a *sifa* samples

The low contributions of *o,p'*-DDT was likely due both to its low percentage in the technical product and its rapid metabolism as indicated in detection frequency compared to other parent molecule. The dominance of *p,p'*-DDE in *sifa* samples can be explained by the relatively rapid conversion of DDT to DDE both abiotically by hydrolysis (elimination of hydrochloric acid, HCl) and biotically under aerobic conditions. *p,p'*-DDD is a minor contributor to total DDT in these samples because it is formed mainly under anaerobic conditions and can be easily mineralised to DDMU. This mineralisation process was likely responsible for the lower contribution of *p,p'*-DDD than *p,p'*-DDT in the samples.

#### C. Hexachlorocyclohexanes (HCHs)

HCHs exist in four common isomers in technical products, namely  $\alpha$ -HCH,  $\beta$ -HCH,  $\gamma$ -HCH, and  $\delta$ -HCH. In this study, only  $\alpha$ -HCH and  $\delta$ -HCH isomers were detected at measurable concentrations. Both  $\alpha$ -HCH and  $\delta$ -HCH were detected in only one sample at concentration from below detection limit to 2 and 8  $\mu\text{g/g}$  w/w, respectively.

In most cases HCH technical mixtures used for pest control are composed of 55 – 88% of  $\alpha$ -HCH, 5-14%  $\beta$ -HCH, 8 – 15%  $\gamma$ -HCH and 2 – 16%  $\delta$ -HCH isomers [16]. Among the HCH isomers,  $\beta$ -HCH has been reported to have a lower microbial degradation rate and higher persistence in mammalian tissues than the other isomers since it resists enzymatic degradation [17]. Absence of  $\gamma$ -HCH is expected since that is the isomer which is very easily metabolised to simpler substances [16]. However, detection of  $\alpha$ -HCH and  $\delta$ -HCH isomers without  $\beta$ -HCH was contrary to the expectation. More samples and investigation are required to give plausible explanation on these observations.

#### D. Other Organochlorine Pollutants

With the exception of DDTs, other organochlorine residues were seldomly detected in *sifa* samples at very low or trace levels. HCB was measured in 44% of analysed samples with concentrations ranging from below detection limit to 25  $\mu\text{g/g}$  w/w. The only cyclodiene pesticides detected in this study is aldrin that was found above detection level in only one sample. It was measured at concentration of 2  $\mu\text{g/g}$  w/w.

#### E. Methoxylated Brominated Diphenyl Ethers

Methoxylated brominated diphenyl ethers is another group of persistent organic compounds that was constantly detected in all *sifa* samples. The measured organobromines include 6-methoxy-2,2',4,4'-tetraBDE (6-MeO-PBDE-47) and 2' methoxy-2,3',4,5'-tetraBDE (2'-MeO-PBDE68) as presented in Fig. 2 and their concentrations in individual samples are presented in Fig. 3. Levels of 6-MeO-PBDE-47 ranged from 0.1 to 7.3  $\mu\text{g/g}$  w/w with mean concentration of 2.64  $\mu\text{g/g}$  w/w (median 0.9 and SD 3.13). The other organobromine, 2'-MeO-PBDE68 was measured at significantly ( $p = 0.0064$ ) higher concentrations than 6-MeO-PBDE-47 with mean concentration of 20.8  $\mu\text{g/g}$  w/w (median 10.7 and SD 20.1) ranging between 0.7 to 51.3  $\mu\text{g/g}$  w/w. Further screening using GC-MS of samples indicated presence of dimethoxy species of the brominated compound but they were not quantified as there were no reliable standards to use.

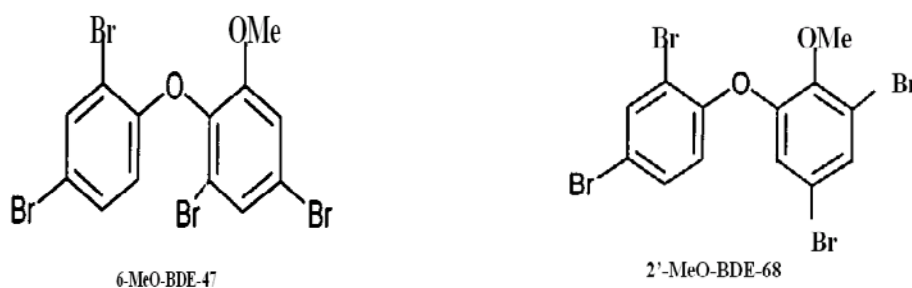


Fig. 2 The two MeO-BDEs compounds measured in *sifa* samples

Radiocarbon investigations of the MeO-PBDEs have shown that the brominated compounds are of natural origin [18, 19] and not associated with the metabolism of anthropogenic polybrominated diphenyl ether (PBDE) fire-retardants. Detection of MeO-BDEs in archived whale oil, which pre-dates the industrial manufacture of PBDE fire retardants, was an additional evidence of their natural origin [20]. MeO-BDEs have been found as natural products in marine sponges [21], and green and red algae [22].

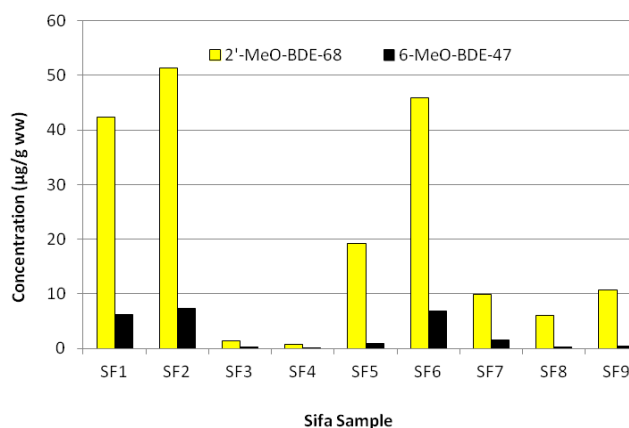


Fig. 3 Variations of 2'-MeO-PBDE68 and 6-MeO-PBDE47 compounds

Both 6-MeO-BDE-47 and 2'-MeO-BDE-68 have been widely reported in different marine organisms [23] and shown to originate from the same sources as their concentrations gave close correlations [24]. Interestingly, in this study, the compounds gave same trend of correlation with value of  $R^2 = 0.952$  (Fig. 4). This correlation indicates that the two compounds resist microbial degradation to the same extent.

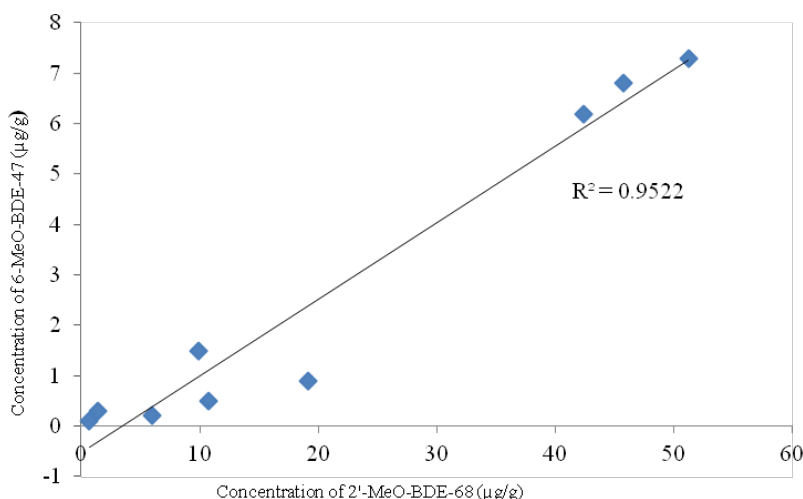


Fig. 4 Correlations of two brominated compounds

Using correlation technique, the compounds 6-MeO-BDE-47 and 2'-MeO-BDE-68 measured in dolphins from Zanzibar have been shown to originate from the same sources [24]. However the potential sources of these compounds are yet to be identified. Detection of MeO-BDEs in *sifa* samples is a clear indication that the compounds are very stable and resistant to microbial degradation. The two brominated compounds MeO-BDE47 and MeO-BDE68 that are presumably of natural origin have been found to bioaccumulate to very higher concentrations more than that of anthropogenic origin [24]. Although little is known about the biological role of these biogenic compounds, researchers are concerned about possible health effects, pointing to their structural similarity with PCBs that may likely result to similar toxic effects and endocrine disruption properties [25]. As there were many unidentified brominated residues further investigation is required to reveal their identity and concentrations.

#### F. Comparison with Related Studies from the Study Area

In comparison, the levels of both organochlorines and brominated compounds found in this study are within the lower range of the levels reported in dolphin from the same area [12]. The levels were much lower than those reported in dolphins caught off the eastern coast of South Africa and other parts of the world [26]. This might be attributed to the difference in nature of samples used between the studies. Whereas this study used *Sifa* from decayed samples which are normally characterised by low pollutants load due to microbial degradation, samples used in the other studies were fresh dolphin's samples.

On the other hand, findings in this study differed from other studies worldwide on types of POPs detected in the samples. No pollutants of industrial origin were detected in this study while the other study reported very high pollutant levels of PCBs, dioxins and furans [27]. Absence of industrially generated pollutants indicates that Zanzibar marine environment is relatively free from industrial pollution.

### G. Fishermen Perceptions of Sifa

Oral interview administered to 9 fishermen using *sifa* reveals that they are using *sifa* as both antifouling and coating material which reduces contact between boat surface and water. It is also used to seal minor openings between two connected pieces of timber on boat surfaces. Regarding the type of materials used for preparation of *sifa*, about 89% (N=8) of the respondents commented that high quality *sifa* is obtained by using fat rich internal organs of carnivorous fish. This provides plenty of oil that ensures effective coating of boat surfaces.

It was observed that all the fishermen were not aware that fat-rich internal organs of big carnivorous fish accumulate high levels of persistent organic pollutants. About 44% (N=4) of respondents suggested that antifouling properties of *sifa* were originated from smell given off by *sifa* and the remaining portion of respondents don't know what causes antifouling effect of the *sifa*.

### H. Environmental Risks Associated with Use of Sifa

Use of *sifa* from decayed internal organs as an antifouling agent to prevent vessel from being damaged by barnacles can have both positive and negative impact on environment and human health. On one hand, use of local antifouling has reduced arguent need of using booster chemical that has been shown to carry detrimental health and environmental impact. On the other hand, *sifa* carries a significant amount of toxic persistent pollutants that may constitute risks to users and their marine environment. Use of *sifa* implies that fishermen and marine water are directly exposed with contaminated material. Although there is no direct ingestion of pollutants from *sifa* by humans and the only contamination route is through passive uptake, that fishermen are using *sifa* without prior knowledge of its chemical content may be considered as potential risk. Once *sifa* is applied on the hull of vessels, it can be easily washed into water bodies. However such small concentration may be inadequate to cause acute toxicity.

## IV. CONCLUSION

Local antifouling agent made up of internal organs of dolphins from Zanzibar contains POPs from both anthropogenic (organochlorines) and biogenic (MeO-BDEs) sources, indicating that the compounds are quite stable to degradation during the decay process. However, the levels were much lower and less diverse in *sifa* than the levels in fresh samples. Preponderance of DDT metabolite (p,p'-DDE) over the parent molecule demonstrates existence of aerobic microbial degradation during the decay processes. The correlations of the two MeO-BDEs suggest that the biogenic compounds are originated from the same source and have similar degradation stability. This study reveals fishermen's lack of knowledge of the presence of chemical contaminants in *sifa* although the levels are too low to pose acute potential risks to users. Based on the findings from this study, it is recommended that the origin of antifouling properties and the possible long-term toxicological consequences of the compounds reported in *sifa* be investigated.

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