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Contamination of the Water Environment in Malaria Endemic Areas of KwaZulu-Natal, South Africa by DDT and Its Metabolites

B. L. Sereda,[1] H. R. Meinhardt[2]

[1] Agricultural Research Council, Plant Protection Research Institute, P/Bag X134, Pretoria, Queenswood 0121, Republic of South Africa
[2] South African Bureau of Standards (SABS), Testing and Conformity Services (Pty) Ltd. P/Bag X191, Pretoria 0001, Republic of South Africa

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Malaria has been a major public health problem in many countries. Since the discovery of the chlorinated hydrocarbon insecticide DDT, chemical control has become the preferred control method in most malaria vector control programmes. However, it has been shown that DDT can enter the environment and persist at significant levels despite having been banned for decades (Blus et al. 1987; Young et al. 1994; Sarkar et al. 2003; Wang et al. 2003). The degradation of DDT is dependant primarily on the evolution production of its metabolites DDE and DDD, which are both very persistent (Chiu et al. 2004). DDT is reported to be exceptionally stable in soil and the aquatic environment (Reich et al. 1986). Ecological toxic effects of organochlorine pesticide residues, and DDT in particular, have been shown in their ability to disrupt functions of, for instance, the endocrine system in biota (Raloff 1984). The implications of these compounds in the environment are related to bioaccumulation and biomagnification in food chains as well as their presence in biological tissues (Ludwicki and Goralczyk 1994, Newsome et al. 1995, Barkatina et al. 2002, Waliszewski et al. 2003). DDT is one of the 12 Persistent Organic Pollutants (POPs) under the Stockholm Convention (UNEP 2003). Because of its persistence and proven long range transport from the source of origin as well as potential impacts on environmental human health, use of DDT is restricted to the permitted until such time that an effective alternative control method has been developed.

In South Africa, DDT was banned from agricultural use during the 1970s, but still remains in use for residual spraying of dwellings for the control of *Anopheles* malaria vectors in the malaria endemic areas of KwaZulu-Natal (KZN), Mpumulanga and the Limpopo Province (Sharp et al. 2000).

The two malaria endemic districts of Ubombo and Ingwavuma in KZN are of particular interest due to the detection of high levels of agrochemical contamination in the water environment (Sereda and Meinhardt, 2003). In addition, there has been a corresponding geographical shift of high-risk malaria areas to control of malaria vector mosquitoes. Its use in malaria vector control will only be the Ingwavuma and Ubombo districts, as reported by Sharp et al. (2000). The phase-out of DDT and its replacement with pyrethroids initiated in 1996 by KZN authorities were not successful due the development of pyrethroid resistance in the malaria vector. As a result of this, a dramatic increase in malaria

Correspondence to: B. L. Sereda

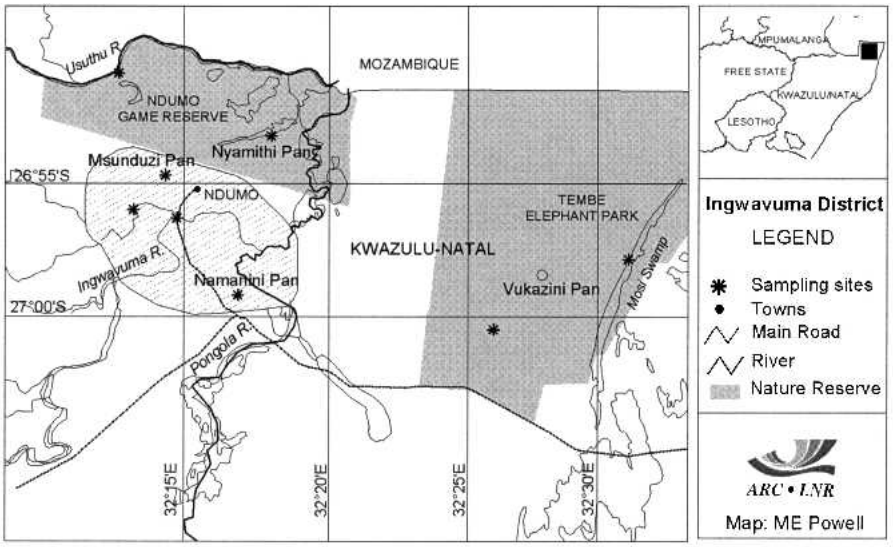

Figure 1. Map of the Ingwavuma district in the KZN, depicting sampling sites.

cases was observed and DDT was reinstated in 2000 (Sereda and Meinhardt. 2003).

Concern has been expressed in that DDT could potentially be introduced into water environment through runoff from DDT-sprayed areas, incorrect mixing procedures and the potential illegal use thereof in agriculture. Furthermore, perpetual input of DDT and accumulation in the natural water bodies may be such, that selection for resistant mosquito individuals could occur (Bowman et al. 2000; Roberts et al. 2000), as cases of vector resistance to organochlorines have been reported from some areas in KZN *(*Sharp *personal communication).* It is thus imperative that the potential implications of vector resistance on the malaria control programme are considered seriously in future.

This study was undertaken to evaluate the potential contamination of natural water bodies by DDT and its metabolites in the KZN malaria endemic areas.

The study further intended to predict the potential risk of insecticide resistance development in malaria vectors.

MATERIALS AND METHODS

The study area is situated in the Ingwavuma (Fig. 1) and Ubombo (Fig. 2) districts of KZN (South Africa). Sampling sites were located at the Makhathini Flats, Ndumo and Ophansi. The Ndumo Game Reserve and Tembe Elephant Park were selected as reference areas (Fig. 1) as malaria spraying does not occur here.

Water and sediment samples were collected for residue analysis from the shallow ends of water bodies where mosquito larvae are expected. Samples were collected

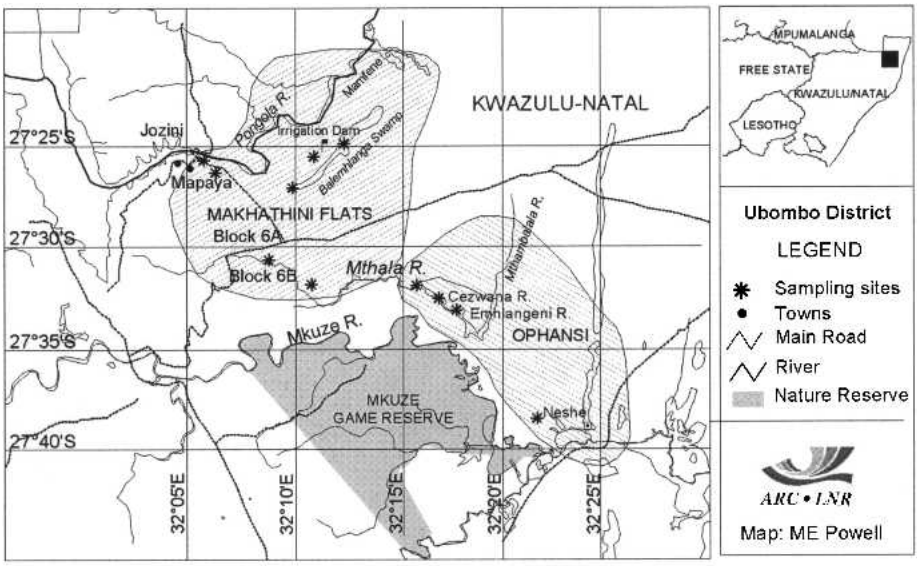

Figure 2. Map of the Ubombo district in the KZN, depicting sampling sites.

on 4 occasions during a 14-month study period. Sampling events were timed to cover the period before, at the beginning and during the malaria-spraying season (November to March) (KZN Malaria Review 1998). Sampling events took place during July/September 2000, November 2000, February 2001 and September 2001, respectively. A total of 178 samples were collected from the study area and 35 samples from the reference areas.

Samples were analysed for residues of DDT and its metabolites p,'p-DDE, o,p'-DDE, p,p'-DDD and o,p'-DDD, referred to as total DDT (ΣDDT). Insecticide residue analyses were conducted according to the procedures based on a multi-residue extraction method (Manual of Pesticide Residue Analysis, DFG 1987).

Samples were collected using a modified soil auger containing a removable metal sample cartridge. The cartridge was pushed into the sediment layer using a T-handle. The cartridge containing the sediment was then removed and sealed with foil lined lids in the field. Water was sampled from the top 20cm water layer directly into 4 litres amber glass bottles.

Where the sampling was done at a distance from the side a rope was fixed to the bottle, and the bottle thrown into the water body, filled and recovered. Samples were stored in cool room at 4°C and 1 litre of water and 200g of sediment were taken for the residue analysis.

The concentrations of insecticides (mean values of duplicate analyses) are expressed in ng/kg for sediment samples and ng/L for water samples. Residue levels were calculated on a wet mass basis and corrected for recoveries, which were ranged from 70 to 100%. Samples were analysed quantitatively using gas chromatography (GC) fitted with an electron capture detector (ECD) and

confirmed using GC- mass spectrophotometry (MS) technology. The limits of detection for DDT and its metabolites were in the range of 0.0003 µg/kg for sediment and 0.00006-0.00007µg/L for water.

RESULTS AND DISCUSSION

Results of analysis show that residues of DDT and its metabolites were present in both the study and reference areas (Figs. 1 and 2). DDT residues not metabolized were detected in 25.8% of samples collected from study area and in 8.6% of the reference samples. The DDT metabolites DDE and DDD were detected in 83.1 % of the samples collected from the study area and in 54.3% of samples from the reference area (Table 1).

Frequency data indicates that all the study sites in the two districts were equally polluted. In Ophansi and Ndumo areas 100% sediment samples were contaminated with metabolites of DDT, while sediment samples collected from Makhathini were contaminated in 94.7% (Table 1).

ΣDDT was detected primarily in sediment samples (Table 1) at concentrations ranging from 1.0 to 13738.0 ng/kg (Table 2) as the metabolite DDE, with the highest concentration detected at 10952.0 ng/kg. In water ΣDDT was found at lower concentrations ranging from 0.2 to 161.0 ng/L.

The highest concentrations of ΣDDT were detected during the September 2001 sampling event in the Makhathini Flats (13738.0 ng/kg) and Ophansi (10855.0 ng/kg) (Table 2).

Table 1. Frequencies of water and sediment samples containing DDT and its metabolites.

Locality		Matrix	No. collected	% containing DDT[1]	% containing metabolites[2]
Study area	Makhathini	W	36	11.1	66.7
		S	57	24.6	94.7
	Ophansi	W	21	14.3	61.9
		S	36	41.7	100
	Ndumo Agric	W	16	37.5	56.2
		S	12	33.3	100
Total for study area		W&S	178	25.8	83.1
Reference area	Tembe	W	7	0	42.8
		S	14	14.3	50
	Ndumo Game Park	W	6	16.6	66.7
		S	8	0	62.5
Total for reference area		W&S	35	8.6	54.3

[1]Samples containing DDT, not decomposed into metabolites, [2]Samples containing metabolites of DDT (DDE and DDD)

The sampling event occurred just before the malaria spraying was initiated, but during peak agricultural activities. These results may indicate that the source of the DDT may be from illegal agricultural use of the compound.

Results of analyses of samples from the reference areas Ndumo Game Reserve and Tembe Elephant Park, (located on the Mozambique border), indicated that these areas were not free from DDT contamination. Total DDT detected in reference sediment samples ranged from 3.0 to 437.0 ng/kg, and in water samples from 2.0 to 51 ng/L, respectively. A maximum total DDT residue level of 437.0 ng/kg was detected in a sediment sample from Tembe Elephant Park (Table 2). As DDT spraying or dumping is not conducted in the parks and as the area is free from agricultural crop production, it is believed that the contaminants found in the Game Parks could have been transported via long-range transport in water.

The residue levels detected in water are comparable with ΣDDT concentration ranges found in Kenya (Lalah et al. 2003), India (Sarkar et al. 2003), Bangladesh (Das and Das 2004) and China (Wang et al. 2003). The residue concentrations of water samples from the Makhatini study were higher than the EC/WHO limit for drinking water (100 ng/L; Lalah et al. 2003).

Table 2. Concentration range of residues of DDT, its metabolites (DDE and DDD), and ΣDDT detected in the KZN.

Locality	Matrix	Concentration range of p,p'- and o'p' isomers of			
		DDT	DDE	DDD	Σ DDT
Makhathini	W[1]	1.9-8.0	0.2-138.0	0.2-15.0	0.2-161.0
	S[2]	5.0 – 134.0	6.0 –10952.0	1.0 – 1433.0	1.0 –13738.0
Ophansi	W	1.0-3.0	0.3-4.0	0.1-5.0	0.5-6.0
	S	10.0 - 7846.0	1.0 – 554.0	1.0 – 1697.0	14.0 -10855.0
Ndumo	W	2.0-9.0	0.2-2.0	0.4-4.0	1.0-12.0
	S	26.0 – 323.0	2.0 – 1596.0	11.0 – 405.0	90.0 – 2173.0
Tembe	W	ND[3]	1.0-5.0	2.0-12.0	2.0-51.0
	S	22.0 – 437.0	3.0 – 21.0	2.0 – 38.0	3.0 – 437.0
Ndumo Game Park	W	3.0	19.0	0.1-24.0	2.0-51.0
	S	ND	3.0 – 13.0	7.0 – 44.0	17.0 – 47.0

[1]Water ng/L, [2]Sediment ng/kg; [3]Not Detected

The highest ΣDDT concentration in sediment detected in this study is two orders of magnitude higher than those found in Kenya (Lalah et al. 2003) and Bangladesh (Das and Das 2004).

These results show that ΣDDT residue levels in sediment are higher than those detected in water, and support the conclusion of Nowell et al. 1999 that organochlorines have the tendency to accumulate in sediment.

In addition to the results reported on in this paper on organochlorine contamination, a parallel investigation performed in the KZN, showed the contamination of the water environment with pyrethroids, organophosphates and carbamates used in local agriculture (Sereda and Meinhardt, 2003).

The presence of DDT and metabolites raises concern with regard to the development of malaria vector resistance towards DDT and agricultural insecticides in the study area. Generally, such insecticide resistance is expected to directly affect the occurrence of malaria, which may threaten the sustainability of the malaria control programme such as discussed by Roberts et al. (2000).
Many countries are faced with malaria vectors that are resistant not only to DDT, but also to other insecticides, possibly as a result of previous selection pressure arising from a pesticide-contaminated environment. According to the WHO (1992), insecticide resistance has been documented in more than 100 species of mosquitoes. The situation in KZN is alarming due to the fact that some vector species resistant to pyrethroids (Hargraves et al. 2000) and other insecticide chemical groups have already been detected (Sharp *personal communication*).
To complicate the situation even further, cross-resistance between chemical groups is possible. The phenomenon of cross-resistance between DDT and pyrethroids has been reported for mosquito species in other international malaria areas (Malcolm 1988a; Omer et al. 1980; Amin and Hemingway 1989; Sharma 1999). Miller (1988) confirmed similarities between pyrethroids and DDT. Both have negative temperature coefficients of toxicity and dual effects on insects – an initial rapid knockdown (*kd*) and a subsequent lethal effect. Knockdown resistance (*kdr*) induced by selection with DDT confers inherent cross-resistance against the *kd* effect of pyrethroids, and vice versa.
Roberts (1994) stated that both insecticidal and behavioural effects of insecticides on insects are important, but the relative importance of one versus the other is controversial. Implications are that future use of DDT for malaria vector control could be compromised because pyrethroid insecticides used could stimulate the avoidance behaviour in arthropods.

The extensive organochlorine contamination detected in the KZN may be due to prior use of DDT, the persistence of the chemical in the environment, and/or its current use for malaria vector control. These findings are disturbing as they indicate the necessity for improving the judicious use and control of DDT. DDT contamination may impact on the water environment in malaria endemic areas. DDT may pose a serious threat not only to human and environmental health directly, but also to human health and social development indirectly, through potentially threatening the malaria vector control programme. Further studies are required to determine the process of selection for resistance in malaria vectors in the study areas.

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