Bull. Environ. Contam. Toxicol. (2005) 75:530–537

DOI: 10.1007/s00128-005-0784-y

Bulletin of Environmental Contamination and Toxicology

Contamination of the Water Environment in Malaria Endemic Areas of KwaZulu-Natal, South Africa, by Agricultural Insecticides

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

Received: 10 October 2004/Accepted: 18 July 2005

Many tropical diseases, transmitted by arthropod vectors are associated with water. Likewise, the three stages of the malaria mosquito life occur in water. Water also plays an important role in the economic development of rural communities. Typically, the intensification of agriculture leads to an increased water use, especially for irrigation, thereby increasing the dependence of the community on water for its sustainability.

The Ubombo and Ingwavuma districts in KwaZulu-Natal province (KZN) of South Africa, have undergone major agricultural developments which have been concentrated in the Makhathini Flats, Ndumo and Ophansi areas. Furthermore government support for the emerging farming sector has resulted in increased agricultural use of insecticides in these areas. These areas are also malaria endemic and therefore under protection of the malaria vector control programme. The malaria vector mosquitoes are controlled primarily through indoor spraying of the insecticides DDT and deltamethrin in dwellings.

It has been shown internationally that pesticides used in agricultural pest management may enter the environment and this could lead to contamination of soil and water by these chemicals (Yousefi 1999). Once a chemical has entered the environment, they are degraded along different routes, depending on the type of chemical involved, the soil type and environmental conditions. The specific environmental conditions prevailing in an area of pesticide use will thus determine the extent of persistence and transport of the contaminants in the environment.
Results of earlier investigations in the Northern KwaZulu-Natal districts of South Africa indicates considerable lack of knowledge, regarding the selection of suitable agricultural insecticides, correct mixing of products and application methods amongst pesticide users in this area (Bouwman et al. 2000; Rother 2000). The authors also found pesticide spray mixtures are prepared adjacent to natural water bodies such as, dams, pans, ponds, and streams. In addition crop fields are made on the banks of these water bodies. Run-off from irrigation or rain from the fields is transported to these water bodies. This is particularly problematic at the shallow ends of water bodies, as pesticides contained in the run-off will end up in

Correspondence to: B. L. Sereda

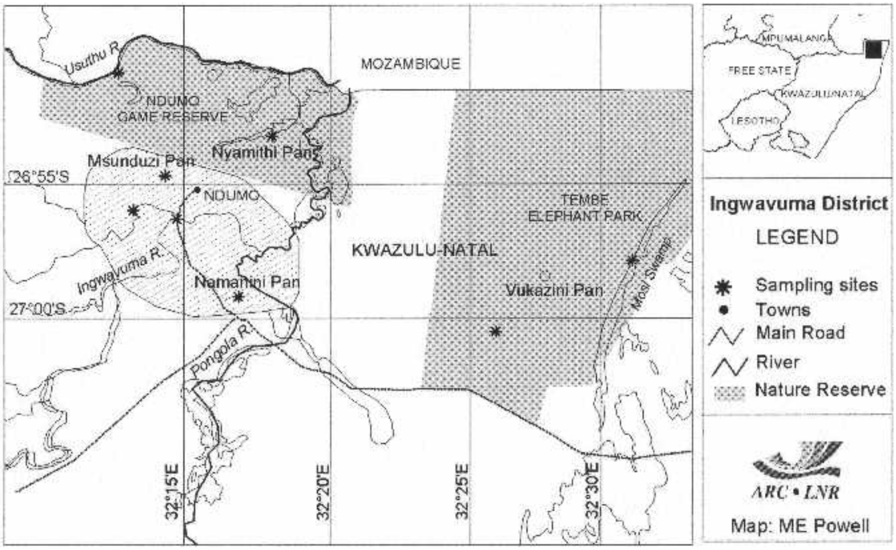

Figure 1. Map of the Ingwavuma district in the KZN, depicting sampling sites.

the water and sediments, possibly contributing to selection for insecticide resistance in mosquito larvae.

Agricultural insecticides are presumed to have contributed to selection for insecticide resistance in many populations of Anopheline mosquitoes (Malcolm 1988; Lines 1988; Georghiou 1990). In South Africa, it has been shown that *Anopheles funestus* from Ndumo exhibited resistance to pyrethroids (Hargreaves et al. 2000). In addition, mosquitoes collected from Makhathini Flats have shown signs of resistance to organophosphates and carbamates (Sharp, *personal communication).*
The study was initiated in order to determine the current situation regarding pesticide use and pollution potential in the study area. The main aim of the project was to establish the risk potential for the development of insecticide resistance in malaria mosquitoes due to the agricultural use of these compounds.

MATERIALS AND METHODS

The study was undertaken in the rural areas of Northern KZN, in two districts namely Ubombo and Ingwavuma. The study sites were located in the Makhathini Flats, Ndumo, and Ophansi areas, whereas the Ndumo Game Reserve and Tembe Elephant Park, both free from pesticides use, were selected as reference areas (Fig 1 and 2). The Makhathini study site is located inside an irrigation scheme, while Ophansi and Ndumo areas depend on natural sources of water only.

The crops cultivated in the study area include rain fed cotton (on limited fields, in comparison with the previous seasons), sugar cane, maize, rice and vegetables.

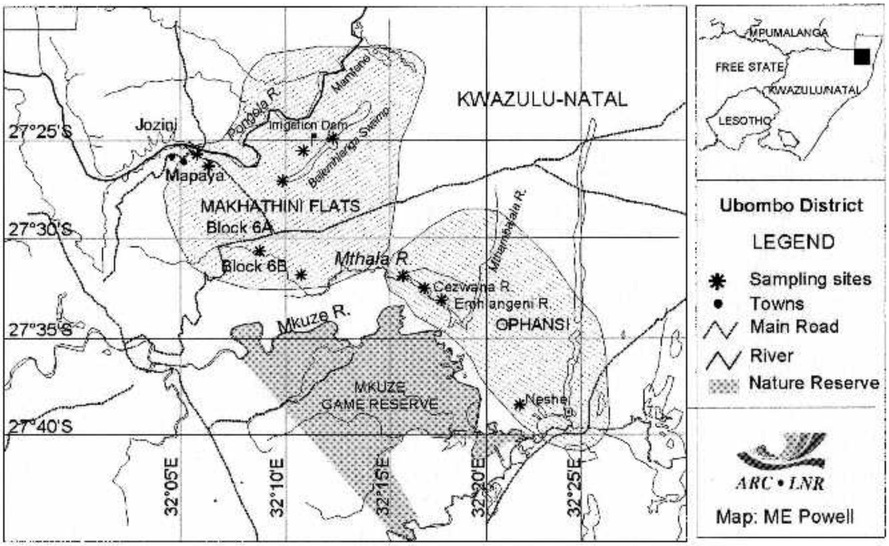

Figure 2. Map of the Ubombo district in the KZN, depicting sampling sites

Data collected during interviews with farmers, Co-ops and extension officers showed that insecticides from all chemical groups are used extensively in the study area. From a list of pesticides used, the most frequently used compounds were selected for analysis. Analysed insecticides included pyrethroids (deltamethrin, cypermethrin, cyfluthrin and permethrin), carbamates (carbaryl, carbosulfan, carbofuran) and organophosphates (fenthion, fenitrothion, demeton SM, dimethoate, monocrotophos and methamidophos).

Water and sediment samples were collected from the shallow ends of water bodies where spillage of insecticide, as well as mosquito larvae, were expected.

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. The samples were collected from the edges of the water bodies up to a maximum of 10 m from the side. 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 sampling events were timed to cover the agricultural spraying season and took place during July to September and November 2000, as well as February 2001. A total of 104 samples were collected from the study sites and 33 samples from the reference sites.

Insecticide residue analyses were conducted according to the procedures based on a multi-residue extraction method (DFG 1987). Pyrethroids were analysed using a GC fitted with an Electron Capture Detector (ECD), and carbamates were analysed using a GC fitted with Nitrogen Phosphorous Detector (NPD) and Flame Ionisation Detector (FID) detectors. Organophosphates were analysed qualitatively only using a GC fitted with a Flame Photometric Detector (FPD). In addition, samples from the September 2000 and February 2001 sampling events were analysed qualitatively using GC Mass Spectrophotometry (MS). Residue levels were calculated on sample wet mass basis and solvent recovery taken into account.

Analyses were performed in duplicate. Recoveries ranged from 70 to 100% for organophosphates and carbamates and 55-65% for pyrethroids. The limits of determination for sediment and water ranged as follows: for pyrethroids were 0.003 μg/kg and 0.0006 - 0.0007 μg/L, 0.0003-0.0010 μg/kg and 0.08-0.30μg/L for carbamates and 0.001-0.035 μg/kg and 0.0002-0.0013 μg/L for organophosphates.

RESULTS AND DISCUSSION

Results of analyses performed during the course of the study show agricultural insecticide contamination of the water environment in the study area. All samples collected contained at least one of the target insecticides, representing all three the chemical groups of interest.

Generally, it may be expected that in areas where the level of knowledge on pesticide use and pesticide application is low, the extent of environmental pollution by these compounds may be high. This could be the case where extensive agricultural development initiatives are implemented in rural areas as part of poverty alleviation initiatives. It can be concluded that the emergent agricultural areas in KZN fall into this category.

Pyrethroid residues are said to decompose rapidly under field conditions, as they are very sensitive to photo- and thermal decomposition (Agnihotri et al. 1986). Despite these properties, residues of pyrethroids were detected in 27.3% – 73.3% of samples collected at Makhathini Flats, 42.3% at Ophansi, and 20% - 72.7% at Ndumo agricultural area (Table 1). The highest residue levels of pyrethroids were found in sediment samples collected during the February 2001-cotton spraying season (Table 2). Residues of the pyrethroid cypermethrin (1651.2 μg/kg) (Table 2) and deltamethrin (90 μg/kg) (data not shown) were detected primarily in the Makhathini Flats. The results indicate the presence of cyfluthrin (467.3 μg/kg) in the water system at Tembe Elephant Park, which was unexpected as no pesticides are used in close proximity to this area. Cyfluthrin and cypermethrin were detected most frequently in the study area. The incidence of pyrethroids indicates a high usage rate of these compounds in the study area.

Organophosphate residues were detected in 43.3 – 77.3% of the samples collected at Makhathini Flats, in 18.7 – 80% of samples from Ophansi, and 36.4 – 40.0% of samples collected at Ndumo (Table 1).

Table 1. Frequency of insecticides residues (water and sediment samples) detected in study and reference areas.

Locality	Mx[5]	Total No collected	Samples containing pyrethroids		Samples containing organophosphates		Samples containing carbamates	
			No	%	No	%	No	%
Study area								
Makhathini	w[1]	22	6	27.3	17 (9)[2]	77.3	10(2)	45.4
	s[3]	30	22(8)	73.3	13(2)	43.3	21(3)	70
Ophansi	w	10	ND[4]	ND	8(2)	80	1	10
	s	16	11(2)	42.3	3	18.7	6	37.5
Ndumo Agric.	w	15	3	20	6	40	3(2)	20
	s	11	8(1)	72.7	4	36.4	3(1)	27.3
Total for study area	w&s	104	50 (10)	48.1	51(14)	49.0	44(8)	42.3
Reference area								
Tembe Park	w	7	1	14.3	4(1)	57.1	2(1)	28.6
	s	14	8(2)	57.1	1	7.1	7	50
Ndumo Game Park	w	5	1	20	4	80	ND	ND
	s	7	2(1)	28.6	1	14.3	5(4)	41.7
Total for reference area	w&s	33	12(2)	36.4	10(1)	30.3	14(5)	42.4

[1]Water, [2]Values in brackets indicate the number of samples containing more than one pesticide, from the same chemical group, [3]Sediment, [4]Not detected, [5]Matrix.

Carbamate residues were detected in 45.4 - 70% of the water and sediment samples collected at Makhathini area, 10.0 – 37.5% samples from Ophansi and 20.0 – 27.3% of the samples collected at Ndumo (Table 2). The highest concentration of carbamates (carbaryl 50.1 µg/kg) was found in February 2001 in sediment collected from Ndumo agricultural area.

The averages of the total frequencies of detection of residues for the three chemical groups of pesticides in the study area were similar. Forty eight percent of samples contained pyrethroids, 49% organophosphates and 42% contained carbamates (Table 1).

If one compares the study areas it is clear that the sites at Makhathini Flats were the most polluted by pesticides. This is also the area where most of the emerging vegetable production occurs. The two other study areas, Ophansi and Ndumo agricultural are characterised by less extensive agriculture and here a lower frequency of insecticide contamination was found.

In this study, pyrethroids and carbamates were detected more frequently in sediment while organophosphates were detected mainly in water (Table 1). The fate of a pesticide and the ability to move from the site of application is affected by the chemical and physical properties of the pesticide, the site characteristics such soil composition, vegetation, temperature, pH etc (Brown and Hock 1990). These factors must each be considered when determining the potential and intensity of a particular compound to adsorb, become transferred or degrade in the environment. Residue data indicates that the game parks Ndumo and Tembe Elephant Park, were contaminated by insecticides. The numbers of samples collected from these reference areas were limited due to the difficulty of collection. However, of the 33 samples collected in game parks 36.4% contained pyrethroids, 30.3% organophosphates and 42.4% contained contaminated carbamates (Table 1). The presence of residues at Ndumo Reserve could possibly be due to the Parks close proximity to agricultural fields on the banks of the rivers feeding into the Ndumo Game Reserve. Tembe Elephant Park on the other hand is void of agricultural surrounds and the origin of pesticide residues within the park remains somewhat of a mystery. It is possible that these residues were transported into the Park from a distance via underground waterways flowing in from the Makhathini area.

An attempt to analyse the seasonal fluctuation of insecticide contamination in samples collected from study as well as reference areas proved fruitless. This is more or less in line with what was found by Lines (1988). It is known that the numbers of factors influencing the contamination of the water environment in the study area are substantial. Generally it was found that the constant and high usage of pesticides in the study area leads to extensive pollution of the environment.

Table 2. Concentration range of pyrethroid and carbamate residues detected in samples collected in study and reference areas.

Locality	Samples containing pyrethroids		Samples containing carbamates	
	w[1]	s[2]	w	s
Study Area				
Makhathini	< 0.0006-40.7	0.01-1651.2	0.12-2.39	0.36-13.23
A.I.[3]	Cypermethrin	Cypermethrin	Carbaryl	Carbosulfan
Ophansi	ND[4]	<0.003-260.84	1.67	3.89-9.95
A.I.	ND	Cyfluthrin	Carbofuran	Carbofuran
Ndumo Agric.	23.19	30.63-350.00	0.6-2.5	3.71-50.1
A.I.	Cypermethrin	Cypermethrin	Carbaryl	Carbaryl
Reference Area				
Tembe Park	< 0.0006	0.07-467.3	0.35-1.39	3.26-85.31
A.I.	ND	Cyfluthrin	Carbofuran	Carbosulfan
Ndumo Reserve	< 0.0006	0.02	ND	1.89-50.33
A.I.	Cyfluthrin	Cyfluthrin	ND	Carbosulfan

[1]Water µg/L, [2]Sediment µg/kg, [3]Active ingredient detected at highest concentration, [4]Not detected

Many authors have shown that agricultural chemicals select for resistance in *Anopheles* species (Hemingway et al. 1986; Lines 1988; Malcolm1988; Hemingway et al. 1998). Georghiou (1990) found that more than 90% of all insecticides produced have been used for agricultural purposes, particularly in rice and cotton, and this use has created serious problems in mosquito control programmes. The author suggests that insecticide resistance in 17 species of mosquitoes in various countries occurred because of indirect selection pressure from agricultural pesticides.

Roberts (1994) concludes that extensive use of pyrethroids in agriculture and following environmental contamination causing an increase in the level of resistance, may preclude the long-term use of these chemicals for the control of vector-borne diseases. Resistance to pyrethroids is in itself a matter of concern for malaria mosquito control, because pyrethroid treatment of bed-nets is currently a preferred alternative to conventional indoor spraying (Curtis 1998). Coetzee et al. (1999) stated that records of resistance to pyrethroids in *Anopheles gambiae* in West Africa raised concern about the future use of pyrethroids for malaria control in Southern Africa. Hargreaves et al. (2000) reported pyrethroid resistant *Anopheles funestus* mosquitoes in some sectors of the Ndumo area in KwaZulu-Natal. This particular species was eliminated from South Africa for four decades primarily due to DDT spraying. *Anopheles funestus* is more dangerous than its summer counterparts as it breeds all over in swamps, wetlands and water resources even during cooler winter months. Hargreaves et al. (2000) reported evidence of this species, responsible for malaria transmission, inside pyrethroid sprayed houses. It was the first time that *Anopheles funestus* had been shown to exhibit pyrethroid insecticide resistance. This situation is alarming because the genes for resistance to carbamates and organophosphates have been also detected in South Africa (Sereda and Meinhardt 2003).

It is believed that major selection pressure for the development of mosquito resistance exists in the study area. It is thus crucial to ascertain the relative contribution of the different insecticide classes to the development of resistance. The presence of pyrethroids, organophosphates and carbamates resistance is of great concern and demonstrates severe consequences involved in designing an efficient, malaria vector control programme for malaria endemic areas of South Africa.

Acknowledgments. The authors thank the Water Research Commission of South Africa, which sponsored this research, the staff of the KwaZulu-Natal Department of Health for their co-operation, and Ms M Powell for compiling the maps. Special thanks to Ms M Cloete and Mr A Havemann of the Pesticide Analytical Laboratory of the ARC, PPRI for sample analysis.

REFERENCES

Agnihotri NP, Jain HK, Gajbhiye VT (1986) Persistence of some synthetic insecticides in soil, water and sediment-Part 1. J Entomol Res10 :147-151

Bouwman H, Meinhardt R, Sereda BL, Cloete MM (2000) The risk of insecticide (pyrethroid) resistance for malaria control in South Africa. UNEP Proceedings. Workshop on the Management of Persistent Organic Pollutants (POPs), for the SADC region. Lusaka, Zambia. 14-16 February 2000

Brown CL, Hock WK (1990) The fate of pesticides in the Environment and Groundwater Protection. Agrochemical Factsheet 8, Penn State University Cooperative Extension

Coetzee M, Horne DWK, Brooke BD, Hunt RH (1999) DDT, dieldrin and pyrethroid insecticide resistance in African malaria vector mosquitoes: an historical review and implications for future malaria control in southern Africa. South African J Sci 95:215-218

Curtis CF, Miller JE, Hodjati M, Kolaczinski JH, Kasumba I, Denholm JA- Pickett (ed), Devonshire AL (1998) Can anything be done to maintain the effectiveness of pyrethroid-impregnated bednets against malaria vectors? Insecticide resistance: from mechanism to management. A discussion organized by Philosophical transactions of the Royal Society of London, Series B, Biol Sci 353:1769-1775

DFG-Deutsche Forschungsgemeinschaft (1987) Manual of Pesticide Residue Analysis. Pesticide Commission. VCH Publishers (UK) Ltd, Method S19: 383

Georghiou GP (1990) The effect of agrochemicals on vector populations. Roush R.T. Tabashnik BE, eds. Pesticide Resistance in Arthropods. New York and London: Chapman and Hall 183-202

Hargreaves K, Koekemoer LL, Brooke BD, Hunt RH, Mthembu J, Coetzee M (2000) Anopheles funestus resistant to pyrethroid insecticides in South Africa. Med Vet Entomol 14:181-189

Hemingway J, Jayawardena KGI, Herath PRJ (1986) Pesticide resistance mechanisms produced by field selection pressure on *Anopheles nigerrimus* an *An.culicifacies* in Sri Lanka. Bull WHO 64:753-758.

Hemingway J, Coluzii M, Bradley D (1998) Insecticide resistance in malaria vectors: a new approach to an old subject. The malaria challenge after one hundred years of malariology. Papers from the Mariology Centenary Conference, Academia Nazionale dei Lincei, Roma, 16-19 Nov 1998. Parasitologia - Roma. 4: 315-318

Lines JD (1988) Do agricultural insecticides select for insecticide resistance in mosquitoes, a look at the evidence. Parasit Today 4:517-520

Malcolm CA (1988) Current status of pyrethroid resistance in Anophelines. Parasit Today 4:513-515

Roberts DR (1994) Insecticide resistance issues in vector-borne disease control. American J Trop Med Hyg 50:21-34

Rother HA (2000) Influences of pesticide risk perception on the health of rural South African women and children. African News Occup Health Safety 10:42-46

Sereda BL, Meinhardt HR (2003) Insecticide contamination of the water environment in malaria endemic areas of KwaZulu-Natal. Water Research Commission Report No.1119/1/03

Yousefi VO (1999) Agrochemicals in South Africa. African Newsletter Occup Health Safety. 1-99