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The status of pesticide pollution in Tanzania

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Abstract

The paper summarises the findings of recent studies carried out to assess the levels of pesticide residues in water, sediment, soil and some biota collected from different parts of Tanzania. Although the intention is to cover the whole country, so far the studies have focused on areas with known large-scale pesticide use (Southern Lake Victoria and its basin, TPC sugar Plantations in Kilimanjaro region, Dar es Salaam coast, Mahonda–Makoba basin in Zanzibar) and a former pesticide storage area at Vikuge Farm in Coast region). Analysis of the cleaned extracts in GC-ECD/NPD revealed the dominance of organochlorines in all samples. Generally, low levels of residues were found in areas associated with agricultural pesticide use but the levels in the former storage areas were substantially high. DDT and HCH were dominant in all the studied areas. In the former areas, levels of $\sum$DDT in water, sediments and soil were up to 2 $\mu g\,L^{-1}$, 700 $\mu g\,kg^{-1}$ and 500 $\mu g\,kg^{-1}$, respectively, while those of $\sum$HCH were up to 0.2 $\mu g\,L^{-1}$, 132 $\mu g\,kg^{-1}$ and 60 $\mu g\,kg^{-1}$, respectively. The levels in aquatic biota were much higher than those in the water most likely due to bioaccumulation. In the former storage area at Vikuge the levels of pesticides in the topsoil were alarmingly high. Their concentrations were up to 282,000 mg kg^{-1} dry weight for $\sum$DDT and up to 63,000 mg kg^{-1} for $\sum$HCH. A herbicide, pendimethalin [*N*-(1-ethylpropyl)-2,6-dinitro-3,4-xylidine], was also found at concentrations up to 41,000 mg kg^{-1} dry weight. Thus the total pesticide content in the soil was almost 40%. Following these findings the area is now earmarked to be a demonstration site for a proposed GEF project 'Bioremediation of POPs impacted soils in East Africa'.

Keywords: Pesticide residues; Tanzania; Lake Victoria; Obsolete pesticides

1. Introduction

Tanzania, with an area of 945,067 km^2 and a population of about 34 million is, principally, an agricultural country with agriculture being the economic mainstay of the economy, employing over 70% of the population. Pesticides are used mainly in agriculture and public health. All pesticides used in the country are imported, although most are formulated in the country. Recent data on pesticide importation and consumption is scanty, with the statistics in published literature being at least a decade old. The per capita consumption of pesticides increased from 1/3 kg in 1977 to 1/2 kg per capita in 1988 [1]. Between 1989 and 1992 Tanzania imported an average of 14,000 tons of pesticides per year [1]. Insecticides, herbicides and fungicides account for over 90% of all pesticides used in the country, mainly on cotton, coffee, maize and paddy. Prior to the studies summarised in this paper, there had been occasional studies on pesticide residues in environmental samples and foodstuffs [2]. In the past 5 years, we have established a pesticide unit at the Chemistry Department, and studied pesticide residue levels in water, sediment/soil and biota in different parts of the country. So far, the studies have focused on areas with known pesticide use—Lake Victoria basin [3,4], sugar cane plantations in Kilimanjaro region [5], coastal areas of Dar es Salaam and Coast regions [6] and Zanzibar [7], and a former storage site at Vikuge [8–10]. There are further ongoing studies on the Rufiji Delta and sugar plantations in Morogoro region.

2. Materials and methods

Sampling and residue analysis including quality assurance measures followed essentially the procedures described by Åkerblom [11].

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2.1. Materials

All the reagents and solvents used were of analytical grade. Pesticide standards ordered from Dr. Ehrenstorfer GmbH (Ausburg, Germany) were used. Working standard solutions were made by diluting these stock standards and mixtures of standards of different concentrations were used in most cases for the screening of the pesticide residues. All glassware used had glass or Teflon stoppers.

2.2. Study areas, sampling and storage

Water, soil, and sediment samples were collected from Southern Lake Victoria basin (Mwanza and Mara regions), TPC sugar plantations at Arusha Chini, Kilimanjaro Region, Dar es Salaam and Coast Regions, Vikuge Farm, Coast Region and from Mahonda–Makoba basin in Zanzibar. Fish were collected from various landing sites of Southern Lake Victoria, while crabs were collected from mangrove swamps in Dar es Salaam. Water samples were collected in 1 L bottles and preserved with 10% NaCl and refrigerated until extraction. Sediment, soil and biota samples were kept in aluminium foils and frozen below −18 °C, while waiting for analysis.

2.3. Extraction

Unfiltered water samples (preserved with 10% NaCl), were extracted by liquid–liquid extraction (LLE) [11]. Each sample (1 L) was quantitatively transferred to a 1 L-separating funnel and the bottle rinsed with dichloromethane (30 mL), and combined with the sample in the separating funnel. The combined contents were then successively extracted with dichloromethane (3 × 50 mL). The organic layer was filtered through a plug of cotton wool topped with anhydrous sodium sulphate (ca 30 g) for drying. The combined extracts were concentrated in vacuo at 30 °C, and the final extract was made up in 2 mL cyclohexane:acetone (9:1 v/v), ready for GC analysis.

The soil samples (20 g in each case) were mixed with saturated ammonium chloride (14 mL) to open up the soil structures, swirled, and allowed to stand for 15 min. The mixture was extracted with cyclohexane:acetone (1:1 v/v, 100 mL) by shaking intermittently for 1 h, followed by extraction in ultra-sonic bath for 30 min, and renewed intermittent shaking for 2 h. The sample was allowed to settle, after which distilled water was added cautiously along the side until the organic phase rose to the neck of the flask. The organic phase was pasteur–pipetted into an E-flask, was dried with anhydrous sodium sulphate (ca 15–30 g) and finally filtered.

Sediment samples (10 g each) were mixed with anhydrous sodium sulphate (ca 30 g) and ground to form free flowing powder. The powder was extracted successively, using (50, 3 × 20 mL) of cyclohexane:acetone (1:1 v/v), by shaking for some time before extracting in an ultra-sonic bath for about 5 min. The combined, filtered extracts were shaken well with saturated sodium chloride (200 mL) and dichloromethane:cyclohexane (15:85 v/v, 50 mL). The organic phase was subsequently subjected to a clean-up process.

Blended biota tissues (from fish and crabs) (10 g in each case), were ground with sodium sulphate and cleaned sea sand to free-flowing powder. The powder was successively extracted with dichloromethane (50, 3 × 20 mL) and the solvent evaporated in vacuo, after which the extract was dissolved in cyclohexane (3.5 mL). Part of the extract (0.5 mL) was transferred to a tared vial and kept in a hood to evaporate the solvent. The fat weight was determined and percent fat in the sample was calculated.

2.4. Clean up

Water extracts in all cases were deemed sufficiently clean and were thus not subjected to clean up procedures. Each filtered soil and sediment extract was concentrated and the solvent changed to cyclohexane:ethyl acetate (1:1 v/v, 2 mL). The extract (1 mL) was cleaned up by size exclusion chromatography (SEC; Biorad SX-3, ethylacetate:cyclohexane 1:1 v/v as eluent), and the appropriate fraction was collected. This was later concentrated and the solvent changed to cyclohexane:acetone (9:1 v/v, 2 mL) for GC analysis.

The biota extracts were cleaned by SEC as above. In some cases the extracts were passed through Extrelut® disposable columns before being cleaned by SEC. The eluting solvent for the Extrelut® column was acetonitrile saturated with hexane.

2.5. Analysis and quantification

Varian Star 3400 and Hewlett Packard 5890A gas chromatographs equipped with ^{63}Ni electron capture (ECD) and nitrogen–phosphorous (NPD) detectors, and megabore and capillary columns (with non-polar and semi-polar stationary phases), were used for analysis. Nitrogen was used both as a carrier and make up gas in the ECD at a flow rate of 0.5–1 mL min^{-1} and 30 ± 1 mL min^{-1}, respectively. For NPD, helium was used as a carrier gas at a flow rate of 0.5–1 mL min^{-1}, and nitrogen as make up gas at a flow rate of 29 ± 1 mL min^{-1}. The temperature programme was 90 °C held for 1 min, 30 °C min^{-1} to 180 °C, 4 °C min^{-1} to 260 °C, where it was held for 12 min. The injector and detector temperatures were 250 and 300 °C, respectively. External reference standards were used for identification and quantification. Identity was confirmed by running on a second column with different polarity. Representative samples were subsequently analysed on a GC–MS for further confirmation of the results.

2.6. Method performance

Blank and recovery experiments were run for all the four matrices using standard methods [11]. For recovery

experiments, pesticide mixtures in acetone were added to the pesticide-free matrix. Spiked soil and sediment samples were stored cold overnight before extraction. In most cases the recoveries of the detected pesticides ranged between 70 and 120% (S.D. 5–20%), and when so, final results were not corrected for recoveries. Detection limits varied for the different pesticides and samples, but were in general 0.02–0.3 $\mu g\,L^{-1}$ in water, 0.1–1 $\mu g\,kg^{-1}$ soil (dry weight), 0.5–4 $\mu g\,kg^{-1}$ sediment (dry weight), and 0.3–2 $\mu g\,kg^{-1}$ fish (fresh weight). In many of the Vikuge samples the detection limits were substantially higher, since the extremely high content of some pesticides forced us to massively dilute the samples before analysing.

3. Results and discussion

With the exception of the former storage site at Vikuge, results revealed fairly low levels of pesticide residues, mainly organochlorines as shown in Tables 1–6. The concentrations varied between samples of different matrices, with the lowest levels detected, as expected (due to the hydrophobicity of most organochlorines) in water samples. However, there was a marked difference in levels between wet and dry seasons, with higher levels in the former. Alarmingly high levels were detected in soil and sediments collected from the former storage site at Vikuge, where, in 1986, a 170 m^3 'donation' of partially expired pesticides from Greece were stored in an open shed that eventually collapsed. Some pesticides were dominant in many samples while in other samples they were found to be below their average method detection limits.

3.1. Pesticide residues in water

Pesticides detected in water were DDT and its metabolites (DDD and DDE), HCH isomers and dieldrin (Table 1). These residues were dominant in the wet season samples while in the dry season most of the residues were below their average method detection limit.

Relatively high levels of total DDT in environmental samples can be related to past use of the pesticide in agricultural fields. Substantial amounts of pesticides have been reported to be used in horticultural fields along River Msimbazi in Dar es Salaam, Mwanza cotton farms as well as TPC sugarcane plantations. DDE and DDT were more dominant in wet season samples from Dar es Salaam indicating some elapse of time from application to sampling. DDE is an aerobic degradation product of DDT, and this degradation is assumed to take place in aerated soils. Thus it is suggested that DDE is transferred from treated soils to water bodies [12]. In Lake Victoria basin and TPC plantations, *p,p′*-DDT levels were dominant with lower levels of DDE and DDD, indicating more recent use of the technical pesticide. In all the water samples from the storage site at Vikuge, there were higher concentrations of *p,p′*-DDT than of its metabolites. Of the DDT metabolites detected from Vikuge samples, *p,p′*-DDD showed higher concentrations than *p,p′*-DDE, indicating the presence of DDD (rothane or TDE) in the 'donated' consignment [9].

Most of the environmental samples showed the domination of α- and β-HCH isomers implying the use of technical HCH rather than lindane. Likewise, results from storage site (Vikuge) indicated higher proportions of α- and β-HCH isomers in the total HCH, suggesting the presence of technical HCH in the stored consignment. Despite the fact that technical HCH has been banned from use in many countries including Tanzania [13,14], studies have indicated some current use of the product. Dieldrin was detected in samples from Dar es Salaam coast and TPC. Other pesticides such as endosulfan, aldrin and heptachlor were also detected from TPC at low concentrations with mean levels below 0.1 $\mu g\,L^{-1}$. Thiabendazole, carbosulfan and chlorpro-

Table 1
Levels of pesticide residues in water ($\mu g\,l^{-1}$) (mean, detection frequency)

Location	Water body	*N*	∑DDT	∑HCH	Dieldrin
Dar es Salaam coast	Rivers and marine	16	0.05–0.8 (0.4, 100%)	bdl	0.2–2.5 (0.6, 100%)
Lake Victoria and basin	Rivers and lake	11	bdl–1.5 (0.2, 27%)	bdl–0.2 (0.04, 27%)	bdl
TPC-Moshi	Rivers	13	bdl–0.08 (0.02, 46%)	bdl–0.03 (0.01, 38%)	bdl
Mahonda–Makoba basin, Zanzibar	Rivers and marine	11	bdl–1.4 (0.3, 45%)	bdl–1.1 (0.3, 91%)	bdl
Vikuge storage site	Pond	4	0.2–0.35 (0.2, 100%)	bdl–0.4 (0.2, 75%)	bdl
	Well	7	0.2–33 (9, 100%)	1–5.7 (1.7, 100%)	bdl

bdl: below detection limit.

Table 2
Levels of pesticide residues in sediments in dry season ($\mu g\,kg^{-1}$ dry weight) (mean, detection frequency)

Location	Sediments	*N*	∑DDT	∑HCH	Dieldrin
Dar es Salaam coast	Rivers and marine	12	7.9–57 (28, 100%)	bdl	bdl–45 (8.4, 66.7%)
Lake Victoria and basin	Rivers and lake	9	bdl–12 (2.0, 20%)	bdl	bdl
TPC-Moshi	Rivers	12	bdl–716 (204, 67%)	bdl–61 (14, 42%)	bdl
Mahonda–Makoba basin, Zanzibar	Rivers and marine	6	bdl–20 (4.1, 20%)	bdl–106 (34, 67%)	bdl
Vikuge storage site	Drainage ditch	n.a	n.a	n.a	n.a

bdl: below detection limit.

Table 3
Levels of pesticide residues in sediments in wet season ($\mu g\,kg^{-1}$ dry weight) (mean, detection frequency)

Location	Sediments	*N*	∑DDT	∑HCH	Dieldrin
Dar es Salaam coast	Rivers and marine	12	6.4–51 (18.2, 100%)	bdl–2.7 (0.6, 25%)	2.3–48 (9.4, 100%)
Lake Victoria and basin	Rivers and lake	9	bdl–705 (142, 78%)	bdl–131 (58, 78%)	bdl
TPC-Moshi	Rivers	12	bdl–444 (115, 67%)	bdl–57 (14.3, 67%)	bdl–7.2 (0.9, 17%)
Mahonda–Makoba basin, Zanzibar	Rivers and marine	9	bdl	bdl–68 (23, 80%)	bdl
Vikuge storage site	Drainage ditch	8	1×10^4–2×10^5 (5.4×10^4, 100%)	2.8×10^5–74×10^6 (2.4×10^6, 100%)	bdl

bdl: below detection limit.

Table 4
Levels of pesticide residues in soils during dry season ($\mu g\,kg^{-1}$ dry weight) (mean, detection frequency)

Location	Soil	*N*	∑DDT	∑HCH	Dieldrin
Lake Victoria and basin	Agricultural fields	40	bdl–20.4 (1.4, 65%)	bdl–3.4 (0.74, 59%)	bdl
TPC-Moshi	Sugarcane plantations	13	bdl–881 (112, 92%)	bdl–16 (5, 83%)	bdl
Mahonda–Makoba basin, Zanzibar	Rivers and marine	14	bdl–57 (17, 57%)	bdl–170 (45, 56%)	bdl
Vikuge storage site	In and around storage site	na	na	na	na

bdl: below detection limit.

Table 5
Levels of pesticide residues in soils in wet season ($\mu g\,kg^{-1}$ dry weight) (mean, detection frequency)

Location	Soil	*N*	∑DDT	∑HCH	Dieldrin
Lake Victoria and basin	Agricultural fields	13	bdl–97 (42, 100%)	bdl–59 (27, 63%)	bdl
TPC-Moshi	Sugar cane plantations	13	bdl–1146 (184, 92%)	bdl–37 (9, 69%)	bdl–14 (1.0, 8%)
Mahonda–Makoba basin, Zanzibar	Rivers and marine	19	bdl–19 (3.76,20%)	bdl–108 (11.3, 19%)	bdl
Vikuge storage site	In and around storage site	7	9.4×10^5–2.8×10^8 (7.3×10^7, 100%)	bdl–6.3×10^7 (2.1×10^7, 87%)	bdl

bdl: below detection limit.

Table 6
Levels of pesticide residues in biota ($\mu g\,kg^{-1}$ fresh weight)

Location	Biota	*N*	∑DDT	Dieldrin	∑Endosulfan
Lake Victoria	Fresh water Tilapia and Nile Perch	30	bdl–24 (3.4, 57%)	bdl	bdl–80 (9.6, 57%)
Coastal DSM	Coastal water fishes, crabs and fresh water fish	16	5.5–76 (22, 100%)	bdl–3.6 (0.5, 20%)	bdl

bdl: below detection limit.

fam residues were detected in surface waters from Vikuge at concentrations of up to 0.17, 3.1 and 1.5 $\mu g\,L^{-1}$, respectively. Thiabendazole at a concentration of 0.6 $\mu g\,L^{-1}$ was also detected in ground water samples. The higher levels and detection frequencies indicated by the wet season water samples were attributed to high levels of suspended particles in run off water from the source (applied fields or storage sites) since the samples were analysed without filtering. Most of the detected pesticides are highly hydrophobic and hence likely to be attached to suspended matter.

The levels detected in surface water were far below the maximum residue levels, (MRL) as set for water quality criteria for toxic and deleterious substances for coastal and marine water (50 $\mu g\,L^{-1}$ for DDT and 4 $\mu g\,L^{-1}$ for γ-HCH). However, the levels detected in drinking water (well water) near Vikuge (with mean levels of 9 and 1.7 $\mu g\,L^{-1}$ of DDT and HCH, respectively) were above the WHO and EU limits for drinking water quality. The limits set by EU for drinking water are 0.1 and 0.5 $\mu g\,L^{-1}$ for individual compounds and total compounds, respectively for all pesticides [15]. The MRL recommended by WHO for DDT and HCH residues is 1.0 $\mu g\,L^{-1}$ [16].

3.2. Pesticide residues in sediments

Pesticide residues found in sediments were the same as those detected in water samples and generally reflected a similar pattern. DDT, HCH and dieldrin were dominant in the samples, both in dry and wet seasons (Tables 2 and 3). Higher levels of the residues were found in sediments than in their corresponding water bodies, which is an indication of their hydrophobicity. The results indicated relatively high residue levels in the sugar cane plantations with mean levels

of up to 204 and 14 $\mu g\,kg^{-1}$ dry mass of total DDT and HCH, respectively, in the dry season (Table 2). The mean levels in the wet season were 115, 14 and 0.9 $\mu g\,kg^{-1}$ for DDT, HCH and dieldrin, respectively (Table 3). Surprisingly, very high levels of DDD were detected in sediments which could probably suggest the use of DDD, once used as a pesticide of its own, rather than of technical DDT in the sugarcane plantations.

Whereas sediments from Dar es Salaam coast and TPC sugar plantations showed no significant differences in detection frequencies and levels of residues between the two seasons, samples from Lake Victoria showed a notable difference. Pesticide residues in dry season samples from Lake Victoria and its basin were generally below average detection limits, but there was a massive enrichment of pesticide-containing sediments run off during the wet season (Tables 2 and 3).

The very high levels of total DDT found at Vikuge, (with levels of sum *p,p′*-DDT and *o,p′*-DDT accounting for more than 60% of total DDT in all sediment samples), highlights the persistence of DDT residues in the environment. Besides the implication of high amounts of technical DDT in the stock, presence of high levels of *p,p′*-DDD in sediments further suggests the presence of *p,p′*-DDD in the stock. High proportions of α- and β-HCH in samples from Vikuge again suggest the presence of technical HCH in the donated consignment.

3.3. Levels of pesticide residues in soils

The residues found in soil samples were more or less the same as those found in water and sediment samples although the levels were highly variable. Samples from Lake Victoria and its basin revealed generally low levels of the residues with low frequencies of detection during the dry season (Table 4). Higher levels of DDT and HCH were detected in soil samples taken in wet season, with domination of *p,p′*-DDT and α- and β-HCH. Substantially higher levels of DDT and HCH were found in samples taken in both dry and wet seasons from TPC sugarcane plantations (Tables 4 and 5). Some samples from TPC indicated high levels of α-HCH compared to the other isomers implying recent use of technical HCH product.

Results from surface soil at Vikuge revealed very high residue levels (Table 5). The total DDT in surface samples, in which *p,p′*-DDT accounted for more than 60% of total DDT like in sediments, suggests that a large amount of technical was DDT stored in the area. α-HCH was detected in high amounts, which is again indicative of contamination by high amount of technical HCH product. The total pesticide content in the soil was almost up to 40%, a fact that classifies the soil to be heavily contaminated and to be in need of decontamination measures. A GEF-UNEP project proposal ‘Bioremediation of POPs Impacted Soils in Eastern Africa’, ear-marking Vikuge as a demonstration site has been submitted.

Whereas DDT and HCH were frequently found in all environmental samples dieldrin was only found in the coastal areas of Dar es Salaam and, to lesser extent in samples from TPC. The presence of dieldrin in Dar es Salaam area can be attributed to past use of aldrin in protecting buildings from termites. With time the latter is oxidized to dieldrin, which is more easily washed away. Occurrence of this pesticide in the sugar plantation can also be related to the past use in controlling termites in the sugar plantations.

3.4. Pesticides residues in biota

Pesticide residues in biota were analysed in samples from Dar es Salaam coast and Lake Victoria and its basin. The detected residues were DDT and its metabolites, dieldrin and endosulfan (Table 6). DDT residues were detected in biota from both sampling locations. Whereas dieldrin was detected only in samples from Dar es Salaam coast, endosulfan was detected in samples from Lake Victoria only. The detected pesticide residues reflected the type of pesticides that have been used in the respective areas. DDT and dieldrin have been used for public health and even in agriculture, while dieldrin and aldrin have been used against termites in godown construction. In this case, washings from applied area is likely to be drained to rivers and then taken up by biota.

In the Dar es Salaam samples about 80% of the total DDT detected were contributed by DDE, its most stable metabolite that usually indicates past use of DDT. In some of the Lake Victoria biota, relatively high levels of fresh DDT were detected in comparison with its metabolites, contrary to what were detected in Dar es Salaam coast. Likewise, the levels of α- and β-endosulfan residues were higher in some samples than the metabolite endosulfan sulphate. DDT was detected in all biota samples from Dar es Salaam coast with average concentrations of 22.4 $\mu g\,kg^{-1}$ fresh weight, ranging from 5.5–76 $\mu g\,kg^{-1}$ fresh weight. Maximum DDT in Dar es Salaam biota was found in mangrove crabs (*Scyla serrata*) from Salender Bridge. The average concentration of total DDT detected in biota samples from Lake Victoria was 3.4 $\mu g\,kg^{-1}$ fresh weight and the concentrations ranged from bdl to 24 $\mu g\,kg^{-1}$ fresh weight. The residues were detected in only 57% of the samples and the maximum concentration was found in *Tilapia* spp. collected from Kome island in Sengerema district. Endosulfan residues of up to 80 $\mu g\,kg^{-1}$ fresh weight with average of 9.6 $\mu g\,kg^{-1}$ fresh weight was also detected, in 57% of the samples from Lake Victoria. The dieldrin residues from Dar es Salaam biota were from the crabs and catfish (*Glossgobius biocellallus*). Interestingly, biota found to have dieldrin are burrowers, signifying the possibility of dieldrin ingestion from sediments.

3.5. Suitability of biota for human consumption

The mean values and ranges of total DDT found in biota from both Dar es Salaam coast (5.5–76 $\mu g\,kg^{-1}$ fresh

weight) and Lake Victoria (bdl–24 $\mu g\,kg^{-1}$ fresh weight) were significantly below the FAO/WHO maximum acceptable limits in fish and sea food (200 $\mu g\,kg^{-1}$ fresh weight) [16] and Canadian maximum allowable limit in fish (500 $\mu g\,kg^{-1}$ fresh weight) [17]. The acceptable daily intake (ADI) of total endosulfan is given as 0.006 mg kg^{-1} b.w [18]. This means that; for an average adult of 60 kg body weight, a daily intake of 0.36 mg of pesticide per day is tolerable. The maximum levels detected in the study (0.2 mg kg^{-1} fresh mass) are still well below the MRL in fish.

4. Conclusion

The pesticide residue levels attributable to agricultural and public health use in the country are generally low. This is not the case with old storage sites like Vikuge, which is among the several contaminated sites in the country. This means that Tanzania has more to worry about obsolete pesticides than pesticides used in agriculture and public health. Another point of concern is the high use of DDT and technical grade HCH as pesticides in agriculture. These are both banned in the country, and farmers have to be sensitised and the importation controls strengthened to prevent them from reaching the poor farmers, whose low purchasing capacity is a key factor in their use of these cheap but banned products.

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