Atmospheric Concentrations of Organochlorine Pesticides in the Northern Lake Victoria Watershed

WEJULI M.S. [1], MUIR D. [2], HECKY R. E. [3], MAGUNDA M. K. [1], AND FELLIN P. [4]

[1]*Kawanda Agricultural Research Institute*
P.O. Box 7065
Kampala, Uganda

[2]*Environment Canada NWRI,*
867 Lakeshore Road Burlington ON Canada
L7R 4A6

[3]*University of Waterloo*
200 Univ. Aven. West, Waterloo ON Canada
N2L 3Gl

[4] *AirZone Inc.,*
2240 Speakman Drive
Mississauga, ON Canada
L5K 1A9

ABSTRACT

Organochlorine pesticides may still be in use in the Eastern African region for agricultural purposes and for the control of mosquitoes. Atmospheric concentrations of organochlorine pesticides are expected to be higher in the tropics compared to temperate regions due to prevailing high temperatures. However, no study has been conducted to investigate atmospheric transport and deposition of pesticides in East Africa. In this study atmospheric concentration of organochloride pesticides in the air were determined at Kakira near Jinja from November 1999 to March 2000. Total DDT was in the range 64.8-610.9 pg/m^3, dieldrin 23.6-90.8 (average 45.7) pg/m^3, α-endosulfan 32.5-206.1 (average 92.4) pg/m^3, lindane (γ-HCH) 20.3-183.6 (average 70.5) pg/m^3, chlordane and heptachlor and hexachlorobenzene (HCB) ranged from <0.1- 10.1 pg/m^3. The insecticide p,p'-DDT metabolite, p,p'-DDE was the predominant DDT isomer ranging from 22.6-390.1 pg/m^3. However, o,p'-DDT isomer was frequently detected (11.2-62.8 pg/m^3) and its metabolite p,p' DDE was relatively high ranging from 29.9-109.9 pg/m^3. The most predominant organochloride pesticides in decreasing average concentration levels DDT (total), α-endosulfan and lindane, p,p-DDT, p,p'-DDT, p,p'-DDE, dieldrin and o,p'-DDT. Average levels of p,p'-DDT and p,p'-DDE at Kakira were lower than those reported for Brazzaville but higher than those reported for Lake Baikal and Southern Ontario while α-HCH, γ-HCH, total chlordane, and hexachlorobenzene (HCB) concentrations for Kakira were lower than those reported for Brazzaville, Lake Baikal in Russia, Southern Ontario. In comparison with data from Senga Bay, Lake Malawi in Malawi, average DDT (total), p,p'-DDE, p,p'-DDT, o,p'-DDT, lindane (γ-HCH) and ΣHCH levels were higher and average dieldrin, α-HCH, chlordane and HCB levels were lower. The presence of these chemicals in air at Kakira was most likely due to both local and regional inputs.

Keywords: Concentration levels, organochloride pesticides, air, Lake Victoria

INTRODUCTION

Lake Victoria is the second largest lake in the world. However, the quality of its waters and fisheries has been affected by the land-based activities such as agriculture. The increasing population pressure and socio-economic activities within the basin have led to increased agricultural activities. Increasing population pressure has also led to increased land pressure, which in turn, has prompted the need to increase agricultural production per unit area rather than increasing production by increasing acreage. This has compelled farmers to use agrochemicals in order to increase and sustain productivity. Increasing agrochemical use and the purchase of chemicals from uncontrolled sources has led to indiscriminate use. A survey conducted in the Uganda Lake Victoria catchment revealed continued use of organochlorine pesticides lindane (γ-HCH) and α-endosulfan (Wejuli and Magunda, 1998). The updated Agrochemicals Database (1999) of the Land Use Management Component of Lake Victoria Environmental Management Project (LVEMP) showed that over 1659.7 kg α-endosulfan and 398.7 kg lindane (γ-HCH) were used in the Uganda catchment area of the lake during 1999. Some chlorinated chemicals have also been reported to be present as micro-contaminants in other pesticides, for example, DDT in dicofol and hexachlorobenzene (HCB) in lindane (Bailey, 1998). DDT is also still used as a vector control in Southern and Eastern Africa. Chlorinated chemicals are very persistent in soil, air and water. Post-application volatilisation and subsequent atmospheric transport is a primary means by which these chemicals may be deposited in areas far from their source of application. Volatilisation is the source of pesticides found in air, rain, or fog that indicate local, regional and global transport (Eisenreich *et al.,* 1981; Glotfelty *et al,* 1987; Karlsson *et al.,* 2000). Consequently, the Lake Victoria system may be receiving chlorinated compounds from outside its watershed in addition to

o,p'-DDT. Average levels of p,p'-DDT and p,p'-DDE at Kakira were lower than those reported for Brazzaville but higher than those reported for Lake Baikal and Southern Ontario while α-HCH, γ-HCH, total chlordane, and hexachlorobenzene (HCB) concentrations for Kakira were lower than those reported for Brazzaville, Lake Baikal in Russia, Southern Ontario. In comparison with data from Senga Bay, Lake Malawi in Malawi, average DDT (total), p,p'-DDE, p,p'-DDT, o,p'-DDT, lindane (γ-HCH) and ΣHCH levels were higher and average dieldrin, α-HCH, chlordane and HCB levels were lower. The presence of these chemicals in air at Kakira was most likely due to both local and regional inputs.

Keywords: Concentration levels, organochloride pesticides, air, Lake Victoria

INTRODUCTION

Lake Victoria is the second largest lake in the world. However, the quality of its waters and fisheries has been affected by the land-based activities such as agriculture. The increasing population pressure and socio-economic activities within the basin have led to increased agricultural activities. Increasing population pressure has also led to increased land pressure, which in turn, has prompted the need to increase agricultural production per unit area rather than increasing production by increasing acreage. This has compelled farmers to use agrochemicals in order to increase and sustain productivity. Increasing agrochemical use and the purchase of chemicals from uncontrolled sources has led to indiscriminate use. A survey conducted in the Uganda Lake Victoria catchment revealed continued use of organochlorine pesticides lindane (γ-HCH) and α-endosulfan (Wejuli and Magunda, 1998). The updated Agrochemicals Database (1999) of the Land Use Management Component of Lake Victoria Environmental Management Project (LVEMP) showed that over 1659.7 kg α-endosulfan and 398.7 kg lindane (γ-HCH) were used in the Uganda catchment area of the lake during 1999. Some chlorinated chemicals have also been reported to be present as micro-contaminants in other pesticides, for example, DDT in dicofol and hexachlorobenzene (HCB) in lindane (Bailey, 1998). DDT is also still used as a vector control in Southern and Eastern Africa. Chlorinated chemicals are very persistent in soil, air and water. Post-application volatilisation and subsequent atmospheric transport is a primary means by which these chemicals may be deposited in areas far from their source of application. Volatilisation is the source of pesticides found in air, rain, or fog that indicate local, regional and global transport (Eisenreich *et al.,* 1981; Glotfelty *et al,* 1987; Karlsson *et al.,* 2000). Consequently, the Lake Victoria system may be receiving chlorinated compounds from outside its watershed in addition to direct inputs from usage in Kenya, Tanzania and Uganda. The objective of this study was to analyse organochlorine pesticides in air as a basis for estimating depositions to Lake Victoria from the atmosphere and determine possible sources of these pollutants.

MATERIALS AND METHODS

This study was conducted at Kakira sugar plantation, located at 0°30'N-33°17'E, in Mayuge District. Soils at the estate are mainly Rhodi-ferralic nitisol.

Air samples were collected with a TE-PUF Poly-Urethane Foam High Volume Air Sampler (Tisch Environmental Inc., Ohio, USA) using the US EPA Method T04, beginning in November 1999. The sampler was set up on the northern catchment of Lake Victoria at Kakira sugar plantation. Approximately 250 m^3 of air was drawn periodically through the sampling unit over a 24 h cycle. Minimum and maximum temperatures, and wind direction were recorded during sampling. The samples were sent to the National Water Research Institute, Canada, for analysis. Detailed analytical procedures as in (Strachan and Hunehault, 1984; Hart *et al.,* 1992; Environment Canada, 1999; Karlsson *et al.,* 2000). The PUF plugs were extracted using a soxhlet apparatus with hexane or hexane/dichloromethane in a 1:1 ratio. Extracts were evaporated, the solvent changed to isooctane, and then further evaporated with nitrogen. Samples were fractionated by column chromatography with either neutral silica (activated) or florisil (1.2% deactivated) eluted sequentially with mixtures of hexane and dichloromethane (Fellin *et al.,* 1996; Environment Canada, 1999). One fraction contained chlorobenzenes and p,p'-DDE and the other majority of chlorinated organic pesticides (Fellin *et al.,* 1996; Environment Canada, 1999). Samples were evaporated to 1.0 ml with nitrogen.

Analysis was performed by gas chromatography (GC) with ^{63}Ni electron capture detector (ECD) as described by Environment Canada (1999). Enantioselective analysis of α-HCH was performed with a GC with a mass selective detector (MSD). Mass spectrometric and GC inlet conditions are described in Muir *et al.* (1999). The instrumental detection limit for the organochlorine compounds was 0.1 pg/m^3 (Hoff *et al.,* 1992).

The instrumental detection limit was taken as 0.1 pg/m^3 (Hoff *et al.,* 1992). The average and standard deviation (Std Dev) values were based on the number of values above instrumental detection limit (NAD).

RESULTS AND DISCUSSIONS

Temperatures, Wind Directions and Pesticide levels

The maximum, minimum and average air temperatures, and wind directions on the days of sampling are given in Figure 1. The highest temperature was 32.5 °C recorded on 13 Feb 2000 and the lowest was 15.5 °C recorded on 23 November 1999. The highest average temperature was 24.8 °C recorded on 8 March 2000 and 11 January 2000, respectively. The lowest average temperature was 21.7 °C recorded on the same days. There were no linear correlations between average temperature and pesticide levels in air. This could be a result of pesticides not originating from local sources but rather regional locations. The organochlorine pesticides, their isomers and metabolites analysed in air are given in Table 1. The average concentrations were highest for DDT (total), followed by α-endosulfan, lindane, and dieldrin, and the lowest were chlordane, heptachlor and hexachlobenzene.

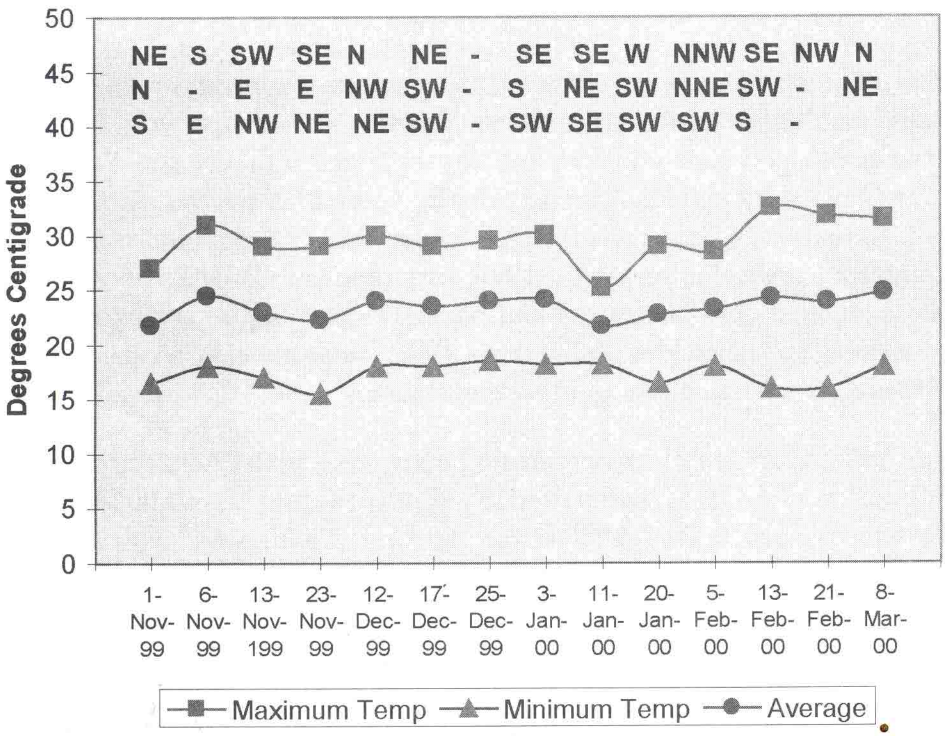

Figure 1: Maximum, minimum and average air temperatures, and wind directions on the days of sampling at Kakira, Northern L. Victoria Watershed

Table 1: **Concentrations in pg/m³ of oganochlorine pesticides in air at Kakira, Northern L. Victoria watershed from 1 Nov. 1999 to 8 Mar. 2000.**

Sampling date	p,p'-DDE	p,p'-DDD	p,p'-DDT	o,p'-DDE	o,p'-DDD	O,p'-DDT	DDT (total)	Dieldrin	α-Endosulfan	HCH (total)	α-HCH	γ-HCH	(cis)α-Chlordane	(trans)γ-Chlordane	Chlordane (total)	Heptachor	HCB
1-Nov	48.8	1.4	37.5	23.5	BD	12.9	124.0	41.8	206.1	33.5	13.2	20.3	BD	1.5	1.5	0.5	7.9
6-Nov	46.0	100.8	390.1	11.3	BD	62.8	610.9	49.8	100.5	29.7	11.7	18.0	BD	BD	BD	0.3	7.3
13-Nov	47.7	12.1	52.1	4.8	BD	27.1	143.7	46.0	94.9	37.9	14.2	23.8	BD	1.8	1.8	0.3	8.4
23-Nov	109.9	12.1	80.3	21.5	6.7	32.9	263.5	60.3	173.2	59.5	25.1	33.7	BD	3.1	3.1	0.5	10.1
12-Dec	63.1	2.0	29.1	4.4	BD	15.0	113.6	38.7	87.7	24.4	3.5	20.1	3.2	1.4	4.6	0.4	3.7
17-Dec	72.0	1.3	35.6	3.7	0.8	21.8	135.1	48.9	109.8	179.5	6.6	172.3	2.1	2.1	4.2	0.4	5.9
25-Dec	53.2	0.9	33.0	2.8	BD	20.4	110.3	34.6	60.0	40.7	3.8	35.6	1.2	1.0	2.2	0.4	4.4
3-Jan	41.0	1.0	22.6	3.5	3.5	12.7	84.3	30.2	81.2	44.0	3.5	40.2	1.5	1.6	3.1	0.3	2.9
11-Jan	77.6	1.7	37.4	3.8	2.8	23.6	146.9	57.7	76.7	128.1	1.4	126.3	1.7	1.8	3.5	0.6	3.4
20-Jan	52.9	0.7	22.0	3.4	0.7	11.2	90.8	90.8	72.3	135.4	3.0	129.4	1.2	0.8	2.0	0.4	7.3
5-Feb	55.9	1.2	26.2	3.7	BD	13.6	100.6	27.5	39.6	81.1	2.8	77.8	1.3	1.1	2.4	0.2	5.9
13-Feb	42.9	1.1	22.7	3.4	BD	12.5	82.7	23.6	32.5	80.4	1.6	77.9	BD	BD	BD	BD	BD
21-Feb	29.9	1.1	19.2	3.2	BD	11.4	64.8	61.3	113.6	185.1	1.0	183.6	0.9	1.1	2.0	0.2	3.0
8-Mar	51.9	1.1	31.7	3.2	BD	17.4	105.2	28.2	45.6	33.4	2.9	27.6	0.8	0.6	1.4	0.2	2.9
NAD	14	14	14	14	5	14	14	14	14	14	14	14	9	12	12	13	13
Average	56.6	9.9	60.0	6.9	2.9	21.1	155.5	45.7	92.4	78.0	6.7	70.5	1.5	1.5	2.7	0.4	5.6
Std Dev	20.9	26.5	96.3	7.0	2.5	13.7	139.5	18.0	48.6	56.3	6.9	58.9	0.7	0.7	1.0	0.1	2.4

BD = value below instrumental detectable limit

DDT

DDT was a frequently detected pesticide with levels ranging from 64.8-610.9 pg/m^3 (total) (Fig. 2). The insecticidally isomers p,p'-DDT and o,p'-DDT ranged from 19.2-390.1 and 11.2-62.8 pg/m^3 , respectively. The isomer p,p'-DDT is the main component of DDT. Technical products contain $\leq$ 30% o,p'-DDT (Worthing and Walker, 1987). DDT's maximum occurred on 6 November 1999 with minor maximum on 23 November 1999. (Fig. 3). Maximas were probably due to new applications either locally or within the region before or on the date of sampling. The persistent p,p'-DDT metabolite, p,p'-DDE that are the predominate breakdown DDT products ranged from 41.0 to 109.9 pg/m^3 , with 12 out of 14 (85.7%) values greater than the parent compound. In the environment, extensively processed technical DDT is reported to have ratios of p,p'-DDE/p,p'-DDT in the order of 3.1 (Schmitt *et al.*, 1990; Strandberg *et al.,* 1998). The ratios of p,p'-DDE/p,p'-DDT obtained in this study ranged from 0.1-2.4 further suggesting continued use of DDT in East Africa region. The major components contributing to total DDT were p,p'-DDT, p,p'-DDE, o,p'-DDT and o,p'-DDE (Figures 4 and 5), however, p,p'-DDD was relatively high (100.8 pg/m^3) on 6 November 1999 and another peak observed on 23 November 1999. This suggests that the DDT applied around 6 November probably contained relatively high quantities of p,p'-DDD compared to o,p'-DDT, which could have been one of the major breakdown products during storage. Metabolite p,p'-DDD is less persistent than p,p'-DDE (Fig. 6).

The overall average p,p'-DDT values were 6 times and 3 times higher than those reported for Lake Baikal region (Russia), and Southern Ontario (Canada), were 20 times less than those reported for Brazzaville (Congo). Average p,p'-DDE values were comparable to those reported for Southern Ontario, but were about 10 times lower than those reported for Lake Baikal region and 6 times higher than for Brazzaville (Karlsson *et al.,* 2000). The data obtained were also compared with data from Lake Malawi area (Karlsson et al., 2000) and is presented in Fig. 5. The average values of p,p'-DDE, p,p'-DDT, o,p'-DDTand DDT (total) were 2.8, 5, 3.5 and 6 times higher than those observed in Lake Malawi area, and suggests probably that more DDT is still in use in the Lake Victoria region than is in the Lake Malawi region.

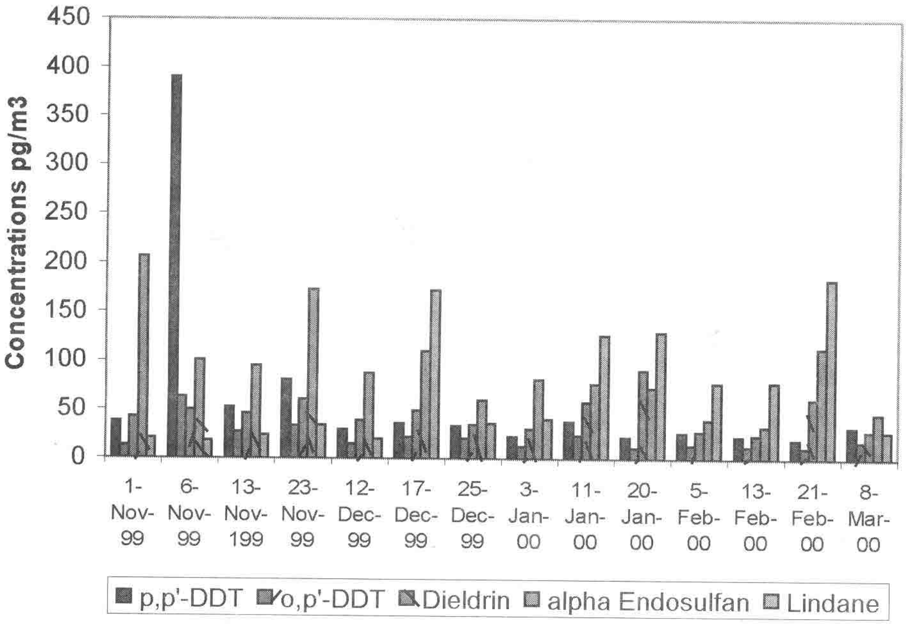

Figure 2: Concentrations of major organochlorine pesticides in air at Kakira, Northern L. Victoria Watershed

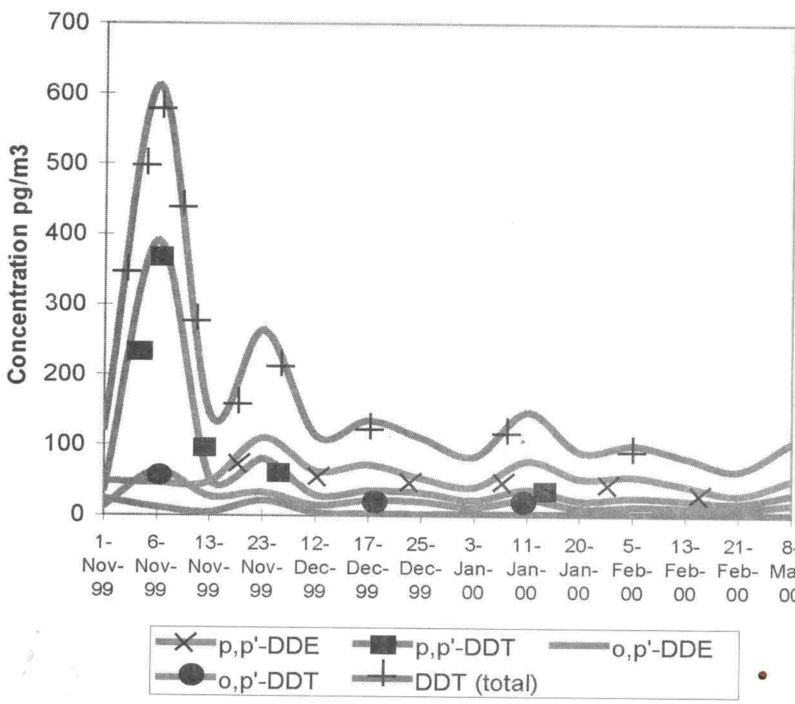

Figure 3: Concentrations of DDT isomers and major metabolite in air at Kakira, Northern L. Victoria Watershed

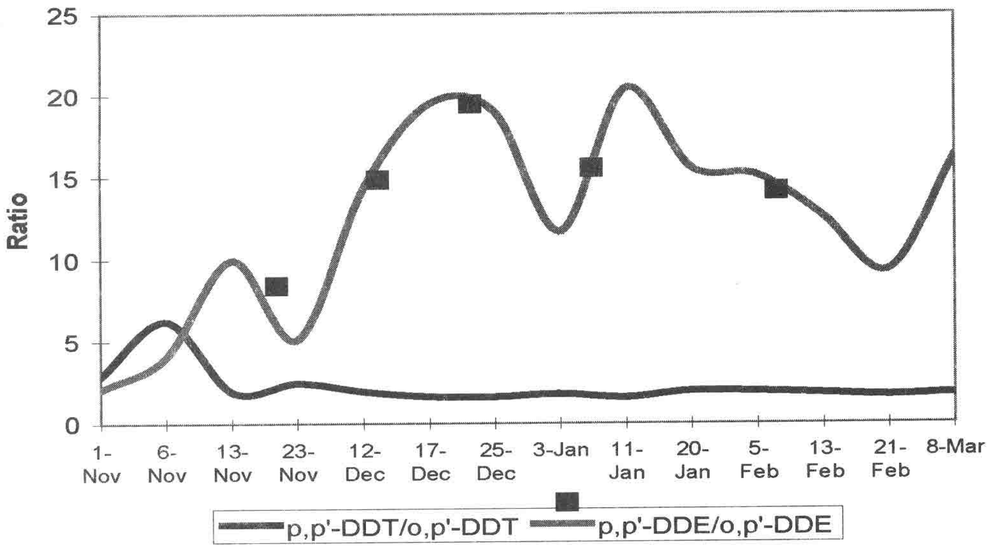

Figure 4: Ratios of selected DDT isomers and metabolites in air at Kakira, Northern L. Victoria Watershed

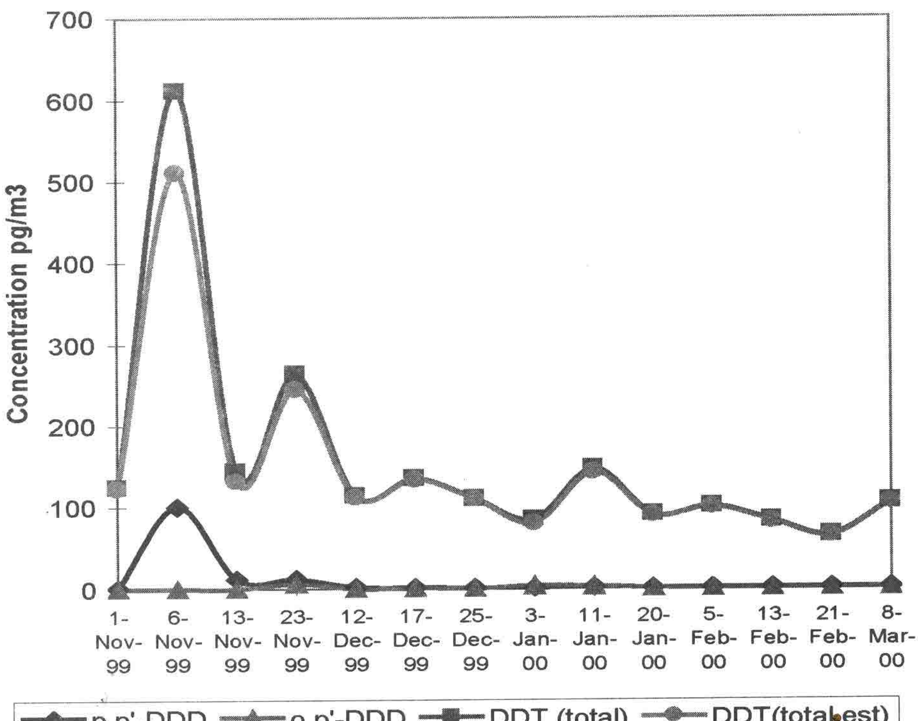

Figure 5: Contribution of DDT metabolites DDD to total DDT concentrations in air at Kakira. DDT (total est) refers to total DDT without the two metabolites

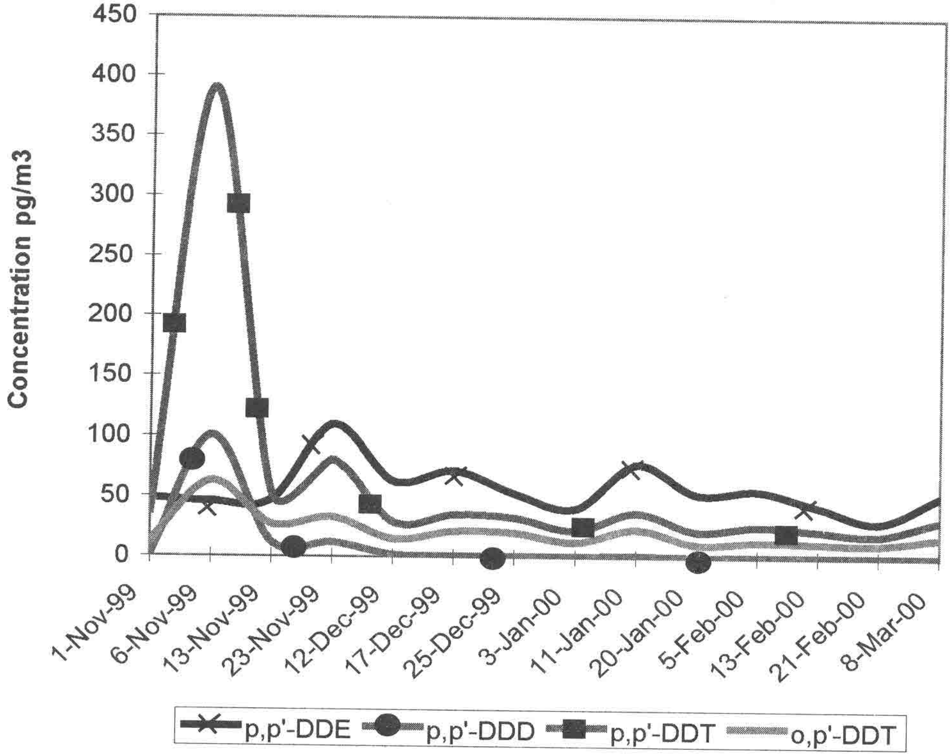

Figure 6: Comparison of trends of metabolites p,p'-DDE and p,p'-DDD in relation to mother compound p,p'-DDT and isomer o,p'-DDT

Hexachlorocyclohexane (HCH)

HCH detected had values of HCH (total) ranging from 29.7-179.5 pg/m^3 (Table I). The isomers α-HCH and γ-HCH ranged from 1.0-25.1 and 18.0-172.3 pg/m^3 respectively (Table I). Technical HCH is reported to have a ratio of α-HCH/γ-HCH of 4-7, while lindane contains at least 99% γ-HCH (Iwata *et al.*, 1993). The ratio of α-HCH/γ-HCH for the period sampled was 0.01-0.75 (depicted in Fig. 5) suggesting continued massive use of lindane. This is also supported by data from the Agrochemicals Database (1999), which reports that over 400 kg lindane were used during 1999. Lindane (γ-HCH) levels were about 6 times lower than observed in Brazzaville, 2 times lower than for Southern Ontario region, comparable to Lake Baikal and, about 10 times higher than what has been reported for the Arctic and Antarctic (Karlsson *et al.*, 2000) and 3 times higher than reported for Lake Malawi region (Fig. 7).

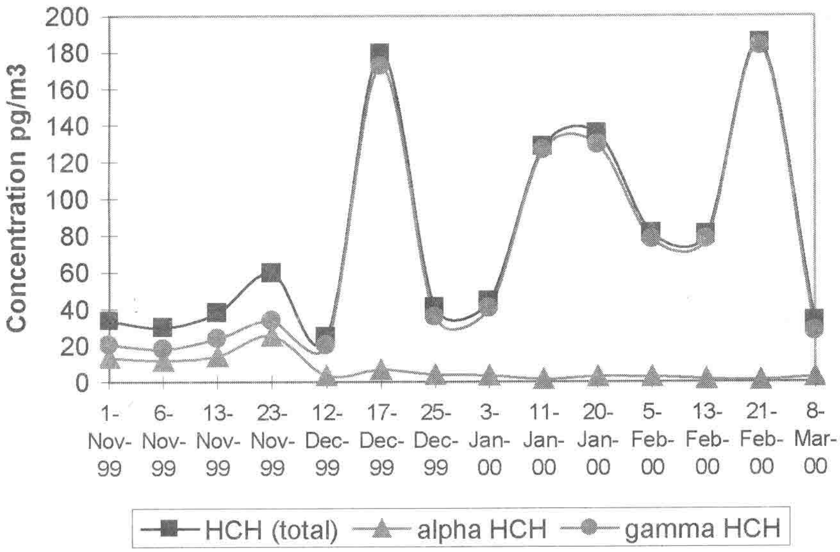

Figure 7: Concentrations of hexachlorocyclohexane (HCH) isomers in air at Kakira, Northern L. Victoria Watershed

Dieldrin and α-endolsulfan

Dieldrin and α-endosulfan were frequently detected with values of 23.6-90.8 pg/m^3 and 32.5-206.1 pg/m^3 respectively. Use of over 1700 kg α-endosulfan in the region in 1999 has been reported (Agrochemicals Database, 1999). The dieldrin values are relatively higher compared to the p,p'-DDT values. This is an indication that probably dieldrin is still in use. Dieldrin values were 1.7 times lower than those for the Lake Malawi region while α-endosulfan was 3.9 times higher (Fig. 8).

Chlordane, Heptachlor, Hexachlorobenzene (HCB)

Chlordane, heptachlor and HCB concentrations were low ranging from <0.1-10.1 pg/m^3, respectively. Total chlordane concentrations were <0.1-4.6 pg/m^3, (cis) α-chlordane <0.1-3.2 pg/m^3, and (trans) γ-chlordane <0.1-3.1 pg/m^3 (Table 1). Heptachlor and HCB values were <0.1-0.6 and <0.1-10.1 pg/m^3 respectively. These low concentrations suggest that the three chlorinated pesticides are not largely used or not in use in this region and presence may be due to regional rather than local use. The average total chlordane value was comparable to those reported for Lake Malawi, Lake Baikal and the Arctic but were about 5 and 10 times lower than those for Brazzaville and Southern

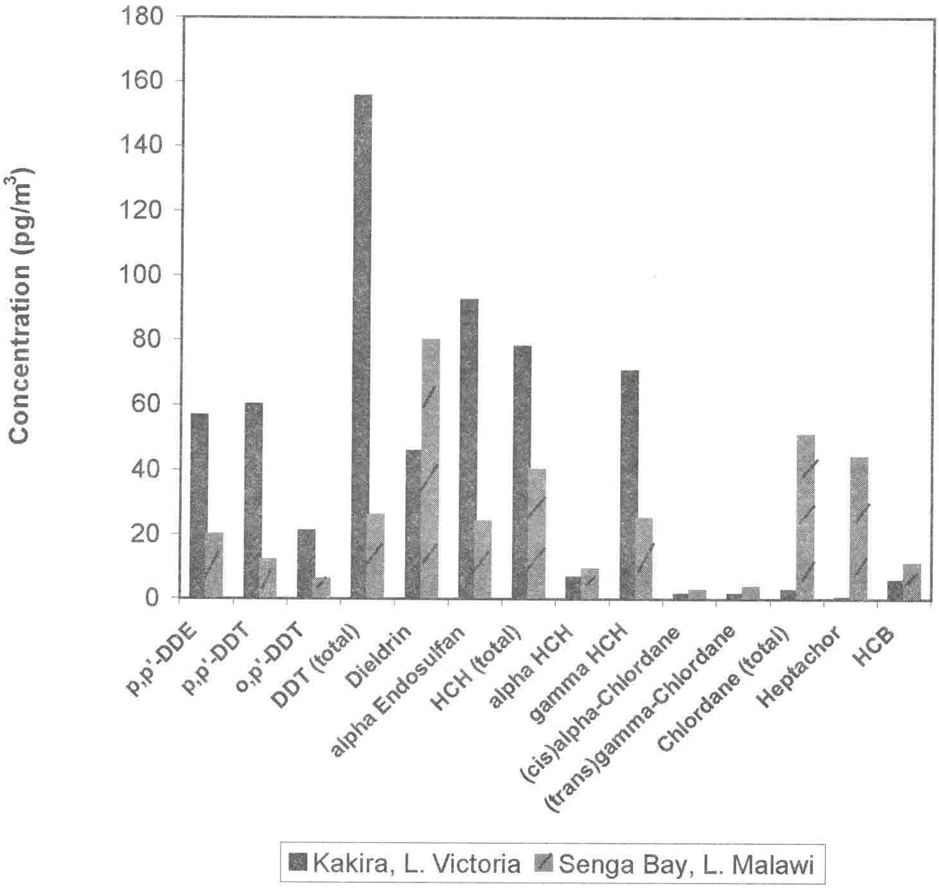

Figure 8: Comparison of average air concentration of organochlorine pesticides in L. Victoria area Nov. 1999-Mar. 2000 and L. Malawi area Feb 1997-May 1998 (Source Malawi Data: Karlsson *et al.*, 2000)

Ontario respectively. The average heptachlor value was 110 times lower than the Lake Malawi value (Fig. 8). The average HCB concentration was comparable to the value for Lake Malawi but was 20, 10 and 15 times lower than that of Lake Baikal, Southern Ontario, and the Arctic, respectively.

CONCLUSION AND RECOMMENDATIONS

The atmospheric concentrations of organochlorine pesticides indicate that they are still in use in the Lake Victoria region. Organochlorine pesticides are still used for agricultural purposes and for the control of mosquitoes, despite the restriction about their importation. Volatilisation from land and water surfaces and subsequent atmospheric transport of these pesticides may pose environmental pollution to areas far from their sources of application. There is need to address this issue and investigate the cycling processes of these substances within the Lake Victoria.

ACKNOWLEDGMENTS

Financial support from the Lake Victoria Environmental Management Project (LVEMP), technical assistance from the National Water Research Institute of Environment Canada, Management of Kakira Sugar Works Jinja for their high level of support, and Mr. J. Francis Oundo of the Agronomy Section of Kakira Sugar Works for collecting the samples.

REFERENCES

Agrochemicals Database, (1999). LVEMP Land Use management Component, Kawanda Agricultural Research Institute, Kampala-Uganda.

ATSDR., (1994). Agency for Toxic Substances and Diseases Registry (ATSDR)/ US Public Health Service, Toxicological Profile for 4,4'-DDT, 4,4'-DDE, 4, 4'-DDD (Update). ATSDR. Atlanta, GA.

Bailey, R., (1998). Global mass balance for Hexachlorobenzene. Report for Chemical Manufacturers Association.Great Lakes ecosystem. Environ. Sci. Technol.15:30-38.

Environment Canada, (1999). Great Lakes Quality Agreement Annex 15, Integrated Atmospheric Deposition Network Sampling Protocol Manual (SPM); Report ARD 94-003; Atmospheric Environment Service.

Fellin, P., L. A. Barrie, D. Dougherty, D. Toom, D. Muir, N. Grift, L. Lockhart and B. Billeck., (1996). Environ. Toxicol.Chem. 5:253-261.

Glotfelty, D. E., Seiber, J. N. and Liljedahl, L. A., (1987). Pesticides in fog. Nature (London). 325:602-605.

Hoff, R. M., Muir D. C. G. and Grift, N. P. (1992). Environ. Sci. Technol. 26:266-275.

Iwata, H., Tanabe S. and Tatsukawa, R. (1993). Mar. Pollut. Bull. 26:302-305.

Karlsson, H., Muir, D.C.G., Teixiera, C. F., Strachan, W. M. J., Hecky, R. E., Mwita, J., Bootsma, H.A., Grift, N.P., Kidd K.A. and Rosenberg, B., (2000). Persistent chlorinated pesticides in air, water and precipitation from the lake Malawi area, Southern Africa. Environ. Sci. Technol. 34:4490-4495.

Muir, D. C. D., Stern G. and Karlsson, H., (1999). Organohalogen Compd. 41:563-568.

Schmitt, C. J., Zajicek J. L. and Peterman, P. H., (1990). Arch. Environ. Contam. Toxicol. 19:748-781.

Strachan, W. M. J and Hunehault. (1984). Environ. Sci. Technol. 26:1048-1052.

Strandberg, B. *et al.,* (1998). 32:1754-1759.

Wejuli, M. S. and Magunda, M. K., (1998). Land Use Management Annual Technical Report. KARI-NARO, LVEMP.