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 are still in use for agricultural purposes and for the control of mosquitoes in the Eastern African region. 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 this region. In this study atmospheric concentration of organochlorine pesticides in air were determined at Kakira near Jinja from November 1999 to-date. Their ranges were as follows: DDT (total) 64.8-610.9 pg/m^3, dieldrin 23.6-90.8 pg/m^3, α-endosulfan 32.5-206.1 pg/m^3, lindane (γ-HCH) 20.3-183.6 pg/m^3 while chlordane, heptachlor and hexachlorobenzene (HCB) ranged from <0.1-10.1 pg/m^3. The insecticide p,p'-DDT was the predominating 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 predominate organochlorine pesticides in decreasing average concentration levels were DDT (total), α-endosulfan, lindane (γ-HCH), 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 Brazaville, Lake Baikal, Southern Ontaria, Arctic and Antarctic. 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 whereas average dieldrin, α-HCH, chlordane and HCB levels were lower. The presence of these chemicals in air at Kakira is most likely due to both local and regional inputs.

Keywords: Organochlorine pesticides, air pollution, 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 and 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 1659.7 kg α-endosulfan and 398.7 kg lindane (γ-HCH) were used in the Uganda catchment area of the lake during 1999. Other organochlorine pesticides have also been reported to be present as impurities 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 process by which pesticides are 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). Therefore, because of this atmospheric transport, the Lake Victoria system may be receiving chlorinated pesticides 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

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 were also recorded during sampling. The samples were sent to the National Water Research Institute, Canada for analysis. Detailed analytical procedures are available elsewhere (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 for 24 hours. Extracts were evaporated, the solvent changed to isooctane, and then further evaporated with nitrogen. A fractionation of samples was 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 chromatograph (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 elsewhere (Muir *et al*., 1999). An instrumental detection limit of 0.1 pg/m^3 for organochlorine compounds in air was used (Hoff *et al.,* 1992).

Results and discussions

The maximum, minimum and average air temperatures on the days of sampling are given in Figure 1. The highest temperature was 32.5 ^oC recorded on 13 Feb 2000 and the lowest was 15.5 ^oC recorded on 23 November 1999. The highest and lowest average temperatures were 24.8 ^oC recorded on 8 March 2000 and 21.7 ^oC recorded on 8 March and 11 January 2000, respectively. The correlations between average temperatures and pesticide levels in air were very weak. This could be an indication that the pesticides detected did not specifically originate from local sources but probably from both local and regional locations. The organochlorine pesticides analysed in air are given in Table 1. The average concentration was highest for DDT (total), followed by α-endosulfan, lindane and dieldrin and lowest for chlordane, heptachlor and hexachlobenzene.

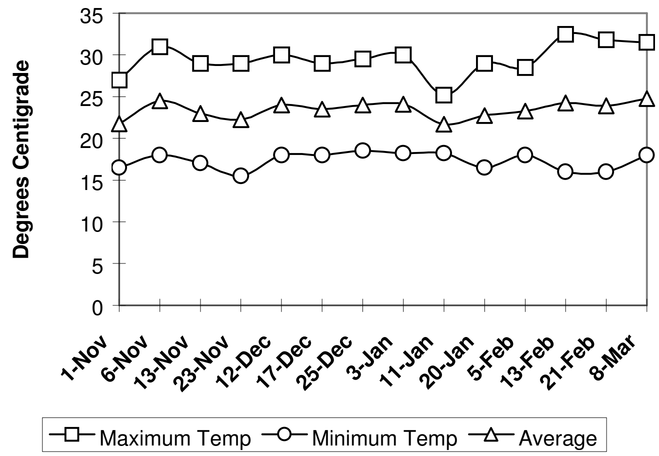

Figure 1. Maximum, minimum and average air temperatures on the days of sampling at Kakira, northern Lake Victoria watershed

DDT

DDT was the most frequently detected pesticide (Figure 2) with levels ranging from 64.8-610.9 pg/m^3 (total) and, 19.2-390.1 and 11.2-62.8 pg/m^3 for the insecticidally active isomers p,p'-DDT and o,p'-DDT respectively (Table 1). The isomer p,p'-DDT is the main component of DDT, however, technical products contain ≤ 30% o,p'-DDT (Worthing and Walker, 1987). The highest values occurred on 6 November 1999 with other high peaks observed on 23 November and 17 December 1999, and 11 January 2000 (Figure 3). These relatively very high values are probably an indication of new applications either locally or within the region before or on the date of sampling.

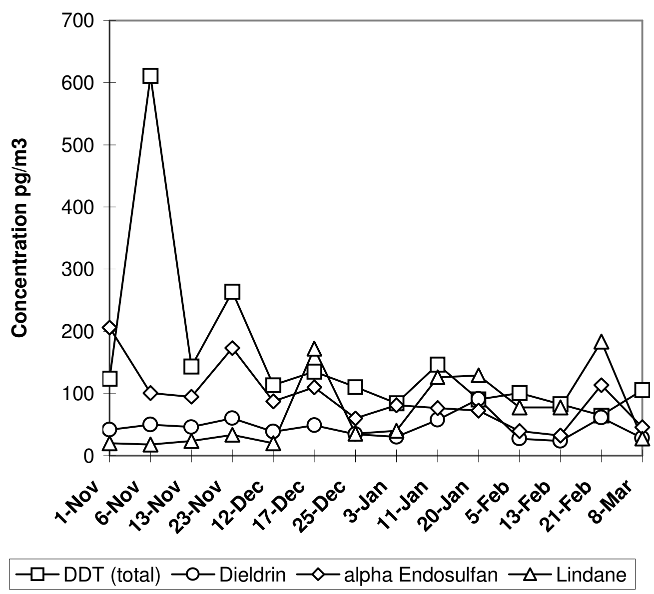

Figure 2. Concentrations of major organochlorine pesticides in air at Kakira,nNorthern Lake Victoria watershed

Table 1. Concentrations in pg/m^3 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	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

Instrumental detection limit is taken as 0.1 pg/m3 (HOFF *et al.*, 1992); NAD = number of values above instrumental detection limit; BD = value below instrumental detectable limit; average and standard deviation (Std Dev) values are based on NAD.

The persistent p,p'-DDT metabolite, p,p'-DDE was the predominant breakdown DDT product ranging from 41.0-109.9 pg/m^3 (Table 1) with 12 out of 14 (85.7%) values greater than the parent compound. In the environment, technical DDT which has been extensively broken down is reported to have ratios of p,p'-DDE/p,p'-DDT in the order of 3.1 (Schmitt, *Et Al.,* 1990; Strandberg, 1998). The ratios obtained in this case ranged from 0.1-2.4 suggesting continued use of DDT in this region. The major components contributing to total DDT were p,p'-DDT, p,p'-DDE, o,p'-DDT and o,p'-DDE (Figures 3 and 4), however, p,p'-DDD was relatively high (100.8 pg/m^3) on 6 November 1999 (Figure 4) and another peak observed on 23 November, 1999. This suggests that the DDT applied around 6 November contained p,p'-DDD which could have been a breakdown product during storage.

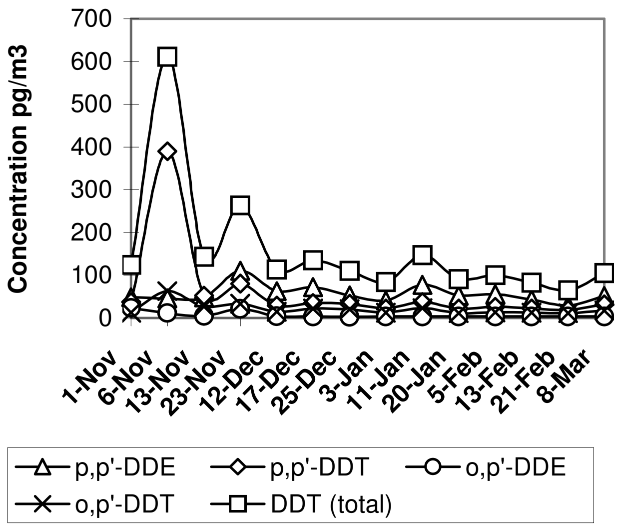

Figure 3. Concentrations of DDT isomers and major metabolite in air at Kakira, northern Lake Victoria watershed

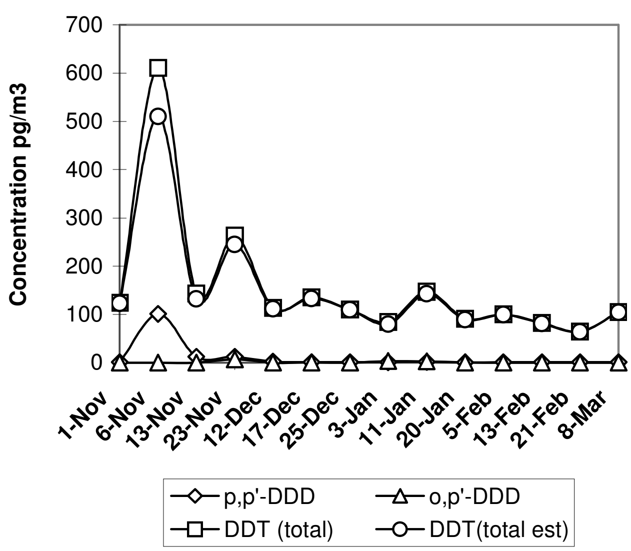

Figure 4. Contribution of DDT metabolites DDD to total DDT concentrations in air at Kakira. DDT (total est.) refers to total DDT without the two metabolites

The average p,p'-DDT values were about 10 times higher than those reported for Lake Baikal region (Russia), 3 times higher than those of Southern Ontario (Canada), and 20 times less than those reported for Brazaville (Congo). Average p,p'-DDE values were comparable to those reported for Southern Ontario, but were about 10 times those reported for Lake Baikal region and 6 times lower than Brazaville (Karlsson *et al.,* 2000). The data for the other DDT component for regions mentioned above were not available. The data obtained were compared with data from Lake Malawi area (Karlsson *et al.*, 2000) and are presented in Figure 6. 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. This suggests that probably a lot more DDT is still in use in the Lake Victoria region than is in the Lake Malawi region.

Hexachlorocyclohexane (HCH).

HCH detected had values of HCH (total) ranging from 29.7-179.5 pg/m^3 (Table 1). The isomers α-HCH and γ-HCH ranged from 1.0-25.1 and 18.0-172.3 pg/m^3 respectively (Table 1). Technical HCH is reported to have a ratio of α-HCH/γ-HCH of 4-7, while lindane contains approximately 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 Figure 5) suggesting continued massive use of lindane. This is also supported by data from the AGROCHEMICALS DATABASE, (1999), which reports use of 398.7 kg lindane during 1999. Lindane (γ-HCH) levels were about 6 times lower than observed in Brazaville, comparable to Lake Baikal and Southern Ontario regions, about 10 times higher than what has been reported for the Arctic and Antarctic (Karlsson, *et al.,* 2000) and 2.8 times higher than reported for Lake Malawi region (Figure 6).

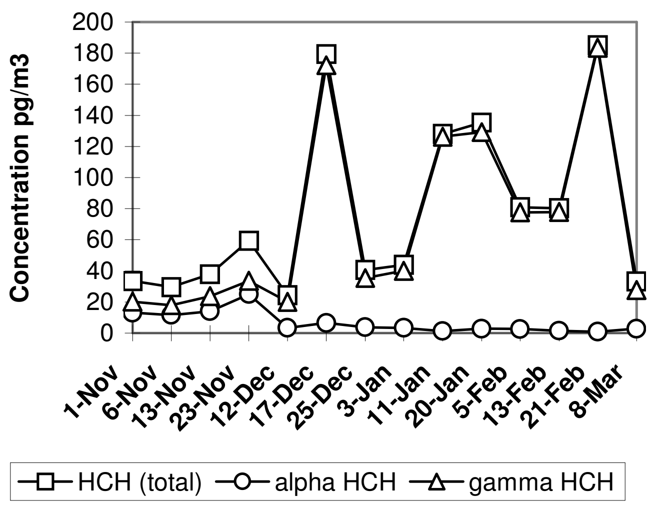

Figure 5. Concentrations of hexachlorocyclohexane (HCH) isomers in air at Kakira, Northern L. Victoria Watershed

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 respectivey. Use of 1669.7 kg α-endosulfan in the region in 1999 has been reported (AGROCHEMICALS DATABASE, 1999). The dieldrin values are very much comparable to the p,p'-DDT values and are relatively elevated. 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 (Figure 6).

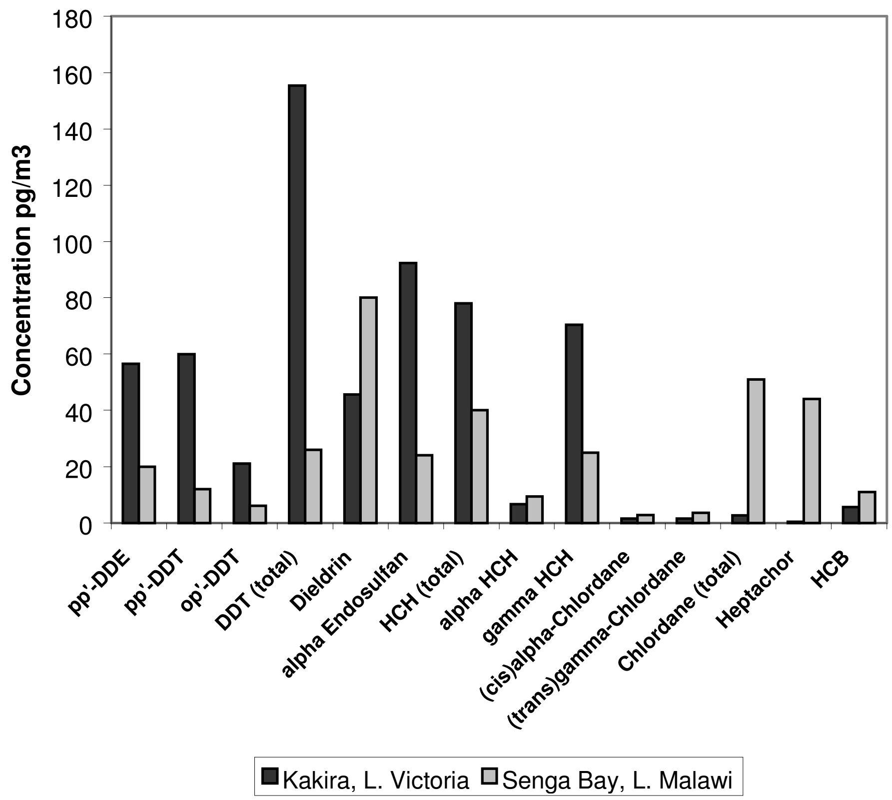

Figure 6. Comparison of average air concentration of organochlorine pesticides in L. Victoria area Nov 1999-Mar 2000 and L. Malawi area Feb 1997-May 1998 (source of Malawi Data: Karlsson *et al.,* 2000)

Chlordane, heptachlor, hexachlorobenzene (HCB)

Chlordane, heptachlor and HCB were detected in small amounts ranging from <0.1-10.1 pg/m^3. 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 probably 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 about 5 and 10 times lower than those for Brazzaville and Southern Ontario respectively. The average heptachlor value was 110 times lower than the Lake Malawi value (Figure 6). The average HCB value was comparable to the value for Lake Malawi region. However, it was about 20 times lower than that for Lake Baikal, 10 times that for Southern Ontario and about 15 times that for the Arctic.

Conclusion

Organochlorine pesticides are still in use in the Lake Victoria basin for agricultural purposes and for the control of mosquitoes. Although the importation of chlorinated pesticides was restricted in Uganda between 1993-1999, lindane and α-endosulfan are still in use and there are strong indications that dieldrin and DDT continue to be used. Volatilisation from land and water surfaces and subsequent atmospheric transport pose environmental pollution to areas far from their sources of application. Tropical regions may therefore still be global sources of chlorinated chemicals and there is need to address this issue regionally and globally. Degradation within the lake may be an important removal process of these chemicals from the environment. It is therefore important to monitor levels of these chemicals in the lake waters and investigate their cycling processes within the lake region.

Acknowledgments

Financial support from Lake Victoria Environmental Management Project (LVEMP), technical assistance from the National Water Research Institute of Environment Canada, Management of Kakira Sugar Works Jinja for accepting the equipment to be placed at their meteorological station and Mr. 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.

Bailey, R. 1998**.** Global mass balance for Hexachlorobenzene. Report for Chemical Manufacturers Association.

Eisenreich S.J., Looney B.B. and Thornton J.D. 1981**.** Air-borne organic contaminants in the Great Lakes ecosystem. *Environ. Sci. Tec*hnol.**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**.**, Barrie L.A, Dougherty D., Toom D., Muir D., Grift N., Lockhart L. and Billeck B. 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.

Hert, K.M., Isabelle L.M. and Pankow J.F.. 1992. *Environ. Sci. Technol*. **26**:1048-1052.

Hoff, R.M., Muir D.C.G. and Grift N.P**.** *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., Iwita 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. Techn*ol. **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. 1998. **32**:1754-1759.

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