REPORT

A Preliminary Evaluation of the DDT Contamination of Sediments in Lakes Natron and Bogoria (Eastern Rift Valley, Africa)

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Abstract Dichlorodiphenyltrichloroethane (DDT) is still used in Africa for the indoor control of malaria and it may represent a potential hazard for wildlife. The littoral sediments of two alkaline-saline lakes, Natron (Tanzania) and Bogoria (Kenya), in the Eastern Rift Valley, supporting large populations of lesser flamingos (*Phoeniconaias minor*), were analysed for DDT residues. Physical–chemical analyses (temperature, conductivity, pH and dissolved oxygen) were also performed on the water of the two lakes and in the tributaries of Lake Natron, to evaluate the influence of the environmental variables on pollutant occurrence. At Lake Natron, around 1 km from the sediment collection sites, tree leaves of *Acacia tortilis* were also collected. The main metabolite found in all sediment samples was pp'DDE, whilst equal concentrations of pp'DDT and pp'DDE were measured in acacia leaves. The levels of DDTs measured in the sediments were within 5.9–30.9 ng g^{-1} d.w., reaching the maximum value in a tributary of Lake Natron. On the whole, the contamination of Lake Natron and Lake Bogoria basins seems to be quite moderate. Nevertheless, the pp'DDE/pp'DDT ratio equals 1 in the *Acacia tortilis* leaves, which makes one suppose that the input of the parent compound was rather recent and could have been from aerial transport or dust from relatively close-by old pesticides storage sites.

Keywords Obsolete contaminant pollution · Soda lakes · Sediments · *Acacia* leaves · Tanzania · Kenya

INTRODUCTION

Alkaline–saline ("soda") lakes in the East African Rift Valley (Fig. 1; latitude from 4°35′ N to 14°30′ S and a N–S distance of around 2,100 km; Spigel and Coulter 1996)

have an outstanding biodiversity value, despite being hostile environments, because together they are home to approximately two million lesser flamingos (*Phoeniconaias minor*) a species considered "near-threatened" by the International Union for Conservation of Nature. Lesser flamingos feed preferentially on *Arthrospira fusiformis*, a planktonic cyanobacterium that is particularly abundant in these lakes (Mlingwa and Baker 2006). Whilst most of the soda lakes—e.g. Nakuru, Elmenteita and Bogoria in Kenya; Manyara in Tanzania—are all used extensively by *Phoeniconaias minor*, only one regular breeding site exists in East Africa, on soda islands in the middle of Lake Natron in Northern Tanzania (Brown 1973; Harper et al. 2003). Despite the high ecological value of this environment, few studies of the Lake Natron basin and its eco-hydrology exist and no data are reported in the literature on the contamination levels of Persistent Organic Pollutants (POPs).

In Africa, pesticides have been used for combating agricultural pests and controlling disease vectors for more than 50 years (Mansour 2009) and the most common organochlorine compound used for pest control has been the insecticide pp'DDT (Dichloro Diphenyl Trichloroethane). In Tanzania, this compound has been restricted, banning it in agriculture since 1991 and in Kenya the last import was in 1985 (Wandiga 2001). At present, however, use is allowed only for Indoor Residual Spraying (IRS) for malaria vector control (Mandavilli 2006). Moreover, "hot spot" pollution areas have been found in a coastal region of Tanzania where obsolete pesticides stocked in a disused farm have been accidentally released since 1990 (Elfvendahl et al. 2004; Marco and Kishimba 2005, 2007).

Although there is no reason to expect a direct negative impact of DDT contamination in the most remote lakes of

Fig. 1 Landscape in Northern
Tanzania, inside the Rift Valley
(Photo: Roberta Bettinetti 2009)

the Eastern Rift Valley, as these areas are sparsely popu-
lated and hundreds of kilometres distant from the recog-
nised pesticide sources, an indirect influence of DDT
cannot be excluded, due to long-range transport and local
occasional IRS in tourist lodges.

Much of the knowledge of the African Rift Valley saline
lakes has come from studies on their chemistry (e.g. Wood
and Talling 1988), biodiversity (e.g. Jones et al. 1994;
Harper et al. 2003), primary production (e.g. Melack 1981,
1988) or explanations for the deaths of large numbers of
lesser flamingos (e.g. Harper et al. 2003; Krienitz et al.
2003; Ballot et al. 2004; Oaks et al. 2006). Little is known
(Saoke 2005) about the degree of pollution in these envi-
ronments by pp'DDT (e.g. Koeman et al. 1972; Gitahi et al.
2002; Kishimba et al. 2004; Mavura and Wangila 2004),
however.

The main objective of this study was to evaluate the
degree of DDT contamination of the river and lake envi-
ronments by surface sediment analyses, in order to ascer-
tain whether such remote environments were still
contaminated. Physical and chemical water parameters
were measured in situ in water where the sediments were
collected, to evaluate the possible influences of pH, tem-
perature, dissolved oxygen and alkalinity on the presence/
proportion of DDT-related compounds recovered in the
sediments. Leaves of *Acacia tortilis* were also sampled in
the Lake Natron area as a bioindication of any recent
atmospheric DDT pollution.

MATERIALS AND METHODS

Study Sites

Lake Natron (Figs. 2, 3, 4a) lies in the Eastern Rift Valley
lake of northern Tanzania just south of the Tanzania–Kenya
border. Lying at an average height of 610 m a.s.l., the lake
spans 75 km in length and 22–35 km in width (Dawson
2008). Four permanent rivers feed Lake Natron from Tan-
zania—the Engare Nyiro, the Peninj, the Moinik and the
Engare Sero, plus a number of seasonal streams. These
drain into the lake from the Ngorongoro Highlands in the
south, Mount Lengai (2,942 m a.s.l.) in the southeast, and
the Nkito Hills in the west (Hughes and Hughes 1992).

The perennial Engare Nyiro River (also known as the
Ewaso Ngiro), the lake's principal affluent, rises in Kenya's
Mau Forest, flows south through the Ngare Ngiro (or
Shompole) Swamp before crossing the border and entering
the lake in the north. Twenty eight springs, most saline or
sub-saline, flow from the base of surrounding volcanoes or
the Rift Escarpment into Lake Natron and in some cases
sustain a number of isolated lagoons around the lake edge.
The maximum depth of the lake is approximately 3–4 m
and whilst the largest single input is derived from direct
precipitation, high evaporation rates (up to 20 mm day^{-1})
and low rainfall characterise the region. During periods of
drought (from June to October) when the water level is low,
a large portion of the lake's bed is exposed, covered by a

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Fig. 2 Lake Natron and its inlet river (Photo: Roberta Bettinetti 2009)

Fig. 3 East tropical African Lakes (modified from Nyamweru 1983)
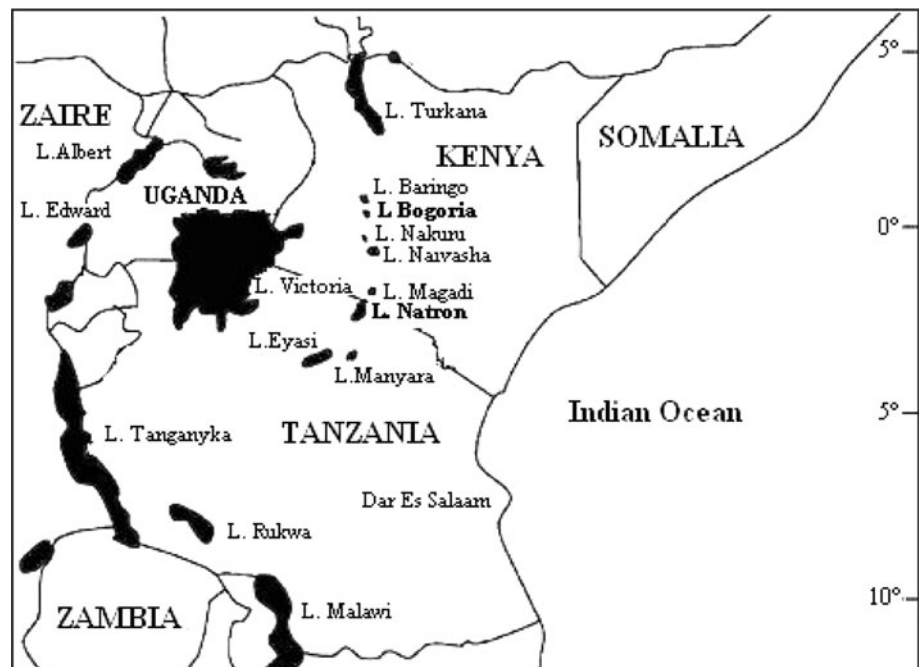
salt crust that dissolves during the rainy seasons (Wetlands International 2002). In this northern and eastern region of Tanzania, the rainfall is bimodal with two rainy seasons. The 'short rains' or Vuli last from October to December, and the 'long rains' or Masika last from March to May. As a consequence, the total surface area of Lake Natron fluctuates considerably, both seasonally and across larger

temporal scales. Figure 4a shows the extent of the exposed salt crust and the distribution of the main lagoons identified from satellite imagery taken in 2009.

In addition to flamingos, in the area around Lake Natron there is a significant population of large mammals and 113 bird species (Yanda and Madulu 2005); the streams and lagoons also support two endemic fish species (*Alcolapia*

alcalicus and A. grahammi). Permanent villages are located along freshwater inflows to the lake. In recent years, the human population has increased significantly, bringing with it an overexploitation of the local natural resources: irrigated small-scale farming has developed around the lake basin and adjacent areas, even where the soil is not particularly productive. Pastoralism by the Maasai, however, remains the dominant landuse in the area. The whole lake in Tanzania is a

wetland of international importance under the Ramsar convention but only in 2009 any staff were appointed by the Wetlands Division of the Tanzanian Ministry of Natural Resources to begin to implement sustainable wetlands policies amongst the communities around the lake.

Lake Bogoria (Figs. 3, 4b) is a deeper, saline and alkaline lake with three basins which lie in a volcanic region in a half-graben basin in the south of Kenya, a little

Fig. 4 Detailed map of Lake Natron (**a**) and Lake Bogoria (**b**) with sampling sites
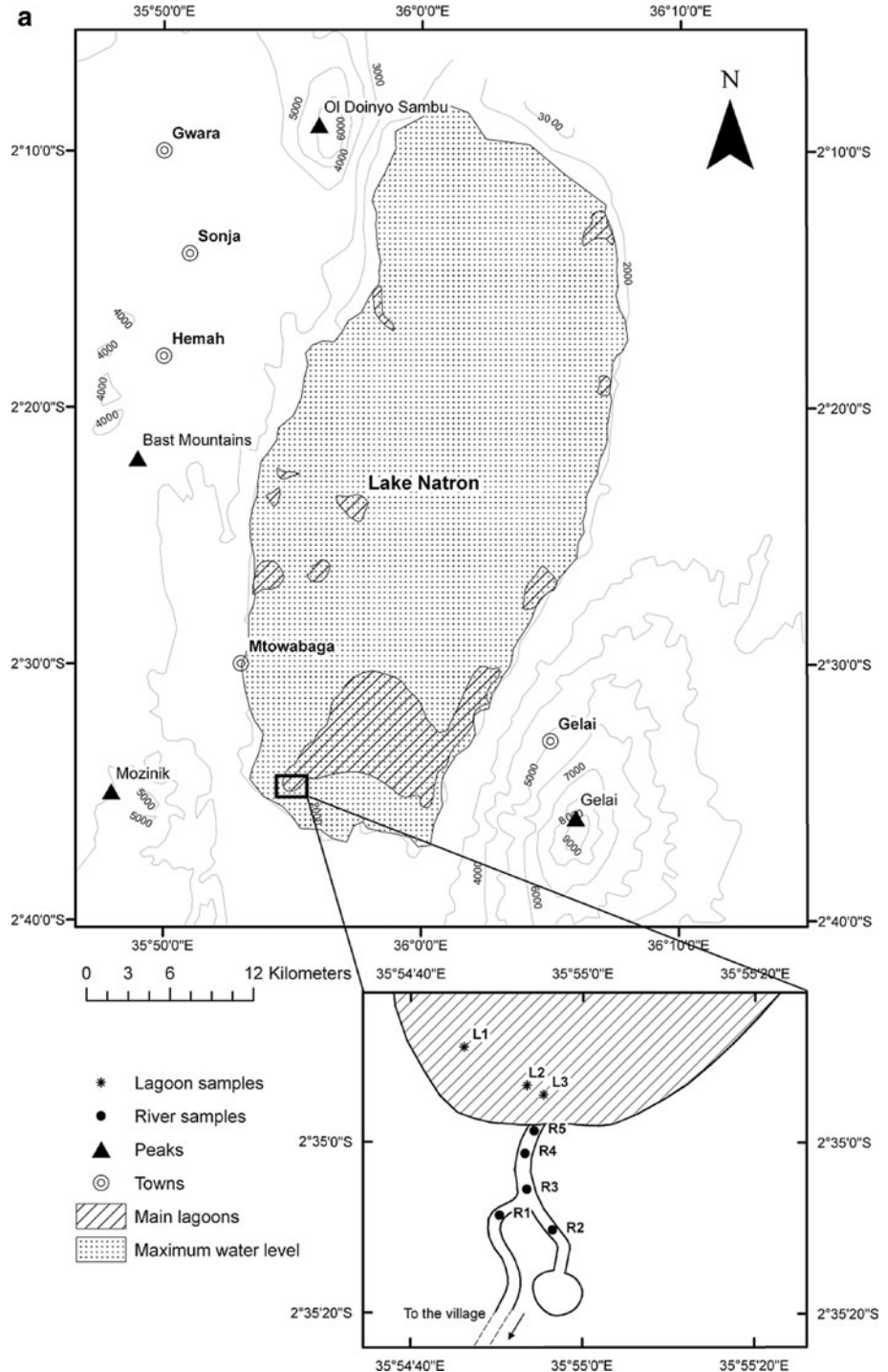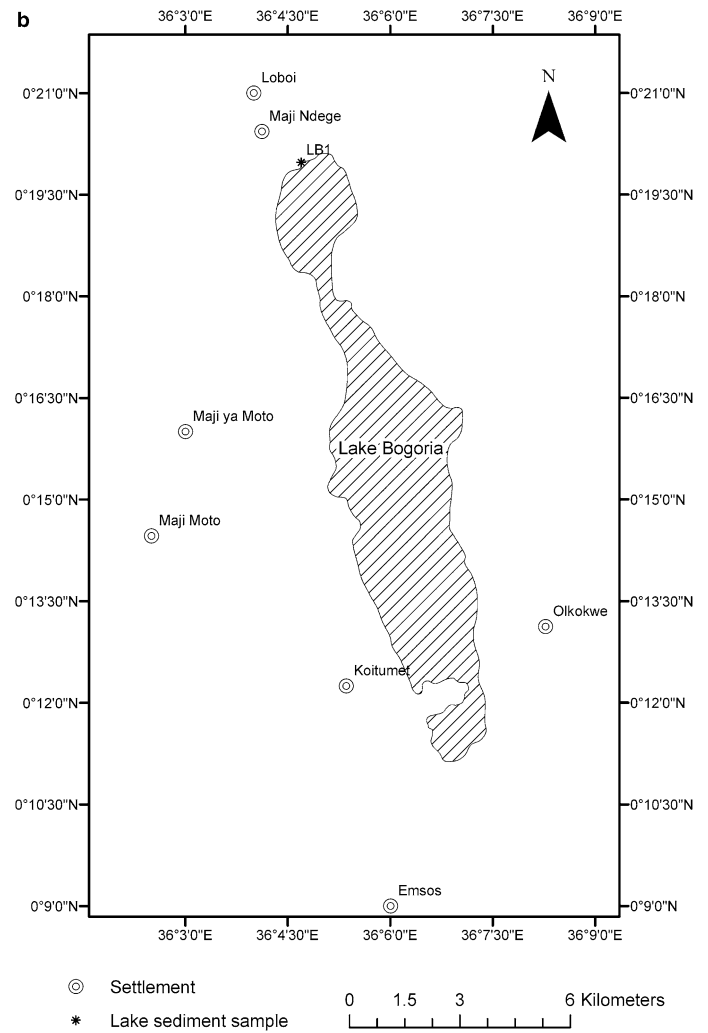
Fig. 4 continued

north of the equator. The southern basin, a relict volcanic crater, is the deepest part (14 m), joined to the rest of the lake by a narrow isthmus. With a mean surface area of 34 km^2 and a mean depth of 5.4 m, it is sustained by two semi-permanent surface water river inflows, two small permanent spring-fed freshwater streams and numerous semi-saline hot springs, most of which emerge from fissures along the western and south-eastern shores of the lake (Harper et al. 2003). The principal source of freshwater is the Sandai-Wasenges River, which drains the Subukia and Iguamiti highlands to the South-East. The river is ephemeral, drying up at the Sandai swamps during the dry season (December–February), and transports huge volumes of suspended solids during the wet season. The lake has no outlet and the intense evaporation has led to high levels of salt and mineral accumulation; however, the water level fluctuations are minimal because of its high volume/area (Wetlands International 2002).

Lake Bogoria is an important conservation area in Kenya holding regionally and nationally endangered species; it was designated as a national reserve in 1974 and in 2001 it was listed as a wetland of international importance under the Ramsar convention. Despite this fact, the land and lake of the National Reserve are at risk because of excessive grazing and erosion (land) and over-stocking and unsustainable arable agriculture in its catchment. The root causes of these problems are poverty and the absence of any effective land use advisory service.

Samples Collection and Chemical Analysis

Surface sediments (the first layer of deposition, 30 g w.w. at each sampling site) were collected by hand using a steel spatula in March 2009 (the end of the dry season), in three different sites in southern area of the Lake Natron and in five sites along the course of two inlet rivers to the lake, (Fig. 4a). One sediment sample was also collected in April 2009 in Lake Bogoria (LB1, Fig. 4b). A sample of *Acacia tortilis* leaves ($n = 6$, from one tree, approximately 2.5 m above the ground) was also collected at 1 km to the south

from the sediment sampling sites of Lake Natron at the same time of sediment sampling. This species was the only tree present in the neighbourhood of the lake. After the collection, leaves and sediments were stored at about 10°C for 5 days and then they were kept frozen until lyophilisation.

At the same sites and times, temperature, conductivity, pH and oxygen concentrations in the water column above the sediment were measured in situ with portable instruments. The organic matter content of the sediments was determined by weight Loss-On-Ignition (LOI) at 550°C (Dean 1974) in laboratory.

Extraction of freeze-dried (Edwards Pirani 1001 freeze-dryer) homogenised sediments and leaves (1 g) was performed in glass microfibre thimbles (19-mm internal diameter × 90-mm external length, Whatman, England) for 2 h with 60 ml of n-hexane (Carlo Erba, Italy, pesticide analysis grade) using a modified Soxhlet apparatus (Velp Scientifica-ECO 6 thermoreactor). For the leaf sample only, organic matter was destroyed with H_2SO_4 (98%, Carlo Erba, Italy) and chlorinated compounds were then recovered by several n-hexane washings. Next, n-hexane extracts were concentrated down to about 2 ml and passed through a Florisil column (4 × 0.7 cm), in the case of the sediment samples, with Cu powder (0.1 g) on the top. Cu powder was previously activated by HCl (18%, Carlo Erba, Italy) and washed with water, acetone and n-hexane. The Florisil column was eluted with 25 mL of n-hexane-dichloromethane (Carlo Erba, Italy, pesticide analysis grade) 85:15 (v/v) mixture and the eluate was concentrated to exactly 0.5 mL. The purified extracts were analysed by gas-chromatography (GC Carlo Erba, Top 8000) coupled with [63]Ni electron capture detector (Carlo Erba ECD 80) using an on-column injection system (volume injected: 1 μl). The column was a WCOT fused silica CP-Sil-8 CB (50 m × 0.25 mm I.D., film thickness 0.25 μm, Varian, USA). The temperature programme used was from 60 to 180°C at 20°C min^{-1}, followed by a run from 180 to 200°C at 1.5°C min^{-1}. A further run from 200 to 270°C at 3°C min^{-1}; an isothermal condition at 270°C was maintained for 20 min, with helium as carrier gas (1 ml min^{-1}) and nitrogen as auxiliary gas (30 ml min^{-1}) was implemented. Sample quantification was performed using external reference standards containing a mixture of HCB (Hexachlorobenzene), HCHs (Hexachlorocyclohexanes), pp'DDT, (pp'Dichlorodiphenyltrichloroethane), pp'DDE (pp'Dichlorodiphenyldichloroethylene) and pp'DDD (pp'Dichlorodiphenyldichloroethane) (Pestanal, Sigma-Aldrich, Germany) in iso-octane (Carlo Erba, Italy, pesticide analysis grade). The detection limit for each chlorinated pesticide was 0.1 ng g^{-1} d.w. Recovery efficiency was tested on reference sediment and it was within 80–100% for the three DDT congeners.

Statistical Analysis

Principal component analysis (PCA) was applied to the correlation matrix of the measured data of sediments at each sampling sites of Lake Natron and rivers (pp'DDT, pp'DDD, pp'DDE and LOI%). In order to treat all variables as if they were of equal importance regardless of their scale of measurement, data were standardised to 0 mean and to 1 standard deviation. PCA calculates a number of linear combinations of the observed variables that explain as much as possible their original variation. Each principal component (PC) results from a linear combination of the original variables multiplied by coefficients (factor scores) whose numerical values are proportional to the contribution of the variable to the component. As a result of an effective ordination process, the first PC accounts for the greatest proportion of the original variance, whilst the second, as well as the following PCs, progressively explain smaller amounts of data variation. PCA was processed using Addinsoft, XLSTAT© ver. 7.5.3.

RESULTS

The conductivities of the inflowing streams to the Natron lagoon show considerable differences according to their source (Fig 4a; Table 1). The lowest conductivity was measured at site R2, a tributary stream fed by underground spring waters, indicated by its slightly lower surface water temperature. Closeby a second stream, R1, also fed by springs further back from the lake, had 20 times higher conductivity, and it was diluted into the lagoon through R4 and R5. Once in the lagoon, however, evaporation at the high temperatures elevated the conductivity to conditions which must be typical of the main lake (it could not be reached because of very flocculent deep mud). At Lake Bogoria where there is little spatial change in concentrations as the lake is deep, conductivity was 68,650 μS cm^{-1}.

The measurements of dissolved oxygen concentrations (DO) reflect the degree of productivity of the lake, recorded during the day so they reflect photosynthesis. The highest DO values at Lake Natron were at sites L2 and L3 and at R4 and R5 where less alkaline water was lying over shallow epipelic (at the surface of mud) cyanobacteria and algae, photosynthesizing highly and providing food for thousands of lesser flamingos (pers. obs.). Low productivity was characteristic of the upstream sites where the oxygen concentration was lower. The particularly high levels of DO at Lake Bogoria reflect the extremely high photosynthesis of planktonic *Arthrospira fusiformis* in this lake, a major lesser flamingo feeding environment.

HCB and HCH isomers in the sediments were always below the detection limit. The maximum value of Total

Table 1 Physical–chemical parameters of the water column, LOI and DDT residues in sediments at different sites of Lake Natron and Lake Bogoria (to identify the location of the stations see Fig. 4a, b)

Water column						Sediments				
Sites	St.	DO (mg l^{-1})	pH	T (°C)	Cond. (µS cm^{-1})	LOI (%)	pp'DDT (ng g^{-1} d.w.)	pp'DDE (ng g^{-1} d.w.)	pp'DDD (ng g^{-1} d.w.)	pp'DDE/ pp'DDT
Lake Natron	L1	0.6	9.93	33.7	106650	20.5	0.6	15.9	1.5	26.5
	L2	17.2	10.01	33.1	14500	15.4	0.6	7.9	1.5	13.2
	L3	19.4	10.32	32.5	12800	13.2	0.4	4.6	0.9	11.5
Natron inlet	R1	2.8	9.50	33.1	39500	15.8	1.6	27.2	2.1	17.0
	R2	3.5	8.43	31.0	1950	10.9	0.7	12.6	0.6	18.0
	R3	4.1	9.18	33.0	21500	6.3	0.9	16.1	1.2	17.9
	R4	12.1	9.21	33.2	12500	8.7	0.8	6.5	0.7	8.1
	R5	13.5	10.20	34.5	13500	11.8	0.8	10.6	1.5	13.2
Lake Bogoria	LB1	>20.0	10.14	31.3	68650	2.7	0.4	10.8	0.7	27.0

Fig. 5 Correlation between DDTs and conductivity (*dotted line* pp'DDE; *dashed line* pp'DDD; *full line* pp'DDT)

y = 0,4739x + 6,1687
R^2 = 0,7126

y = 0,0389x + 0,5287
R^2 = 0,7833

y = 0,0247x + 0,5206
R^2 = 0,8971
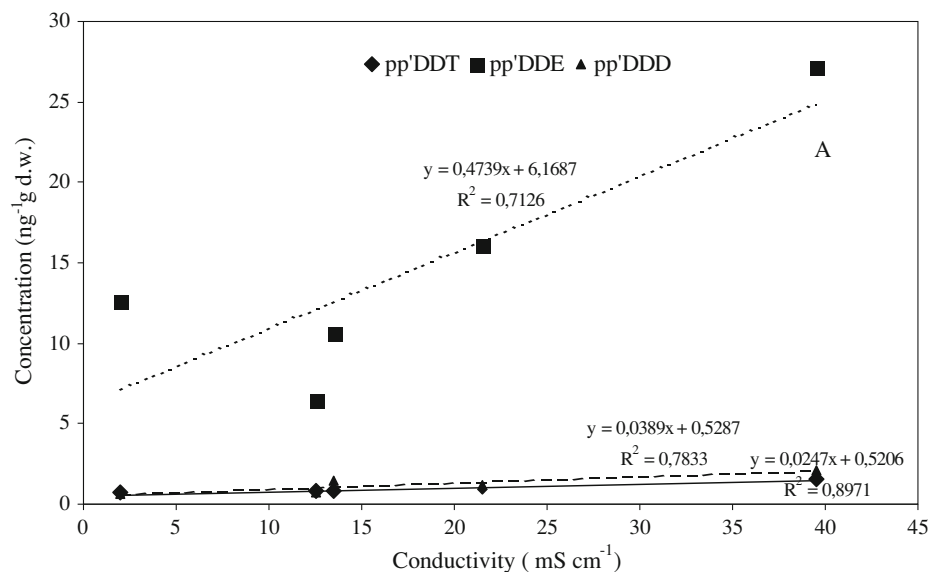
DDT was detected in sediments of site R1 of the river coming from the west (Fig. 4a) which crosses a Maasai village, whilst the river fed by the underground spring (R2) was less contaminated. A significant correlation was found between conductivity and DDTs for river sediments (Fig. 5), which reflects a pollutant dilution effect along the rivers to the lake due to the mixing of spring waters, which should have lower DDT residues, with the river which crosses a Maasai village (R1) and drains a larger basin. The maximum contamination in lagoon sediments was measured farthest site from the river inlet. Comparable levels of DDT residues were found in Lake Bogoria sediments.

Multivariate analysis amongst LOI% content and DDTs for Lake Natron sediments (Lake Bogoria was not considered in this analysis since it was supposed to be affected by DDT pollution sources in a different way, being located in a different watershed) showed that the first two axes of

the PCA accounted for 92% of the data variance (Fig. 6). The first principal component axis (57% of variance) describes a gradient of pp'DDT and pp'DDE from site L3 up to site R1, which presents the highest compounds' concentrations. The second axis (35% of variance) illustrates the increasing percentage of organic matter (LOI) from river samples up to site L1 in the lagoon. pp'DDD does not contribute in the described data variation. It can be observed in the biplot that the Lake Natron samples form a cluster separated by the rivers along a North-East direction, indicating that the organic matter, pp'DDT and pp'DDE differentiate the lagoon samples from those of the rivers. The inputs of organic matter to the rivers and into lake probably have a different origin; in the first case it should come from the humans and their livestock farming, whilst in the second case from the degradation of phytoplankton and faeces of the birds.
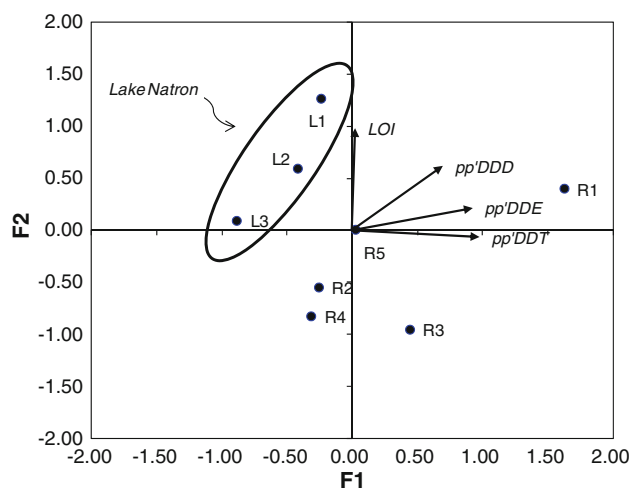
Fig. 6 Bivariate PCA scores plot (F1 vs. F2) for the analytical data. *LOI* organic matter percentage determined by weight loss-of-ignition. The *arrows* show the direction of increasing values of the analysed variables

DISCUSSION

Recent data of DDT contamination of the Rift Valley lakes are quite scanty. Similar levels of DDTs found in the present work were found in the sediments of Lake Nakuru (Table 2; Mavura and Wangila 2004), another Kenyan soda lake in the Rift Valley (Fig. 3). In this case, the parent compound pp'DDT was only found in the rainy season at concentrations higher than its metabolites. In the Tanzanian side of Lake Victoria (Fig. 3), the concentrations of DDTs ranged between the detection limit and 12 ng g^{-1} d.w. (Table 2; Kishimba et al. 2004) during the dry season, whilst higher concentrations were measured during the rainy season. In light of these observations, in the near future the investigation of the contamination by DDT even during the rainy season for lakes Bogoria and Natron seems to be important.

The peak concentrations found in Lake Victoria sediments were ascribed to an old pesticide stockpile at Vikuge (Elfvendahl et al. 2004; Kishimba et al. 2004; Marco and

Kishimba 2005, 2007), which could also be a point source pollution for many kilometres around, even for lakes Natron and Bogoria at a distance of around 500 km. The hypothesis of the aerial transport from such a 'hot spot' pollution site seems to be confirmed by the analysis of the sample of *Acacia tortilis* leaves. Although *Acacia* sp. is not usually used as a micropollutant bioindicator (but it was the only tree species occurring in the area), and as a consequence cannot be used for quantitative comparisons, it can provide useful information for a qualitative evaluation. Only pp'DDT and pp'DDE were detected at comparable concentrations (about 0.5 ng g^{-1} d.w.), indicating that pp'DDD found in river and lake sediments could be formed in soil and/or sediments in anaerobic conditions (Zoro et al. 1974). Moreover, the pp'DDE/pp'DDT ratio equal to 1 in the leaves might indicate a recent local input of the parent compound through the atmospheric transport. The ratio pp'DDE/pp'DDT was particularly high for the lake and river sediments, indicating that most of the parent compound pp'DDT should have a low persistence in this tropical environment, as already demonstrated by in situ studies (Samuel and Pillai 1989). The occurrence of pp'DDT as dominant pollutant in Lake Nakuru sediments (Mavura and Wangila 2004) during the rainy season and its disappearance during the dry season, leads support to this hypothesis.

CONCLUSIONS

DDT pollution was found to be quite variable in the littoral sediments of the Rift Valley lakes. As the capacity of binding the pollutants in sediments seems to depend mainly on their organic matter content, the biotic components may play a role in the patchiness of contamination.

At present, low contamination levels were measured in the sediments of lakes Natron and Bogoria which should exclude any risk for DDT bioaccumulation in top predators and explain why lesser flamingo population is still abundant and healthy in these aquatic environments.

Table 2 DDT residues in sediments of Rift Valley lakes (Fig. 3) in ng g^{-1} d.w.

Sites	Season	pp'DDT (ng g^{-1} d.w.)	pp'DDD (ng g^{-1} d.w.)	pp'DDE (ng g^{-1} d.w.)	ΣDDT (ng g^{-1} d.w.)	References
Lake Nakuru	Dry	bdl	2.3	7.5	9.8	Mavura and Wangila (2004)
	Rainy	13.6	bdl	2.2	15.8	
Lake Victoria (Southern basin)	Dry				bdl-12	Kishimba et al. (2004)
	Rainy				bdl-131	
Lake Natron	Dry	0.4–0.6	0.9–1.5	4.6–15.9	5.9–18.0	Present research
Lake Bogoria	Dry	0.4	0.7	10.8	12.0	Present research

bdl Below detection limit

However, further investigation should be undertaken to verify if pollution increases during the rainy season as occur in many other lakes located in the same region. Moreover, as the lesser flamingo is a filter feeder species with a sieve-feeding mechanism to harvest phytoplankton, it ingests around 72 g d.w. day^{-1} of cyanobacteria and algae (Vareschi 1978), with consumption rates sometimes exceeding the primary production rates (Vareschi and Jacobs 1985), so its load of pesticide residues should be checked by other methods, such as eggs analysis. Finally, a new sampling campaign should be carefully organised, bypassing the difficulties linked to the logistic matters of these remote areas, addressing particular attention to the contamination of the different components of the food chain, such as the one of the phytoplankton communities.

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