PERSISTENT ORGANIC POLLUTANTS IN FREE-RANGE CHICKEN EGGS FROM ARUSHA, TANZANIA: LEVELS AND HUMAN HEALTH IMPLICATIONS

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Introduction

During the last century Tanzania has experienced extensive population growth, urbanization accompanied by industrial and agricultural progress. These activities have led to increased use and discharge of industrial- and agrochemicals¹. In addition, Tanzania holds tons of old obsolete pesticide stockpiles². A large number of these chemicals, with well-documented toxic potency, are persistent organic pollutants (POPs). With the rapid growing population, access to protein rich food sources is essential. Eggs constitute an important protein source in Tanzania, in addition to meat and fish. In addition, eggs are an important part of small farmer's income. The main source of POPs to the hens and further to the eggs is the feed. On a global scale, hen's eggs are frequently used in monitoring studies to assess human exposure to POPs. These investigations are primarily meant to monitor levels of dioxins and dioxin-like PCBs in eggs from commercial and free ranging chicken farms³. Less literature is found on other POPs in hen's eggs⁴. In the present study the main goal was to assess the occurrence and levels of POPs in relation to other studies and to Maximum Residue Levels (MRLs).

Table 1: Results from the questionnaire conducted during sampling of free-range chicken eggs in Arusha

	Number of households/ pools	Total number of eggs	Gardening	Livestock	Housing of chicken yes/no	Feeding chicken yes/rarely/no	Pesticide use yes/rarely/no
Kwamrefu	8	42	7	2	1/7	3/5/0	5/2/1
Kilanyi	9	54	5	1	3/6	5/3/0	0/0/9
Siwandeti	4	24	4	2	1/3	1/0/3	4/0/0
Olmotonyi	6	33	6	6	4/2	4/0/2	4/0/2
USA River	1	6	1	1	1/0	1/0/0	1/0/0

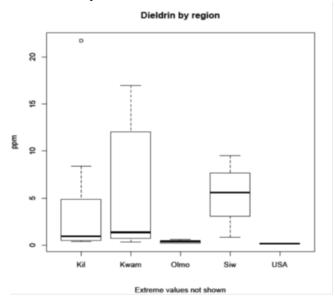
Materials and methods

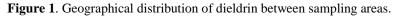
In September 2012, 159 eggs were collected from 28 different households in four townships of Arusha (Kwamferu, Kilanyi, Siwandeti Olmotonyi and USA River) (Table 1). The eggs from each household were pooled, with preferably 6 eggs per pool. 28 pooled samples were thus available for the analyses. Information on housing and feeding of the chicken and use of pesticides was given by the participants (Table 1). The samples were pooled in in Arusha and kept frozen at -20° C until transport and analyses in Norway. Permissions for export and import were granted by responsible authorities. The chemical analyses were performed at the accredited Laboratory of Environmental Toxicology, Norwegian University of Life Sciences (NS-EN ISO/IEC 17025, TEST 137). The method included liquid/liquid fat extraction and a gravimetrical determination of the lipids. Prior to GC analyses lipids were removed by either use of $\geq 97.5\%$ H2SO4 or using a gel permeation column. Determination of Organochlorine compounds (OCs): HCB, HCHs, chlordanes, DDTs, mirex, toxaphenes, aldrin, endrin, dieldrin, endosulphanes, PCBs and brominated flame retardants (BFRs) including PBDEs and HBCDD were performed with HRGC-ECD, and HRGC-MS. Details of the methods were described earlier⁵.

Results

Chlorinated pesticides: HCB, p,p'-DDE and dieldrin were detected in 100% of the samples. Heptachlorine, cisheptachlorine epoxyd, trans-heptachlorine epoxyd, technazen, and quintozen pentachloranillin were <LOD in all the pooled samples, and will not be discussed further. HCB levels were generally low and in the in the same range at all five areas. However, one sample from Kwamrefu had significantly higher levels of HCB (167 ng/g lw). Σ HCHs levels were generally low, with a median of 0.81 ng/g lw (Table 2), except for one sample from Olmotontyi, which measured Σ HCHs of 66.0 ng/g lw. γ -HCH was the most abundant compound of the HCHs and contributed with 76% to Σ HCHs. Oxychlordane was detected in 32% of the samples, and trans-nonachlor in Organohalogen Compounds Vol. 76, 574-576 (2014) 574

50%. Σ Chlordanes consisted mainly of trans-nonachlor. p,p'-DDE was detected in 100% of the samples, p,p'-DDD in 11% and p,p'-DDT in 96%. Generally, p,p'-DDE, was the dominating compound contributing 79% to Σ DDTs. The DDT/DDE ratio ranged between 0.003-6.64. Dieldrin was detected in 100% of the samples, with a median of 3,606 ng/g lw. In one sample site dieldrin, aldrin and endrin were detected in levels of 98,790 ng/g lw, 6 ng/g lw, and 149 ng/g lw, respectively. The levels of dieldrin at this site were significantly higher than measured at any other sample site (Figure 1). Endosulfan was detected in 50% of the samples. Σ Endosulfan was dominated by endosulfan sulfate, present in 46% of the samples, compared to endosulfan α and endosulfan β detected in respectively 17 % and 10% of the samples. Toxaphenes were detected in 35% of the samples. CHB-26 had the highest maxima of all the toxaphenes.





PCBs: PCBs were detected in low concentrations and the median of the 7 marker PCBs was 1.75 ng/g lw. PCB-153 was the only PCB congener detected in all the samples, and had also the highest maxima of 3.12 ng/g lw. The most abundant PCBs, PCB-153 and PCB-180 contributed 60% to Σ PCBs, of which PCB-153 accounted 34%.

Brominated flame retardants: PBDEs were detected at all the sample sites. BDE-209 was present in 100% of the samples and contributed 80% to Σ PBDEs. HBCDD was detected in 60% of the samples, with levels ranging between <LOD-62.5 ng/g lw.

Table 2. Concentrations (ng/g lipid weight) of some persistent organic pollutants (POPs), in pooled samples of free ranging chicken eggs from Arusha, North Tanzania

Compound	Median	Mean	Minimum	Maximum
<i>N</i> = 28				
НСВ	1.9	8.4	0.6	167
ΣHCHs	0.8	3.2	0.03	66
ΣChlordaner	0.1	0.7	<lod< td=""><td>14</td></lod<>	14
ΣDDTs	6	37	2	324
Dieldrin	7.6	3606	1.8	98790
ΣEndosulfan	1	3.2	<lod< td=""><td>11</td></lod<>	11
ΣCHBs	1.4	4.5	<lod< td=""><td>28</td></lod<>	28
ΣPBDEs	14	13	0.2	347
HBCDD	4.3	13	<lod< td=""><td>63</td></lod<>	63

Discussion

The main findings in this study were the very high levels of dieldrin found in several of the egg samples and the 100% frequency of detection (Table 2). Aldrin and endrin were also detected in one sampling site. Aldrin and dieldrin have been banned since the 1980s in most countries and was included among the first listed chemicals in the Stockhom Convention⁶ because of its high toxicity⁷. In Arusha, three stockpiles were reported to contain Organohalogen Compounds Vol. 76, 574-576 (2014) 575

dieldrin and aldrin in 1998⁸. In 2004 these stockpiles were gone but substantial amounts of aldrin were still registered at a seed producing farm. The maximum level of dieldrin found in this study (98,790 ng/g lw), was approximately ten times higher than the maxima of 1,298 ng/g lw measured in Belgium⁴. Three of the samples exceeded EU MRL for dieldrin in bird egg of 0.02 mg/kg wet weight, whereas the highest level exceeded with more than 500 times. p,p'-DDE and p,p'-DDT were detected in 100% and 96% of the samples, respectively. The high DDT/DDE ratio suggests recent use of DDT in Arusha. Compared to other countries Σ DDT levels in the studied eggs were in the median range and did not exceed EU MRLs. The median HCB levels were low and considered as regional background levels. The finding of one sample with high HCB levels, indicated, however, that seed treated with HCB may be distributed among farmers. HCHs were also in general found in low levels. Nevertheless, markedly higher levels of HCHs were measured from a single sample site and random release of remaining, unused HCH to the environment cannot be excluded.

PCBs: PCB levels were low and were considered as general background levels for the region.

BFRs: PBDEs were detected at all the sample sites, and the levels in this study are higher than detected in chicken eggs from other parts of the world. BDE-209 was the dominating congener in the eggs, which is consistent with the findings in in home-produced eggs from China⁹ and Belgium¹⁰. A recent study from Tanzania reports that BDE-209 was detected in 68% of tilapia (*Oreochromis* sp.) filets from Lake Victoria⁵. In addition to this, results in the present study strongly indicate a release of PBDEs in Arusha and Northern Tanzania from unknown sources. Also the occurrence and variable levels of HBCDD in the studied eggs warrants further research. Possible sources of BFRs may be dumping and disposal sites of obsolescent electric materials and uncontrolled outdoors burning of these in Arusha.

Conclusion

The present study found dieldrin and DDT as the dominating pesticides in free-range chicken eggs from Arusha. These findings are in accordance with earlier environmental observations from Tanzania. The detected levels were generally low, and suggest old residues for most of the compounds. Nevertheless, high levels of dieldrin, the presence of aldrin, in addition to a high DDT/DDE ratio, suggest recent use of these banned substances. The detections of the industrial chemicals PCBs, PBDEs and HBCDD confirm the ubiquitous presence of these persistent contaminants in Tanzania with a short history of industrial activity. Further studies are needed.

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